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Preface

The triennial INTERPOL Forensic Sciences Managers Symposium serves as an unparalleled opportunity for forensic science managers from across the Globe to spend a few precious days together, sharing and exchanging experiences and knowledge and discussing the challenges they face in an increasingly complicated world. The 18th IFSMS meeting represents a continuous engagement and support by INTERPOL in the forensic science domain of over 50 years.

The purpose of the symposium is to create a forum that facilitates:

- the presentation of advances made in scientific methods over the previous three (3) years and to provide a perspective of future developments in forensic science;
- the exchange of information which will enhance scientific methods in criminal investigation and the administration of justice at a national and international level;
- the discussion of challenges encountered by member states and the possible provision of solutions; and
- the exchange and pooling of ideas for future progress.

We live in a rapidly developing world where the delivery of justice is complex and challenging. The need for both a validity of process and procedure, and a confidence in the competence and performance of forensic science staff has led to a global and welcome desire to move forensic science onto an accredited footing in alignment with many other industries worldwide.

In recent years, the analysis and interpretation of many types of scientific evidence have come under scrutiny and the robustness of the scientific underpinning of some evidence types is increasingly being questioned. The needs of the judiciary to have an understanding of the scientific validity of the evidence presented is acute and critically important if they are to make confident decisions on admissibility of that evidence in the service of justice. This provides both challenges and opportunities to assess and address the scientific validity of the current means of analysis of different evidence types, to develop ground truth databases and to determine appropriate means of expressing evaluative interpretations of forensic evidence within differing case contexts. These proceedings provide one gateway to disseminating these important research activities.

The 18th IFSMS has only been possible with the support of INTERPOL and the General Secretary. INTERPOL staff coordinated all aspects of the organisation's involvement including distributing the meeting announcements; organizing registration; arranging the meeting venue, and publishing the meeting proceedings. In particular, the Organizing Committee is extremely grateful for the continuing efforts and support of Serge Eko, Kim Legg and Eileen O'Reilly. IFSMS would not be possible without the significant work of the Organizing Committee, each Coordinating Laboratory and the review paper authors and I am inordinately grateful for their support and efforts throughout the past three years.

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Criminalistics

Firearms, 2013-2015

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Introduction

This review paper covers the advances in scientific methods and general discussions applied to firearms examination, published from 2013 until and including 2015. A literature search was conducted covering articles on this subject published in the main forensic journals:

- AFTE Journal
- American Journal of Forensic Medicine and Pathology
- Forensic Science International
- International Journal of Legal Medicine
- Journal of Forensic Identification
- Journal of Forensic Sciences
- Science and Justice
- The Australian Journal of Forensic Sciences

1. Firearms identification

Following the recommendations made in the National Academy of Science's 2009 report to strengthen the scientific foundations of firearms identification [1] several articles have been published.

1.1. Current topics

In respect to forensic examinations, including firearms examination, two main topics have received specific attention over the last 3 years:

1. Implementing the likelihood ratio approach in forensic casework
2. Implementing context information management in forensic casework.

1.1.1. Implementing the likelihood ratio approach

Two articles have provided a practical introduction to the use of the likelihood ratio (LR) approach in forensic firearms examination. Bunch and Wevers (2013) discuss that a provided categorical conclusion provides a form of a posterior odds, which is not ideal for reports and testimony and is mathematically incorrect as these posterior odds can never equal infinity (probability of 1). They propose three possible solutions to the overall current state of affairs [2]:

1. Continue with the current paradigm, but provide more transparency in reports and testimonies by refraining from concluding in absolute certainties and use phrases such as "practical certainty" and by disclosing the assumption that contextual information is embedded in the prior odds, which are a part of the final conclusion. They view this solution as a less desirable solution.

2. Examiners could provide a report detailing a conclusion based on the complete Bayes' rule, providing posterior odds where the examiner both calculates and assigns the LR and the prior odds. They say that this is neither a desirable solution as examiners are generally not regarded to be the appropriate persons to assign the prior odds.
3. Examiners should only report the LR and could provide some guidance to the trier-of-fact how the posterior odds will be influenced based on the prior odds. This could be done by providing e.g. a sensitivity table.

Bunch and Wevers (2013) provide some detailed examples of how the LR can be assigned in forensic firearms examination. While doing this, they make the distinction between the LR based on the class-characteristics evidence and based on the microscopic comparison evidence, which they argue to be independent from each other. The final LR of a comparison between e.g. a bullet and a firearm would consist of the LR based on the calibre and class-characteristics of the bullet and firearm and their frequency of occurrence in e.g. firearms casework and of the LR based on the comparison of striations and impressions which can be multiplied with each other. This explanation of the way the final LR should be assigned corresponds to what is also argued by Hicks, Biederman, de Koeijer, Taroni, Champod and Evett (2015). They have provided a few examples how the LR can be assigned for different types of forensic examinations [3].

Kerkhoff, Stoel, Mattijssen and Hermsen (2013) have also written a plea, with a step to step discussion, to report the results of firearms examinations as an LR. They first argue that the examiner is supposed to inform the trier-of-fact about the strength of the evidence (which is fundamentally probabilistic in nature) in a scientifically sound way after which the trier-of-fact is responsible to assess the weight of the provided evidence for the particular case. By going through the different possibilities of reporting firearms evidence, from a categorical conclusion, a probabilistic conclusion, an expert opinion and an LR, they provide an accessible discussion on the merits of reporting an LR. They conclude by stating that although reporting an LR is the best approach, it seems harder to explain and understand than most logically flawed formats. Although this is not optimal, the problems which are solved by reporting an LR outweigh this difficulty [4].

Agreeing with each other and with the authors of the former articles [2,4] that reporting an LR is the best method to evaluate forensic evidence Taroni, Bozza, Biedermann and Aitken (in press) and Sjerps, Alberink, Bolck, Stoel, Vergeer and Zanten (in press) disagree with how this should be reported. The former argue that an LR should be reported as a single value based on a "full-Bayesian" approach without additionally considering the uncertainty of the LR [5], while the latter argue that not only an LR value should be reported, but that its uncertainty should also be addressed to inform the trier-of-fact about the essential information needed to assess the reliability of the evidence [6].

1.1.2. Implementing context information management

Part of the criticism towards the traditional forensic sciences, including firearms examination, focuses on the biasing effect of task-irrelevant information on the judgements of examiners in forensic casework [1,7-10]. It has been argued that forensic examiners should acknowledge the risk of contextual and other biases, and minimise their effects by implementing appropriate procedure [9,11-14]. With the growing acceptance that these issues exist and are relevant for forensic casework several procedures have been proposed for various forensic disciplines, including (linear) sequential unmasking [11,15,16], evidence line-ups [14], and the 'case managers' model [9,13]. For firearm examination, an implemented context information management procedure has been published [17]. In this article the design and implementation of the procedure is described guided by a taxonomy of

different sources of context information [18]. After showing that removing all context information, except for the information that is necessary for the examiner to do his/her work, seems to work best, they conclude with a flow-chart of their implemented procedure. Based on the same procedure some of the authors have also written an article which focuses more on the potential influence of bias through a step-by-step discussion of a case. Through this article they show under which circumstances bias might influence the examination and how context information management can help to minimise this possibility [19].

1.2. Validation studies and statistical foundations

Traditionally, validation studies within the field of forensic firearms examination have been based on:

1. Reproducibility of marks
2. Individuality of marks

1.2.1. Reproducibility of marks

Reproducibility has been studied by shooting large amounts of ammunition through one firearm or barrel and by then comparing the marks (seen in the ammunition parts fired over time) to check whether they change over time or are still sufficient to reach a conclusion pointing in the direction of a similar source.

As a follow-up on their previous study [20], Mikko and Miller (2013) fired thirty additional bullets with the same firearm and thirty bullets using a second M240 machine gun with a different barrel. Four examiners were asked to compare the marks in these newly fired bullets with those in some of the bullets fired throughout the earlier reproducibility study consisting of 20,000 shots. In total 164 questioned high velocity bullets were examined, resulting in 164 correct answer and an error rate of zero [21].

Wong (2013) studied the reproducibility of marking in a 1,000 bullets' land engraved areas and cartridge cases' firing pin aperture shear marks from a Ruger P89 pistol, by using both pattern matching and QCMS. They compared every 25th fired bullet and cartridge case to the first fired bullet and cartridge case and to the previously and subsequently collected 25th bullet and cartridge case. For the bullets some differences in the striations were seen, especially in the trailing edge of the land engraved areas. Striations became less well defined over time but each collected bullet could be identified to the first fired bullet. Bullets fired closer in sequence to each other resulted in higher CMS runs. The degree of shearing and the overall quality of the marks were inconsistent for the firing pin aperture shear marks. Even though this was the case nearly all cartridge cases could be identified to the first fired cartridge case [22].

A study based on three 9mm Luger Walther P99 pistols showed that after at least a 1,000 shots the breechface recess, firing pin impression and firing pin aperture shear marks showed reproducibility and that the marks held sufficient individual characteristics for identification [23].

1.2.2. Individuality of marks

Individuality has been studied by comparing the marks in cartridges cases and bullets fired by consecutively manufacture firearm components and by looking into the manufacturing process.

A study [24] consisting of five consecutively manufactured slides from a 9mm Luger Hi-Point model C-9 pistol showed that it was possible to distinguish between these slides based on the breechface impressions. 68 trained examiners received 2 reference fires from each of the slides and 8 questioned cartridge cases. All conclusions were correct and no

inconclusives have been called by the examiners. The authors add that differences in the locations of the firing pin aperture were seen between the shots fired from different slides. This might have unintentionally helped the examiners during the comparison phase. A study [25] utilising 10 consecutively manufactured slides from a 9mm Luger Ruger resulted in an error rate of less than 0.1%. 217 firearms examiners with at least 2 years of training received 2 reference fires from each of the slides and 15 questioned cartridge cases. From the 3255 breechface comparisons, 3239 were correct, 2 incorrect and 14 inconclusive, which resulted in an error rate of 0.0006363 (SE = 0.006617). In the same article reproducibility of the breechface marks is also tested. Additional test fires were compared by 114 participants, which resulted in 570 breechface comparisons, from which 564 were correct, 1 was incorrect and 5 were inconclusive. Overall the results showed that (with a significance level of 5%) firearms examiners were able to correctly identify fired cartridge cases, to correctly identify fired cartridge cases fired at different intervals from each other, there was no significant difference between examiners with less or more than 10 years of experience, and that there was no significant difference between lighting methods, utilised microscopes, or used methods in regard of correct/incorrect conclusions.

When comparing the marks in the firing pin, ejector and breechface marks of 10 consecutively manufactured pistols of two different makes of .32 Auto Turkish pistols it was deemed possible to differentiate between the firearms [26].

When examining and comparing the extractor marks of two sets of six Remington 870 shotguns with either metal injection molded (commercial) or milled (law enforcement) extractors some similarities in marks within each group were seen, but the extractor marks could still be identified back to their corresponding extractor [27].

Based on information from a study, Metal Injection Molding has the potential to introduced subclass characteristics in the resulting components. Comparisons of the marks of five extractors from a calibre .40 S&W M&P pistols illustrated that there was some presence of subclass. The electropolish finish or the melonite process after production of components (used for the firing pins) seemed to diminish the possibility of subclass [28].

The Metal Injection Molded breechface of a .357 Magnum Taurus model Protector Poly was examined. Based on the manufacturing process information that the breechface receives additional machining and is glass microspheres blasted the marks were interpreted as being individual in nature [29].

Vibratory finishing of broaching marks showed that more of the finer striations were removed as finishing time increased. As a result the grosser marks became more pronounced, which could happen for both gross individual and gross subclass marks [30].

1.3. Parameters that affect the identification process

Studying the marks left by modified or re-activated firearms in firearm crime, the authors show that even though some parameters seem suboptimal for identification purposes (e.g. large bore, no rifling, choke that distorts the bullet marks) there are still marks that can be used for identification of the fired bullets and cartridge cases [31].

While discussing the design and components of the Winchester PDX1 410 Defender shotshells the authors also show that marks useful for identification purposes can be found on these fired cartridge cases [32].

Explaining how frangible bullets are constructed and performing test fires, the authors state that the striations resulting from shooting through a barrel changed for one of the used

firearms. While the frangible bullets usually break-up upon impact there were still some intact and potentially identifiable bullets after being fired through the target materials (plywood, drywall, sheet metal) [33].

Firing .45 GAP calibre cartridges with several .45 ACP pistols showed that there was a high tendency for the primers to be pierced by the firing pin. Although this happened there were still sufficient marks available on the fired cartridge cases to be identified to the used firearm [34].

Rusted firearms are sometimes submitted for examination. Different methods of rust removal and restoration of firearms are examined. The marks on fired bullets and cartridge cases before rusting and after de-rusting were compared. The tested soda blasting and Rust Release™ seems to provide the best results [35].

A reprint of “Forensic Ballistics” by Goddard, first published in 1925, was made available in the AFTE journal [36].

When the spring controlling the movement of the firing pin is absent, damaged or otherwise lost its control over the firing pin in top break (hinge frame) firearm, the firing pin can move freely. During opening and closing of the firearm the firing pin can now be damaged and thus altered, which could deceive the examiner [37].

1.4. Identification based on unusual marking

Dutton (2014) discusses a case where a .22 calibre bullet proved to be fatal. While showing marks from the barrel there were also marks resulting from other sources. These marks resulted from separate processes during loading by the use of an aftermarket magazine in conjunction with the exhibit sawn-off rifle. The main focus lies on the interpretation of these extraordinary marks [38].

Breechface recess marks of 20 Beretta model Px4 Storm, calibre 9mm Luger were compared. The marks are visible on the rim of the cartridge cases but demonstrate poor to excellent reproducibility. When they do reproduce they can be used for identification purposes [39].

Reproducible cycling marks were seen on the nose of cycled cartridges or fired bullets resulting from contact with the feed ramp and frame of the .380 Auto, Ruger model LCP pistol [40].

An identification between a fired cartridge case and a highly corroded firearm recovered from a chemical toilet was still possible using the chamber marks. This was probably possible because the chamber had been preserved by the live cartridge that was present in the chamber [41].

A 9mm Luger Hi-Point model C9 pistol was submitted for comparison to a questioned bullet. During examination the breechface showed features not seen in other Hi-Point C9 pistols. It is hypothesised that the relative softness of the alloy allowed the heads of the cartridges to displace material during firing and thus led to the obliteration of the factory-made breechface marks over time [42].

When comparing test fired cartridge cases and crime scene cartridge cases unusual marks were observed. Examination of the firearm showed that these resulted from tampering with the bolt face, extractor and ejector of the firearm, a possible attempt to avoid firearm identification [43].

A rifled slug was identified to the smooth bore of a Baford Arms model Thunder, calibre .410 Derringer [44].

1.5. Class characteristics

Adding to the earlier publications [45-48] on the difference in class characteristics between Glock and Smith & Wesson Sigma pistols the authors introduce an additional mark that may help to differentiate. A mark on the rim at 3 o'clock can result from the trigger bar tip of Glock and S&W Sigma pistols. The marks on the rim can be found in cartridge cases fired from both brands, but an additional scratch on the wall of the cartridge cases was only observed on the cartridge cases fired from Glock pistols [49].

The class characteristics of the Czechoslovakian CZ model 83, calibre .32 Auto pistols are discussed. The firing pin impression is round, with a round base including circular detail and located off-centre, approximately at the 4-5 o'clock position. The breech face impression shows vertical lines and the ejector mark is triangular with a rounded edge located at 7-8 o'clock. The extractor mark is located at 2-3 o'clock in the extractor groove [50].

Based on two types of homemade submachine guns, resembling Carl Gustav and UZI submachine guns, it was shown that they might be distinguished using the ejector cut-out shapes [51].

1.6. Subclass characteristics

Two articles showing subclass characteristics have been published. One of these provides an example of subclass visible in the 12 land engraved areas of the pellet and the respective lands in the barrel of a Gamo .22 air rifle. Similar striations are seen in each of the lands [52].

A second article shows subclass characteristics to be present on the breechface marks of Jiminez Arms JA Nine pistols. The article also provides details about the manufacturing process of these firearms [53].

1.7. Proficiency testing

The ENFSI Expert Working group Firearms/GSR sent out the 2009 edition of their proficiency test. The test consisted of ten sets of castings, from which five contained bullets and five contained cartridge cases. Each set consisted of one questioned item and two known items from the same firearm. Sixty-four laboratories (mostly European) returned the answer form, giving a total of 637 conclusions (the three missing conclusions were from examiners who submitted the specific test sets). In total twenty-six conclusions (4%) were false identifications and thirteen conclusion (2%) were false exclusions [54].

With the help of external parties an exploratory double blind testing programme was designed. The external parties mixed fake cases disguised as regular cases in the main case flow. From the total of 29 conclusions no misleading evidence was reported (all conclusions supported the true hypothesis). The article discusses the design considerations of the programme, the details of the tests and described various ways to analyse the test results [55].

Stroman (2014) discusses the criticism from scientific and legal experts over the past years regarding the lack of empirical research and blind testing. A literature search revealed two studies that involved declared double-blind testing. Setting up a study with realistic samples using a declared double-blind testing format the author tried to determine the error rate in firearms identification. Apart from this, the evidential strengths of the different components of the Smith & Wesson model 4006TSW, calibre .40 S&W are discussed [56].

1.8. Instrumental methods

Forensic firearms examinations are traditionally based on examiner based judgments. Although these examiners are highly trained, the call for more objective methods is often heard. Different approaches to objectify the firearm comparisons through the use of 2D and 3D instruments have been studied in the past years.

An imaging technique that is capable of reducing glare, reflection and shadows is said to greatly assist the process of toolmarks comparison. This situation could be reached using the expensive 'far-IR'. Visual comparison resulting from the cheaper 'near-IR' are compared with visual light images. The use of near-IR photography did not reveal more details and could not effectively eliminate reflections and glare, thus showing little advantage when compared to regular visible light photography [57].

Three types of microscopy were addressed for the comparison of the markings in a 9mm Luger bullet fired from the polygonal rifled barrel of a Glock 17 pistol: optical microscopy, comparison scanning electron microscopy (CSEM) and virtual (confocal) microscopy. Optical microscopy resulted in an 'inconclusive opinion', CSEM in a positive identification and virtual microscopy in the conclusion that 'regions of interest and commonality' were found. The authors conclude that optical microscopy is the most efficient, CSEM the most advanced but hardly ever necessary and that virtual microscopy is promising but needs more development [58].

Scanning electron microscopy (SEM) is also addressed as a possible supplement for the traditional optical microscopy. The high magnification, the large depth of field and the independence from oblique lighting issues might make it possible to perform better comparisons. A downside is the risk of contamination if the SEM is also used for gunshot residue examination [59].

Utilising the IBIS® BULLETRAX-3D™ the authors examined the usefulness of this technique to determine the order of placement of different toolmarks. The 3D technique provides more detail to address the order of creation and to examine the directionality of engraved marks [60].

Several automated techniques for the comparison of marks have been proposed where some focus on finding matches between spent ammunition and some also focus on the resulting evidential strength associated with these matches.

As a proposed objective method to support the subjective conclusion of an examiner the striations present in the matched land engraved areas from bullets were translated to a barcode. The barcode was based on the distance of the striations from one shoulder of the land engraved area. Through Principle Component Analysis and Support Vector Machine error rates varied between 19.444% and 1.149%. The second result generated by the majority of the analysed bullets indicates the correct grouping based on barcodes was possible, supporting the examiner's subjective identifications [61].

Using 3D-topography images the striations in the bullets' land engraved areas are represented by a feature profile which is used for determining the consecutive matching striations (CMS) automatically. The method might 1) be used for database searches, 2) it will increase the objectivity, as the subjectivity in defining matching striations is decreased, and 3) it provides a means to create an abundance of data for statistical analysis of the CMS method [62].

An automated technique to compare impressions in cartridge cases based on 3D technology has been developed. By coupling the system to a bivariate evaluative model, the assignment of likelihood ratios was allowed. Based on a dataset of 79 pistols (Sig Sauer 9mm Luger) the system showed high discriminative power (LRs exceeding a billion were predominantly obtained for same source comparison) as well as relatively low rates ($\leq 1\%$) of misleading evidence depending on the considered firearm [63].

Using a database of 3D striation patterns, generated by standard tip screwdrivers and 9mm Luger, Glock firing pin apertures, the error rate using algorithmic methods was studied. The marks were recorded by white light confocal microscopy and multivariate algorithmic methods, such as principle component analysis and support vector machine methodology, were exploited to associate striation patterns with their respective sources. When sufficient data is used to train the algorithm, identification error rates of $<1\%$ were seen [64].

The striations present in fired bullets have been acquired using confocal microscopy to create known match and known non match databases of maximum cross correlation function (CCF_{max}) scores. The authors suggest that when the size of the database is increased (both in numbers and in e.g. used calibres) a cut-off value or threshold could be set to conclude whether bullets came from either the same or different sources [65].

A novel 3D imaging and analysis system for cartridge cases is introduced, TopMatch. This system used the GelSight photometric stereo sensor to measure micron scale surface geometry and a novel feature-based matching algorithm. The algorithm separately takes into account the breechface impression and the firing pin aperture shear mark and then combines their similarity into one confidence score. Based on an experiment using 290 firearms and 700 cartridge cases from 24 firearm manufacturers the system demonstrates different performances based on the quality of the available marks. Tests on clean, well-marked datasets score with a 100% accuracy and no false positives, while for less marked cartridge cases the system correctly identifies approximately 75% of the known matches with no false positives [66].

Applying surface metrology to firearm identification is discussed. Objectivity of firearms identification is improved through measurements of surface topography and the application of unambiguous surface similarity metrics, such as the maximum value of the areal cross correlation ($ACCF_{max}$). Case studies on consecutively manufactured tools (barrels and breechfaces) were performed and demonstrated that all tools could be distinguished from each other using this technique. It is explained that the method is as of now not capable of automatically distinguishing between individual marks and (sub)class marks, making proper pre-processing very important. Through the case studies it is shown that there is a good separation between the scores resulting from known matches and known non matches [67].

The National Institute of Standards and Technology (NIST) proposed the NIST Ballistics Identification System (NBIS) to facilitate accurate ballistics identifications and fast evidence searches. The system will use 3D topography measurements. The marks to be compared will be subdivided in correlation cells and the 'valid' and 'invalid' correlation areas will be identified. Based on the concept of correlation cells a 'contiguous matching cells (CMC)' method using three identification parameters of the paired correlation cells (cross correlation function maximum CCF_{max} , spatial registration position in x-y and registration angle θ) is proposed. The number of matching cells could be used to differentiate between same source and different source comparisons, where an identification threshold of 6 cells is mentioned [68].

Developing NBIS, NIST has performed a study on the breechfaces of 40 cartridge cases fired with 10 consecutively manufactured slides. These initial tests show a significant separation between the known match and known non match distributions of 'congruent matching cells (CMC)' cell counts, without any false positive or false negative identifications, verifying their proposed numerical identification criterion [69].

NIST also proposes an error rate procedure based on the CMC method to establish a statistical foundation to support ballistics identifications, and to provide an error rate report for court proceedings. This error rate report could be similar to reporting the random match probability for DNA evidence. Based on the study using 40 cartridge cases fired with 10 consecutively manufactured slides all the error rate values are within the 'extremely strong level' of likelihood ratios (10^{-6} or less) [70].

1.9. Ballistic imaging database

It is suggested that using spatial, temporal and evidence-status data will improve the ballistic imaging performance of NIBIN. Taking into account both locations of shooting incidents and time between shooting incidents increased matching accuracy [71].

While discussing the performance of BALISTIKA 2010, the authors also discuss that computerised ballistic identification systems should implement algorithms to take into account overall differences in class characteristics between manufacturers of firearms to increase accuracy and decrease search duration [72].

The possibility of a reference ballistic image database, containing images of cartridge cases fired by all firearms in legal circulation, is studied using Evofinder®. The results indicate that, although clear improvements were seen in comparison to a similar earlier study [73], the implementation of such a database should not be considered nowadays because at least 43% of the items in the database need to be compared on screen to find at least 90% of all potentials matches. This is still necessary because of the variable character of marks and of differences between types of used ammunition [74].

Using the LUCIA-BalScan™, the use of the Marks Step Integration method is tested for automatic ballistic imaging searches. The method combines the marks in several (partial or damaged) bullets and creates one Virtual Bullet to increase the performance of ballistic database searches. The method is also applicable to cartridge cases, toolmark examination and shoeprint analysis [75].

By implementing a new protocol at the Denver Police Department Crime Laboratory, the firearm evidence is located and entered much faster into NIBIN, increasing its use towards a forensic intelligence tool [76].

2. Firearms and ammunition miscellaneous reports

2.1. Firearms (and ammunition)

2.1.1. Sound of shots

Tests were performed to investigate a shooting incident and the question how quickly a pistol could be fired by one shooter and then handed off to and fired by another shooter. The results were compared with an eye-witness account, claiming that two shooters passed along one firearm, and with the time lapse between shots from the 911 call. Additionally the video recording capabilities of common smartphones were tested and they seem to be able to make useful recordings of firearm shots [77].

2.1.2. Serial number restoration

A simulated serial number was stamped into samples of stainless steel by hand-hammering, after which they were polished until no trace of the serial number was visible. A method using electron backscatter diffraction (EBSD) in a scanning electron microscope (SEM) is used which allowed for clear visualisation of the erased serial number up until a depth of 760 µm below the surface [78]. This method has not yet been tested on obliterated serial numbers from real firearms.

2.1.3. Microstamping

Grieve (2013) studied whether SEM could be utilised to distinguish more of the six digit alpha-numerical identifiers stamped into the primer by firing pins modified with a gear code. SEM allowed more of the alpha-numerical identifiers and the gear code structure to be distinguished than standard optical microscopy, but complete recognition was still not possible in all cases [79].

Nedivi (2014) questions the added value of microstamping to help solve firearm-related crimes and/or prevent them. Different arguments for this scepticism are explained such as, 1) that law abiding citizens, who will be the ones to buy these firearms, are not usually involved in crimes, 2) that awareness of this identification system would bring owners of these guns to remove the code from the firing pin, and 3) clogging of the gear code will inhibit good reproducibility [80].

2.1.4. Time since discharge

Factors determining local atmospheric corrosivity and the assessment of corrosivity according to ISO standards 9223, 9224, 9226, 8407 and 8565 are explained. Coupled with the visual results from an 18 month atmospheric corrosion study of six different ammunition samples this can serve as a reference for rough estimations of time since discharge [81]. For the examination of shorter times since discharge modern thermal imaging cameras might be useful. A study showed that it was possible to detect residual elevated temperatures of recently discharged rifles and handguns out to at least one hour after firing. Also movement of firearms, which were in thermal equilibrium with their environment, from colder to warmer environments and vice versa could be distinguished [82].

2.1.5. Firearms and pregnancy

The influence of exposure of pregnant women to lead and loud noise during e.g. test firing on the developing foetus is studied through a literature review. The results of previous studies seem to be contradicting each other. The author recommends additional studies and precautionary safety regulation until the risks are clear, stating "*is it worth the risk?*" [83].

2.1.6. Firearms and ammunition (mis)match

Although in most casework the calibre of the used ammunition corresponds with that of the used firearm this is not always necessary. One such an example is shown for a case where 9mm Luger cartridges were loaded in a pistol chambered for .357 SIG. Test firing was conducted and did not result in failure of the cartridge case wall, and produced bullets with a relatively high, consistent muzzle velocity when compared to other large bore / small bullet mismatches such as 9mm Luger in .40 S&W pistols or .40 S&W in .45 ACP pistols [84].

Another example is where .38 Smith & Wesson cartridges were loaded in the magazine of a .357 Sig pistol. These cartridges were found to feed and chamber but required manual extraction and ejection. The cartridge cases showed extreme expansion due to the larger chambers [85].

Instead of combining smaller calibre ammunition with larger calibre firearms, a case report on an examination on a 12-gauge shotshell shows that it is also possible to replace the load of the shotshell with dimes (US 10 cent coins) and fire these with lethal consequences [86].

In a case where a Florida panther was shot with 000 Buckshot the question was whether these were fired with a rifled or smoothbore barrel. Several .410 bore 000 buckshot shells were examined and fired through a shotgun and a Taurus Judge® revolver. The conclusion was that the pellets were fired through a smoothbore barrel [87].

2.2. Ammunition

2.2.1. Manufacturing marks

Winchester Australia (Limited) is producing a line of 9mm Luger remanufactured cartridges marked as “recycled centrefire cartridges”. These are reloaded cartridges from the Australian Federal Police utilising Glocks. The cartridges show cycling/firing marks from these Glocks but also reloading marks. On the case walls fine striations running parallel to the length of the case from the mouth were seen, circumferential marks below these and what appeared like a shallow channel that was aligned with the long axis of the cartridge case. The cartridge cases also displayed shell holder marks within the extractor groove. The primers also showed impressions marks [88].

Manufacturer marks are also seen on Remington 380 Auto calibre cartridge primers [89]. These types of marks should not be confused with firearm related marks.

2.2.2. Trace analysis

Lead isotope ratios in bullets or bullet fragments can be used to distinguish between different sources of bullets. This has been tested using multicollector inductive coupled mass spectrometry. To be able to assess the evidential strength it is imperative to study the variability within and between boxes of ammunition [90].

Another approach to analyse bullets or bullet fragments is by using X-ray fluorescence spectrometry, as has been shown for fragments of possible lead-free bullets manufactured by Hornady Manufacturing Company [91].

An article reviewing the more common constructions and compositions of modern tracer bullets also addresses how bullet holes in clothing and other object can be recognised to have resulted from tracer bullets with the use of scanning electron microscopy-energy dispersive spectroscopy and elemental mapping [92].

Using qualitative and quantitative X-ray fluorescence spectroscopy the elemental compositions and ratios of copper and zinc from commercially loaded ammunition were compared. Some ratios were specific to a particular manufacturer while others were more generic [93].

Environmental scanning electron microscope with energy dispersive X-ray spectroscopy was used to study the composition of pellets sold as lead free metal .177 (4.5 mm) calibre air gun pellets. These pellets proved to be lead-free. The authors state that a weight difference was observed between lead and lead free pellets which could be used for presumptive identification of diabolo styled lead free pellets. Apart from this, the article describes some references to wound ballistic case reports [94].

2.2.3. Frangible and disintegrating ammunition

Describing a case study involving the use of a 12-gauge breaching round and subsequent injury to a suspect the author also provides information on the manufacturer, the breaching rounds and the shotguns used for door breaching [95].

The Barnes *Varmint Grenade* rifle bullets are described. These bullets contain no lead and rapidly disintegrate upon entry in soft tissue. They are designed for varmint hunters to be used in high velocity rifles. Shooting larger animals will result in non-perforating wounds with short wound paths. The small copper disc that represents the base of the bullet' jacket can be used for identification purposes [96].

2.2.4. Manufacturer identification

Based on the results of a blind study it was shown that the deformation characteristics of air gun pellets could be used to successfully identify the manufacturer of the fired and deformed pellets [97].

2.3. Replicas and casts

McVeigh (2015) demonstrates that it might be possible to make decent replicas of marks by first creating a mold using AccuTrans® AB and then casting these with coloured glue sticks using a hot glue gun [98]. Although cheap and fast the level of detail based on visual assessment of the published photographs seems to be less than that of the more complex replication techniques utilising polymer molds and polyurethane.

Using AccuTrans® AB casts of bullets is proposed to compare the marks in the bullets. The casts will ensure one overall colour, without too much reflection and by cutting open the casts, the marks in the bullets can be compared in a flat surface. Especially when marks are hard to compare, for e.g highly deformed bullets, this might be a practical solution to be able to compare the marks [99].

Taylor (2015) shows the use of casting material and of soft sheet metal to enable the examiner to produce test marks when a firearm cannot be used for test firing. He also shows that hammering a bullet through the barrel of a firearm can create marks to compare to a fired bullet [100].

2.4. Statistics

Through a descriptive retrospective cross-sectional study, carried out on the victims of firearm injuries referred to the mortuary from August 2008 until July 2010 in the Kanpur region of central India, 3154 fatalities were registered. From these 66 resulted from firearm injuries (2.09%), of which 92.42% were males. Homicidal attacks were most often encountered, where unlicensed, illegal country-made firearms were the preferred choice [101]. Apart from the results from this specific study the article provides an overview of comparable studies worldwide and compares some of the main results, e.g. firearm related fatalities in the US vs. those in Europe.

Also in India, it is observed that about 30% of the total firearms submitted for examination consists of country-made firearms. These country-made firearms are reported to be more common in the rural areas and are usually devoid of marks or registration numbers, they look crude and unfinished, their barrels are manufactured from crude iron, they do not have standard specifications and calibrations, are smooth bores and are most often muzzle-loading firearms [102].

Based on a national database of all offenders arrested in the US, the Supplementary Homicide Reports database, it turns out that in 18% of the homicides which occur in conjunction with rape or other sexual offences a firearm is used [103].

2.5. Quality assurance documents

The National Commission on Forensic Science (NCFS, US) posted a few documents proposing policies and positions regarding various aspects of forensic science and practice. One of these documents is entitled “Scientific Literature in Support of Forensic Science and Practice” and provides six criteria for foundational, scientific literature supportive of forensic practice [104].

1. Peer-reviewed in the form of original research, substantive reviews of the original research, clinical trial reports, or reports of consensus development conferences.
2. Published in a journal or book that has an International Standard Number (ISSN for journals; ISBN for books) and recognized expert(s) as authors (for books) or on its Editorial Board (for journals).
3. Published in a journal that maintains a clear and publicly available statement of purpose that encourages ethical conduct such as disclosure of potential conflicts of interest integral to the peer review process.
4. Published in a journal that utilizes rigorous peer review with independent external reviewers to validate the accuracy in its publications and their overall consistency with scientific norms of practice.
5. Published in a journal that is searchable using free, publicly available search engines (e.g. PubMed, Google Scholar, National Criminal Justice Reference Service) that search major databases of scientific literature (e.g. Medline, National Criminal Justice Reference Service Abstracts Database, and Xplore).
6. Published in a journal that is indexed in databases that are available through academic libraries and other services (e.g. JSTOR, Web of Science, Academic Search Complete, and SciFinder Scholar).

The AFTE Board of Directors and Editorial Committee has written down their comments regarding each of the criteria in a document during its public comment stage. This was done due to the document’s potentially far-reaching influence regarding the perceived propriety and academic soundness of the AFTE Journal and articles published therein [105].

In the NCFS’ summary of the public comments they responded to these comments with the following words: “What is considered foundational literature in forensic science must be the same as what is considered foundational literature in any science. This does not mean that other journals are not of value to a given discipline. Conflict-of-interest policies are integral to the definition of foundational literature and goes beyond financial and business considerations. Indexing is critical to ensure accessibility of scientific work to any and all interested parties, as this opens the work to review, replication, evaluation, testing, confirming extension, and application through the normal process of the scientific method.” [106].

The AFTE Certification Committee have published the recently-revised certification and recertification policies and procedures [107].

A recent poll amongst the members of the Association of Firearm and Tool Mark Examiners found that a vast majority of the examiners (94%) use metaphors to clarify or simplify complex portions of their testimony either ‘often’ or ‘occasionally’. McVeigh has written down a list of these metaphors based on the information in the AFTE forums [108].

The Scientific Working Group for Firearms and Toolmarks (SWGUN) have published a few documents to provide a framework of standards:

1. SWGGUN Minimum Qualifications (Entry Level), SWGGUN Minimum Qualifications (Experienced Level) [109]
2. SWGGUN Quality Assurance Guidelines [110]
3. SWGGUN Guidelines: Projectile Path Reconstruction [111]
4. SWGGUN Guidelines: Criteria for Identification [112]

3. Legislation

Sound suppressors have been devices which, based on design and construction characteristic, have been readily identifiable as such under federal and state laws. With technological and manufacturing advancements air guns have become a rapidly growing commercial segment internationally. With increasing calibres, projectile velocities and accuracy their performance is sometimes indistinguishable from that of traditional firearms. Accessory items, such as suppressors are now commonly marketed and used on air guns. As a result international regulation is altering the traditional thinking of what a sound suppressor is and how they are evaluated [113].

4. Technical examination

Technical examination of a firearm can be useful to be able to assess the evidential values of marks on spent bullets or cartridge cases. But apart from this it might also be very useful for the assessment of statements made by victims, eye-witnesses or suspects about the functioning of the firearm. Different articles were published on modified or homemade firearms and on the reconstruction of possible events.

4.1. Modified or homemade firearms

Yasin (2013) discusses the various features (class-characteristics) of the obsolete calibre 7.92x33mm rifles which are being locally manufactured in Pakistan. Also the chambers of AK47 rifles are modified to be able to fire these cartridges. Most of these rifles can now fire both 7.62x39mm and 7.92x33mm cartridges. Especially the addition ring visible on the shoulder of the cartridge case is a useful characteristic to recognise the use of such a converted firearm [114].

A Mossberg 702 Plinkster semi-automatic rifle was examined from which the firing pin was modified in such a way that the rifle could now be fired by slam firing. Feeding a live cartridge in the chamber and releasing the bolt would consistently result in a discharge without a need to pull the trigger [115].

Another such a slam firing firearm was examined. A Bulgarian manufactured Arsenal model SLR-100H was found to fire fully automatically upon closing the bolt due to improper placement of one of the hammer springs [116].

A Mosin-Nagant bolt action rifle was submitted for function testing with a missing bolt head assembly. It turned out that it was possible to fire a primed cartridge case in this state and it was even possible to fire a bullet through the barrel with potentially lethal consequences [117].

The way in which a Japanese type 99 Light Machine Gun can be disassembled is discussed after an examination of such a firearm with a missing firing pin [118].

Apart from changes to firearms, silencers are also commonly found to be homemade. The effectiveness of the homemade silencers was compared to that of commercially available silencers, where the commercial silencers performed better at suppressing sound. In this article the operation of silencers is also explained as well as some advantages of using a

silencers apart from suppressing sound, such as increased precision by stripping away hot gases from around the bullet and a reduction of recoil through added weight [119].

Pal and Pratihari (2014) provide an overview of homemade firearms examined at their ballistics division in India. About 50% of the firearms they examine are homemade. Most of the homemade firearms are manufactured from crude and cheap materials and can vary a lot in quality [120].

An example of a homemade firearm manufactured from hardware material (copper pipe, copper by copper coupling, a copper nipple pipe and plaster casting) is described [121]. Another such a homemade firearm was found to be disguised as a Super Soaker® water gun [122].

Instead of manufacturing a homemade firearm from hardware material it is shown that a World War II flare gun can be modified to chamber a 45-70 Government cartridge and is capable of firing these [123].

4.2. Reconstruction of events

Although most firearms are designed to be safe to use some firearms can be fired under unintended conditions. One such an example is given for the FN model High Power 35. The magazine safety can be annulled by manually pushing the safety lever inside the pistol. Additionally, by manually pressing in the hammer sear a shot can be fired without pulling the trigger [124].

Glock pistols without a trigger spring appear to be inoperable if loaded and attempted to fire by pulling the trigger, however it is possible to fire them when the trigger is held to the rear prior and during loading, after which the trigger can be released and pulled again [125].

Drop tests with Raven Arms P-25 and MP25 .25 Auto calibre pistols showed that these can be discharge in that way. Especially the firearms with a thinner cam, discharge more easily than those with a thicker cam [126].

It is shown that when feeding short 12-gauge cartridges (not intended for the firearm) in a Winchester Model 1300 pump action shotgun it is possible to create a potentially unsafe firing condition. Under certain conditions the trigger and safety button can retain the hammer in a partially-cocked position, not captured by the sear. Releasing the safety will release the hammer resulting in an unintended firing of the firearm [127].

4.3. Firearms background

Detailed information about the history and the manufacturing process of the Jennings / Bryco / Jiminez Arms pistols is discussed [128] as well as the historical background, principle variants and manufacturer marks from the AK-47 and its AK variants [129]. The characteristics (design, performance, marks and GSR production) are discussed from the unique Russian 5.45mm PSM pistol and its ammunition [130].

5. Shooting incident reconstruction

5.1. Research

While reconstructing a shooting incident a lot of factors have to be taken into account. Research within this field has especially focused on reconstructing bullet trajectories, bullet ricochet behaviour, bullet behaviour and shot patterns from shotguns used to estimate shooting distances.

5.1.1. Bullet trajectory reconstruction

Based on test fires at known angles of incidence on sheetrock, wood pressboard and sheet metal, using a Walther P4, 9mm Luger pistol with full copper-jacketed bullets the accuracy and precision of the ellipse method has been studied. Both an ellipse template and callipers have been used to measure the dimensions of the primary bullet hole. The results show that the calculated angles of incidence were approximately within 10° of the known angle of incidence for angles of incidence up to approximately 60°. For higher angles of incidence the accuracy further decreased [131].

Based on shots fired with multiple calibres on laminated glass windshields, the deflection of most calibre bullets (except two from .45 ACP) when perforating the glass was lower than the generally proposed range of -5° to +5°. For these test the windshield was placed under an angle of 30° and the firearm at a downward slope of 5° [132].

5.1.2. Ricochet behaviour

The critical ricochet angle (the angle of incidence at which 50% of the fired bullets of a given ammunition type ricochet from a given object) was studied for .32 Auto and 9mm Luger FMJ bullets on different types of wooden boards. The critical ricochet angle is higher for .32 Auto bullets than for 9mm Luger bullets and increases with increasing wood density and Janka hardness. The results indicate that there is a strong linear relationship between both wood density and Janka hardness and the critical ricochet angle [133].

Instead of the more cumbersome Janka hardness test (the force in Newton required to push a 11.28 mm steel ball into wood to half the ball's diameter) another method to determine wood hardness is proposed, using steel BBs. Using this simple and inexpensive method it can be tested whether the hardness of wood found at the crime scene is the same as that used for reference testing [134].

Empirical test shots on asphalt using a Keltec model P-11, 9mm Luger pistol and FMJ bullets showed that at angles of incidence of 20°, 30° and 45° the bullet was destabilised upon impact and tumbled post ricochet. All bullets were fragmented to varying degrees, increasing with increasing angles of incidence. Ricochet angles seemed fairly consistent for different angles of incidence and were lower than the respective angles of incidence [135].

5.1.3. Bullet behaviour

The penetration depth of bullets in solid material, including bodies of firearm victims, wood of trees, and certain construction materials or objects, can be used to assess the impact velocity of the bullet. By performing empirical tests with downloaded cartridges this assessment of the impact velocity can be used to assess the shooting distance [136].

While most studies on bullet behaviour during flight focus on stable bullets there is essentially no data available on instable bullets. Haag (2013) studied the behaviour of ricocheted, destabilised or instable bullets. These bullets showed to have a very high aerodynamic drag due to their yawing and/or tumbling in the air, where deformation of the bullet only added to this drag. Ricochet, deflection and perforation events and shooting bullets through oversized bores reduced the bullet's velocity. The differences in calibre, bullet weights and designs had no major influence on the bullet's effective ballistic coefficient (BC) when they were destabilised or tumbling in flight. The BCs of destabilised and/or tumbling bullets are many times lower than those from stable flying bullets (example shows a factor 10), in the order of 0.02 to 0.03, regardless of calibre and bullet weight and design [137].

The so-called 1 pound and 1 inch diameter “standard bullet” served as the basis for the development of the ballistic coefficient (BC). While weights of modern bullets deviate highly from this standard bullet it is still widely used. A scaled down, proportionally correct reduced standard bullet (RSB) is studied and the use of ballistic radar tracking to assess the BC is shown [138].

5.1.4. Shot patterns

A literature search on the relation between shotgun barrel length and shot patterns size revealed discrepancies in the results. Additional tests were performed which were unable to identify a specific relationship. When cutting down the barrel down past the choke construction, the diameter of the resulting shot patterns varied depending on the used ammunition and from test shot to test shot [139].

Apart from the length of the barrel, intermediate target materials can also have an influence on shot pattern distributions, affecting shooting distance determinations. Test with an 12-gauge shotgun and number 0 and 9 pellet cartridges showed that the spread of pellets was significantly larger in the presence of all intermediate target materials (glass, mica auto glass, tempered auto glass, fibreboard, flat iron, aluminium and grey cotton fabric) for pellet number 9 ($p < 0.05$), but not for grey cotton fabric with pellet number 0 ($p = 0.33$) [140].

5.2. Methods

A method is explained where for the bullet trajectory reconstruction of e.g. a car no setting up of a “box” around the car, using tripods and strings, is necessary. Taping paper to the ground, running parallel with the car is suggested as a method to overcome minor problems with strings that might impede easy access to the car. Using plumb bobs, the determined trajectories can be projected on the paper [141].

Especially when evidence is not abundant on a crime scene even casts from bullet holes in e.g. sheet metal can provide valuable information for the examiner. Bullet calibre might sometimes be assessed, but also class characteristics and sometimes even individual characteristics, sufficient in detail for comparison with test fires [142].

5.3. Case reports

Apart from studies on different parameters that influence reconstruction work some casework examinations show how the results from such studies can be used in practice.

New York City’s notorious Son of Sam shootings are presented including the microscopic comparison of the marks [143].

Based on a bullet trajectory reconstruction a likely scenario of events could be built around the conclusions in a case regarding a self-defence claim [144].

Approximately 50 years after the assassination of the 35th President of the United States, John F. Kennedy, multiple publications have been written about this event. The content of these publications varies, and some discuss the proposed second shooter known as the grassy knoll, resulting from discussions about the large wound in the upper right of the President’s forehead and the sudden rearward movement of the head [145].

Haag (2014) studied the merits of this claim and concluded that the rearward movement of the head could have resulted from the acceleration and propelling of the brain matter and skull fragments forward and upward from the head. As a consequence an immediate rearward propulsion of the head is expected [146].

Focusing on the Western Cartridge Company 6.5mm Carcano bullets used for the assassination, the difference in fragmentation behaviour of the first and second gunshots are discussed [147].

Although President John F. Kennedy was hit by two bullets the unaccounted for third bullet has also been studied. Through a step-by-step review and analysis of the assassination and going into the exterior and terminal ballistics of the 6.5mm WCC Carcano bullet the author concluded that the missing shot was the first shot fired and must have hit the asphalt of Elm Street at a relatively steep angle of incidence and subsequently self-destructed [148].

The so-called “Magic” bullet in the JFK assassination that passed through two individuals and remained intact is discussed. Taking into account the exterior, terminal and wound ballistics of the novel 6.5mm WCC Carcano bullet the author explains through a step-by-step analysis of the bullet and its journey that there is nothing “magical” about the bullet [149].

The articles about the JFK assassination [146,147,149] triggered an additional letter to the editor [150], a subsequent response from the author [151] and a separate response [152].

6. Wound ballistics

During the examination of firearm related incidents multiple factors can be taken into account. The field of terminal ballistics is of importance when reconstructing the trajectories of bullets through the examination of the evidence, but there is also another source of information: wound ballistics. As a specific topic within the field of terminal ballistics the results from examinations might provide more insight into the occurred events.

6.1. Research

When the parameters of a case, including the injuries, are taken into account to formulate a conclusion about the question whether it was a homicide or a suicide, Bayesian analysis, using pooled data from a systematic review, can be used to provide information about the strength of the evidence [153].

To study the effect of the damage from direct shots or from bullet fragments on military helmets to the head, an anatomically correct skull/brain model has been developed. When using a ballistic gelatine brain or when completely filling the polyurethane skull with ballistic gelatine, the obtained fracture patterns compared favourable to the report of actual high velocity rifle wounds to heads [154].

Another study with ballistic helmets shows that there is a high risk that the deformation of these helmets upon bullet impact will cause severe injury to the head, called “behind armour blunt trauma”. The authors make a plea that this should be taking into account when designing these helmets [155].

Studying bullet hole shape after ricochet, fabric and human tissue were used. Four classifications of shape were distinguished: nearly round, polygonal, slit-like and letter-like. Shots were fired on different target materials at angles of incidence of 10°, 20°, 30°, 40° and 50°. The shape of the gunshot wound seems to be dependent on the angle of incidence [156].

Tests were performed to study the lethality of unconventional projectiles (tree branches, stones, disposable foam earplugs, cotton applicator swabs and chewing gum) fired by a M-16 assault rifle using 5.56mm blank cartridges. The respective energy per unit area J/cm^2

was calculated and the pieces of tree branches, stones, disposable foam plugs, cotton applicator swabs and un-used chewing gum were found to be potentially lethal [142].

Adding to the discussion about the lethality of air guns a study has been performed using a 4.5 mm air pistol and a 5.5 mm air rifle. Test shots on 15% transparent ballistic candle blocks at 1 m and 10 m shooting distance showed that especially shots at the neck, eyes and thorax region can cause fatal injuries based on the penetration depths. The authors recommend that kinetic energy limits should be the main point of air gun regulations [157].

Another study also looked at the injury potential of fired air pellets. Test shots were fired on different combinations of ballistic gelatine and on ballistic gelatine with embedded animal organs and/or applied animal skin. Some of the possible physical parameters are discussed that might help to predict the degree of damage, but from the study it was not possible to define a limit which could be proposed as safe [158].

According to the Polish Weapons and Ammunitions Law air guns with a kinetic energy of the fired pellet below 17 J are not regarded as firearms. Test using 20% ballistic gelatine were performed at various shooting distances and the results were compared to autopsy findings. The findings demonstrated that these air guns can cause serious injuries and that the gelatine blocks do not fully reflect the properties of the human body [159].

Using ballistic gelatine blocks the injury potential of air gun pellets was studied. Loosely applying different items of clothing on the gelatine blocks decreased the injury potential [160].

An uncommon sabot air gun projectile was studied. The results showed that the plastic sabot cup surrounding the sub-calibre copper-coated lead projectile discarded well in flight for high energy air guns ($E > 17$ J). Separation was also observed for low-energy air guns ($E < 7.5$ J). While the velocity was similar to that of a diabolo type reference pellet (RWS Meisterkugel) the energy density of the pellets was up to 60% higher [161].

A study on the effects of a 9mm Luger bullet's penetration was done by comparing the behaviour seen in ballistic gelatine with that in a numerical simulation model that was built using the finite element method (validated through the test shots). The response of the ballistic gelatine in the process of bullet penetration can be divided into four stages: 1) the smooth attenuation, when there is no bullet instability, 2) the bullet's rolling stage, when the bullet rolls due to instability, its velocity drops sharply, and the kinetic energy is rapidly transferred to the gelatine, 3) the full penetration stage, when the bullet penetrates through the gelatine in a relatively stable backswing position and 4) the expansion and contraction stage, when the temporal cavity in gelatine continues to expand and then contracts. The effect of the bullet's impact velocity and angle of incidence on the temporal wound cavity, its velocity attenuation, its rolling angle, its resistance and energy variation were investigated [162].

The relation between the kinetic energy of contact shots to the head and bursting of the head was studied. 35 cases were examined and compared with respect to firearm, ammunition, entry site and projectile energy. Bursting, disruption of at least 50% of the head, was associated with energies < 2700 ft-lbs in 12/22 cases and energies > 2700 ft-lbs in 13/13 cases. No relation between bursting and either wound site, ammunition type or projectile fragmentation was found [163].

Studying the wounding potential of number 8 shotgun pellets fired by a 12-gauge shotgun it is shown that fabrics reduce the penetration of pellets into tissue. This effect was greater at

increasing shooting distance (beyond 40 yd (36.6 m) and for thicker denim and cotton fabrics [164].

To investigate the trauma potential of pistol crossbows different tests were performed on these crossbows which operate on remarkable low energy levels [165].

A study on the impact parameters and efficiency of 6.8/15 calibre captive bolt guns shows that the bolt velocity ranges from $v = 42$ to 54 m/s and that the kinetic energy values range from $E = 224$ to 369 J. The efficiency of the captive bolt stunner (ratio of the kinetic energy of the stunner's bolt to the potential energy of the industrial blank cartridge) was found to vary between 36 and 46% [166].

An experimental and numerical study on the indirect effect of rifle bullets on bones shows that when the velocity of bullets increases, the stress on bones also increases. Depending on the distance between the bullet and the bone, the bone will or will not fracture, but is in both situations affected by the stress wave [167].

Examining contact shots has shown that muzzle imprint marks can be seen on the entrance wound of victims. It has been accepted that these result from intruding powder gasses expanding under the skin which as a results balloons back against the muzzle. The current study shows that in a considerable number of cases not only excoriations (abrading or wearing off the skin) but also intradermal haemorrhages may cause an imprint pattern reflecting the muzzle, its relief and/or contours [168].

Full metal-jacketed rifle bullets with lead cores and open bases can experience deformation as they jaw during soft tissue penetration. The amount of deformation depends upon the strength of the bullet and the velocity upon penetration when it goes into jaw. The jaw behaviour of the bullet depends upon its design (length, ogive shape, ogive length, centre of gravity, and pre-impact stability) when it penetrates soft tissue. The specific relationship between bullet deformation and bullet velocity must be worked out through empirical testing [169].

A database search through the Miami-Dade Medical Examiner Department's computer database for homicides from 1997-2011 resulted in a total of 2647 homicides from firearm injuries. Two of these cases (both on January 1st) fit the criteria for fatal injuries from celebratory gunfire, "falling bullets" [170].

6.2. Case reports

Two fatal cases involving discharged spherical lead projectiles fired from muzzle-loading blank powder firearms are discussed. In contact and close-range shots the deposition of GSR is greater than for similar shots with smokeless powder. The wad might be found in the wound channel. Apart from this the mostly spherical shape of the projectiles cause maximum tissue damage at the entrance side. Similar results with penetration depths up to 25 cm were seen in ballistic soap covered with pig skin [171].

To be able to compare the injuries found during autopsy to those of the suspected black powder replica of a Colt Navy of 1851, test fires were made while shooting at 20% ballistic gelatine blocks. Solid spherical projectiles caused extensive injuries, especially in the initial segment of the wound canal. Based on the presence and location of the wad in the wound channel the shooting distance could be also determined [172].

Tests using ballistic gelatine and the suicide victim's blank firing pistol showed that the extent of the temporary cavity after firing 5 g of black powder roughly corresponded to the wound cavity in the precordial region of the deceased [173].

The lethality of air rifles is demonstrated by a fatal victim following a shot through the heart by a 0.177 (4.5 mm) calibre pellet. The pellet, with an energy density of 1.9 J/mm² penetrated through two layers of cotton fabric and several layers of tissue [174].

The distance of 60 ft. (18 m) between a shotgun and a suspected suicide victim, by a head shot, triggered a literature search [175]. Multiple publications were found reporting cases of victims who were able to act following penetrating ballistic head injury. The authors discuss that three conditions seem to be necessary to support the hypothesis of delayed incapacitation: 1) the use of a slow and lightweight projectile with low velocity, 2) absence of injuries of vital and motor areas, and 3) absence of evidence of overpressure injuries.

A case is described where a man was killed by a 9mm calibre gunshot wound to the head. The skull showed a "keyhole" defect which occurs when a bullet strikes the skull tangentially, usually fragmenting the bullet [176].

7. Training material and books

Since 2008, the NFSTC has put a firearms examiner training course online [177]. This course was made in collaboration with AFTE members and is based on the AFTE training manual.

The book *Their Arrows will Darken the Sun: The Evolution and Science of Ballistics* by Mark Denny [178] has been reviewed by James L. Roberts [179]. In his review he states that the book is easy to read and covers the subject well. Apart from this he says that it is written from the perspective of a physicist, which might sometimes be confusing for an examiner because of the use of different nomenclature. The review responds to some errors in the book and appeals for a second edition.

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Criminalistics

Forensic Geosciences,

2013-2016

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1. Introduction

The objective of this review is to provide peer reviewed publications, books and other activities on forensic geoscience (geology, geosciences and soil science) since the previous review carried out in 2013 (1). This review is based on articles in academic journals, books and book chapters, reliable internet resources, academic societies' web pages and publications, police and forensic magazines and mainstream publications. The number of cited peer reviewed journal articles was over 250. This review will refer only to related conference proceedings. It covers the period from the date of completion of the last review from 1st January 2013 to 1st June 2016.

This field of study is experiencing a period of expansion. Published articles alone would not cover all of the advances in forensic geology that have been achieved over this time. In the 20th century, it was rare to find forensic geology papers and presentations in any media, but it is now easier to find them as the numbers and access to them through the internet have increased significantly. One of the reasons for this rapid expansion in the development of forensic geology/soil science is due to the establishment of global organizations such as the International Union of Geological Sciences (IUGS) (2), Initiative on Forensic Geology (IFG) (3). This group was established specifically to promote and develop forensic geology globally.

The term 'forensic geology', also referred to as 'geoforensics' and 'forensic geoscience' and their definition is still subject to some debate. Ruffell (4) has reviewed these definitions and pointed out that forensic geology now includes a range of sciences related and/or applied to the forensic discipline. This review includes the allied disciplines of geology (mineralogy, sedimentology, microscopy), geophysics, geomorphology, soil science, microbiology, anthropology, and taphonomy, all which have been used successfully as tools to aid forensic (domestic, serious, terrorist and international) crime investigation. 'Forensic' means pertaining to the law.

The main areas where the geosciences contribute to forensic provision are as trace evidence, and also in crime reconstruction and search, aiding the police and criminal justice systems, while also delivering on aspects such as environmental forensics (e.g. 5), wildlife crime, and counter terrorism.

This review is divided into several sections, broken down into specific topic areas, which relate to forensic geoscience. Evidence, Geophysics, Soil DNA, Search, Environmental Forensics, Method Development and Taphonomy will be featured, as these are the areas that have contributed most of the scientific papers over the period of this review. In addition, a review of developments and activities globally in forensic geoscience will be outlined. Papers which were not linked to geology or soil science were not included. The first section predominantly covers refereed publications, while the second section of the review predominantly outlines the related books and book chapters.

2. Refereed Publications

2.1 Evidence

Over 10% of the published papers had a focus on evidence (6 to 22). These consider both inorganic analysis (16) and organic approaches (e.g. numbers 7, 12), with several focussing on Soil DNA, an area of growing research interest (e.g. 13 and see separate section). Some papers have also proposed forensic lab approaches for the provision of evidence for Australian soils (20 to 22).

Some papers have explicitly looked at evidence in relation to transfer and persistence (e.g. 23 and 24). Others have focused on transfer in relation to pollen; these papers can be found in the forensic botany section below (e.g. 25, 26).

2.2 Method Development

Over 15% of the peer-reviewed papers have covered new developments. New method development has spanned the areas of organics (e.g. 27), isotope chemistry (e.g. 28) and soil microbiology/DNA (e.g. 29), sample handling approaches (e.g. 22, 23 and 30) and in relation to infestation estimates (31) and search (32, 33). There has been interest in inorganic characterisation (e.g. 34 to 35 and 39), with application of synchrotron radiation as a tool being tested in Japan (36).

2.3 Soil DNA

Interest has risen recently in the use of soil DNA in the context of its use as soil trace comparisons and as potential evidence, including consideration of a wide range of taxa (29, 30, 37 and 42 to 51 and 211); with over 20% of the refereed publications in this area. The use of specific groups of organisms such as testate amoebae for time after death estimations (40) and soil fungi in the understanding of decomposition (41) and mites (59) has been investigated. Research on tools and techniques has been carried out (e.g. 43, 44, and 45) and some validation work (46 to 58). More research is required in this interesting area before it can be used safely in court; there are still concerns over extraction effects and temporal and spatial effects.

2.4 Botany

Papers which have focussed on forensic botany and palynology comprise less than 5% of the total number of papers in forensic geosciences. Protocols and case examples have been described (61 and 62), and papers on the use of palynology for use in indoor crime scenes has been developing (25, 26 and 60). The role of forensic botany in crime scene investigation has been reviewed (63).

2.5 Environmental Forensics

Over 12 % of the papers were in the area of environmental forensics (e.g. 67 to 82). Environmental (as well as criminal) case work and related research has benefitted from advancement in forensic geoscience. A review of environmental versus criminal application

can be found (5). Some papers, focusing on inorganic analysis (11, 12 and 16), have considered the use of inorganic databases relevant to environmental forensics (1 and 2).

Unfortunately environmental crime is still a major problem in nations where waste has been buried (e.g. Ireland, UK, Italy), and is an increasing problem with surface dumping of waste. A recent report was published by [UNEP-INTERPOL](#) concerning the rise of environmental crime (64), and networks have focussed on the topic (e.g. 65, 66). Organisations such as SEPA (Scotland) and EA (England) help police and legislate such issues.

2.6 Search

The numbers of publications in the area of search increased significantly. The application of geology to search has taken place since the 1990s, when Laurance Donnelly and Mark Harrison applied geological methods to search for burials in the United Kingdom. As a result, this work has significantly advanced police and law enforcement ground searches for graves and other buried items. Considerable advancement has been made in method development to improve search capabilities, from work on source location using databases (83), VOC detection (e.g. 84 to 96 and 106) to elemental profiling (66). Over 10 % of the papers focussed on such developments.

2.7 Geophysics

Over 15% of the papers spanned the subject of geophysics. Controlled studies using geophysics have advanced considerably over the period 2013 to present, with long-term monitoring of controlled sites having taken place (112 to 116), chiefly by Keele University research groups led by Jamie Pringle in the UK. Ground Penetrating Radar (GPR) and electrical resistivity (ER) appear to be suitable, with winter surveys optimal for such resistivity surveys. Conductivity of Leachate plumes show that values return to normal water levels after 5 years (38). Burial style, soil type and local depositional environment are all key variables needing further consideration (107, 108, 109 and 111). In addition, book chapters also covered this subject area (117 and 118) in addition to a range of related journal papers (119 to 125). Two papers were published on the subject of geomorphology (126 and 127).

In addition to the ongoing research, which is predominantly in the UK, geophysics has successfully been used over this period 2013 to 2016 in case work in the UK police (mainly through Jamie Pringle and Alastair Ruffell). Results from some casework have been published, notably from indoor surveys (98). There have been a few publications on graveyard investigations (with unmarked burials still being difficult to spot) and potential detection techniques (thermal imaging for surface remains, side-scan sonar for water searches, magnetic susceptibility as a complementary technique) (128 to 141).

Geophysical casework continues with police forces, military geophysical and remotely sensed systems and private consultants throughout the world in the UK, Italy, France, Brazil, Argentina and the Philippines. New developments involve unmanned vehicles, both Unexploded Ordnance (UXO) detection (combined EM GPR platforms in remote vehicles linked by bluetooth and or radio with cameras at front and end, currently in Syria).

Geophysics continues to be placed on drones with gamma detectors for nuclear waste/old arms dumps. Spatial location remains at the forefront with increasing accuracy of Global Navigation Satellite System (GNSS), making all geophysics linked in XY space. Underwater imaging advances in similar ways with the development of unmanned seabed and sidescan sonar for side-looking imaging in zero visibility such as the Codaoctopu.

Two recent conferences which featured forensic geophysics were: the European Meeting of Forensic Archaeologists at the new Gendarmie Headquarters (Pontoise, Paris, France, August 2015), and the Geological Society of London meeting held on 3rd December 2014,

entitled 'Forensic Geoscience: Future Horizons'. Other recent conferences on forensic science have included talks on the use of geophysics, e.g. at the Australian New Zealand Forensic Science Society. In addition, geo archaeology was the specific topic of a meeting held in Italy (232).

2.8 Taphonomy

An aligned area to forensic geoscience is taphonomy, which crosses also the boundaries with pathology, anatomy, biology, anthropology and archaeology. There are an increasing number of papers in this area, and one major advancement in this is the setting up of the new human decomposition facility in Australia led by Shari Forbes – the Australian Facility for Taphonomic Experimental Research (AFTER) (142, 143). Already this group are providing new expertise for helping to locate and recover buried evidence, including human remains, drugs, weapons, and currency (see training section below). 30% of the papers could be categorized as relating to assisting with understanding the taphonomic processes (144 to 183).

2.9 Other related publications

Other areas of interest to forensic geoscience which have been published in this period relate to: blast residue in soil (184), statistics (185), plant and animal provenance (186), use of fungi in dust tracking (187), engineering (188) and ballistics (189).

3. Book publications

Several key books and book chapters were published over the period 2013 to 2016 (191 to 229). PhD theses are not included in this review. In 2013, a seminal text was produced covering the wide range of topics relevant to the subject (191). Other books touched on the topic (192 and 193), others featured environmental forensics (e.g. 210) forensic chemistry (e.g. 220) while others featured aspects of taphonomy (e.g. 196, 200, 212) and specialist areas such as diatomology (e.g. 218). In 2016 a book on forensic geoscience, aimed at university students was published in the USA (233). A book on forensic geoscience was published in Italian (230) and one in Chinese (245).

4. Education, Communication and Global training

Communication of forensic geoscience is vitally important. This can be in relation to educating the next generation of forensic geoscientists (e.g. 17) or to the general public (221 and 231). Training in the topic area of forensic geoscience has taken place in many countries across the world, carried out by a few key groups, in particular with a global emphasis, by the International Union of Geological Sciences (IUGS) (2), Initiative on Forensic Geology (IFG) (3). Some examples of activities, events and meetings where forensic geoscience played a main part are listed below.

The numbers of presentations and conferences have increased more rapidly than the previous review period. The success of each event was due to the hard work and commitment of the hosts and organizers. These events have also benefited from the increasing interest in forensic geology and related sciences. A brief summary of meetings, which included sessions on forensic geology, is presented in Table 1.

The number of presentations also introduced in this review illustrate the scope of forensic geology in terms of the techniques, discussions, and internationally growing recognition of the importance of forensic geology. Most of the presentations were presented in English in the previous review; the number of non-English language reports/papers/books has increased significantly.

Year	Date	Conference Title	Conference Link	Venue	Country
2013	18-23 February 2013	65th Annual Meeting of the American-Academy-of-Forensic-Sciences	http://www.aafs.org/meetings/future-past-aafs-meetings/	Washington, DC	USA
2013	07-12 April 2013	Soils and Human Health, European Geosciences Union General Assembly	http://www.egu2013.eu/	Vienna	Austria
2013	15-18 May 2013	Third International St Petersburg Legal Forum		St Petersburg,	Russia
2013	10-12 June 2013	INEF Penn State Conference 2013. A conference for the Environmental Forensics Community	http://forensics.psu.edu/news-events/events/2013/inef-penn-state-conference-2013	The Pennsylvania State University, University Park	USA
2013	16-Jul-13	International Crime Science Conference	https://www.ucl.ac.uk/jdi/events/crime-science-conf/icsc-2013	The British Library, London	UK
2013	2-7 September 2013	25th Congress of the International Society for Forensic Genetics, (ISFG)	http://www.isfg.org/Meeting	Melbourne Convention and Exhibition Centre	Australia
2013	16-Sep-13	Workshop - Improving the Death scene Investigation: Advanced Multidisciplinary Approaches and their Use in court		Universita di Pavia	Italy
2013	27-Sep-13	Northeastern Association of Forensic Scientists (NEAFS), 39th Annual Meeting.		Cromwell, Connecticut	USA
2013	7-9 October 2013	2nd International Conference on Forensic Research and Technology. Forensic Research - 2013	http://forensicsresearch.conferenceseries.com/pdfs/forensic-research-2013-3567-scientific-program.pdf	Jampton Inn Tropicana, Las Vegas, Nevada	USA
2013	08-10 October 2013	17th Interpol International Forensic Science Managers Symposium. Interpol General Secretariat	http://www.interpol.int/INTERPOL-expertise/Forensics/Forensic-Symposium	INTERPOL General Secretariat Headquarters, Lyon	France
2013	14-15 October 2013	Biometric Technologies in Forensic Science	http://www.ru.nl/clst/btfs/btfs-2013/	Nijmegen	The Netherlands
2013	21-25 October 2013	The Russian Federal Centre of Forensic Science (RFCFS) of the Ministry of Justice of Russia and IUGS IFG Soil Forensics and Forensic Geology' Knowledge Transfer event	http://www.forensicgeologyinternational.com/	Moscow	Russia

Year	Date	Conference Title	Conference Link	Venue	Country
2013	06-08 November 2013	The Forensic Science Society and California Association of Criminalists Joint Autumn Conference. Forensic Horizons 2013	http://ialm.apf.it/component/kunena/61-international-meetings/492-the-forensic-science-society-a-california-association-of-criminalists-joint-autumn-conference.html	The Mercure, Manchester Piccadilly	UK
2013	24-27 November 2013	2nd International Conference on Engineering Geophysics and Forensic Geology	https://www.eage.org/events/index.php?eventid=898&Opens=3	United Arab Emirates University, Al Ain	United Arab Emirates
2013	09-10 December 2013	1st Annual International Conference on Forensic Science Criminalistic Research (FCSR 2013)		Hotel Fort Canning, Singapore	Singapore, Southeast Asia
2014	17-22 February 2014	66th Annual meeting of the American Academy of Forensic Sciences	http://www.aafs.org/meetings/future-past-aafs-meetings/	Seattle	USA
2014	01-03 April 2014	The Annual Conference of the International Association of Law and Forensic Science (IALFS). The 1st International association of forensic Toxicologists (T.I.A.F.T.) Middle East Congress	http://www.ialfs.org/ialfs2014/index.php	Ibis World Trade centre, Dubai	United Arab Emirates
2014	08-13 June 2014	20th World Congress of Soil Science	http://www.20wcso.org/	Jeju	Korea
2014	09-11 July 2014	GeoEnv 2014, 10th Conference on Geostatistics for Environmental Applications	http://2014.geoenvia.org/	Paris	France
2014	16th July 2014	8th International Crime Science Conference	https://www.ucl.ac.uk/jdi/events/crime-science-conf/icsc-2014	British Library, London	UK
2014	18-24 July 2014	IX National Congress of Geology (IX CNG) and 2 Geology Congress of Portuguese Speaking Countries (2nd CoGePLiP)	http://www.icog.es/iageth/index.php/p3loki-by/	Faculdade de Ciências da Universidade do Porto	Portugal
2014	04-06 August 2014	International Network of Environmental Forensics, INEF	http://www.rsc.org/events/detail/11468/INEF%20Cambridge%202014	St John's College, Cambridge	UK
2014	31 August - 4 September 2014	22nd International Symposium of the Forensic Sciences of the Australian and New Zealand Forensic Society (ANZFSS 2014)	http://www.aomevents.com/ANZFSS2014	Adelaide Convention Centre, Adelaide	Australia

Year	Date	Conference Title	Conference Link	Venue	Country
2014	6-8 October 2014	3rd International Conference on Forensic Research and Technology. Forensic Research 2014		San Antonio, Texas	USA
2014	13-18 October 2014	20th Meeting of International-Association-of-Forensic-Sciences (IAFS) part of World Forensic Festival (WFF2014)	http://www.wff2014korea.org/s	COEX, Seoul	South Korea
2014	24-26 October 2014	The British Diatom Meeting	http://www.isdr.org/meetings/british-diatom-meeting	Baskerville Hall, Hay-on-Wye, Wales	UK
2014	07-08 November 2014	UK Chartered Society of Forensic Sciences Annual Autumn Conference: The changing face and pace of trace evidence	http://www.bafs.org.uk/index.php/events/upcoming-external-events/351-chartered-society-of-forensic-sciences-events	University of Leicester	UK
2014	02-04 December 2014	Forensic Geoscience Group and the International Union of Geological Sciences-Initiative on Forensic Geology	https://www.geolsoc.org.uk/expired/NSGG-FGG-Forensic-Geoscience-Future-Horizons	Geological Society of London, Burlington House, London	UK
2015	02-03 February 2015	The Paradigm Shift for UK forensic Science	https://royalsociety.org/science-events-and-lectures/2015/02/forensic-science/	The Royal society, London	UK
2015	16-21 February 2015	67th Annual Meeting of the American-Academy-of-Forensic-Sciences	http://www.aafs.org/meetings/future-past-aafs-meetings/	Seattle	USA
2015	21-22 May 2015	ENFSI Annual Meeting 2015	http://www.enfsi.eu/agenda/enfsi-annual-meeting-2015	Pontoise	France
2015	30 June -2 July 2015	FORensic RESearch and Teaching , FORREST 2015	http://www.theforensicinstitute.com/	Glasgow	UK
2015	3-6 August 2015	5th International Network of Environmental Forensics (INEF) Conference		University of Toronto, Ontario	Canada
2015	19-20 August 2015	International Conference on Environmental Forensics 2015 (iENFORCE2015)		Putrajaya	Malaysia
2015	25-27 August 2015	National Institute of Justice Impression pattern and Trace Evidence Symposium	https://www.forensiccoe.org/Community-Involvement/NIJ-IPTE-Symposium-2015	San Antonio, Texas	USA
2015	28-29 August 2015	4th European Meeting on Forensic Archaeology (EMFA) 2015	http://www.enfsi.eu/agenda/european-meeting-forensic-archaeology-emfa-2015	Pontoise	France

Year	Date	Conference Title	Conference Link	Venue	Country
2015	31 August -5 September 2015	26th Congress of the International Society for Forensic Genetics, (ISFG)	http://www.isfg.org/Meeting	Krakow	Poland
2015	06-11 September 2015	7th European Academy of Forensic Science Conference, (EAFS 2015) Pushing boundaries, working beyond borders	http://www.eafs2015.eu/invited-speakers.htm	Prague Congress Centre, Prague	Czech Republic
2015	28-30 September 2015	4th International Conference on Forensic Research and Technology 2015. From Evidence to Verdict	http://forensicrosearch.com/conferenceseries.com/	Hilton Atlanta Airport, Atlanta	USA
2015	15-18 November	International Conference on Engineering Geophysics:	http://conferences.uaeu.ac.ae/iceg/en/	Crescent Building, UAE University	UAE
	2015	Leading through Creativity, Innovation, and Sustainability			
2016	22-27 February 2016	American Academy of Forensic Sciences, 68th Annual Scientific Meeting. Transformation: Embracing change.	http://www.aafs.org/meetings/aafs-68th-annual-scientific-meeting-las-vegas-nv-2016/	Las Vegas, Nevada	USA
2016	19-20 April 2016	Forensic Expo 2016	http://www.csofs.org/Events/Forensics-Europe-Expo/28510s	Olympia, London	UK
2016	17-22 April 2016	European Geosciences Union General Assembly 2016	http://www.egu2016.eu/	Vienna	Austria
2016	12-13 May 2016	The Forensic and Policing Services Association Conference (FAPSA) 2016. Collaboration, Innovation and Identification: The Future of Forensics	http://www.fapsa.org.uk/fapsa-conference-2016/	College Court, Leicester	UK
2016	5-7 July	FORensic RESearch and Teaching, FORREST 2016	http://www.theforensicinstitute.com/training/forrest-conference/forrest-2016	The Lighthouse, Glasgow	UK

Table 1. Summary of meetings that included sessions on forensic geology (accessed 11 AUG 2016).

The International Union of Geological Science (IUGS) - Initiative on Forensic Geology (IFG) held various training and education events. The IUGS-IFG aim is to develop forensic geology internationally and promote its applications. The specific objectives of IUGS-IFG are to: 1) Collate and disseminate data and information on forensic geology applied to policing and law enforcement, criminal, environmental and civil investigations. 2) Promote international meetings, seminars, conferences and training. 3) Develop a 'Committee' to act as principal advisers, collaborators and active participants. 4) Develop an international

network whereby each 'member' will act as a principal contact in their respective country for the collation and dissemination of information on forensic geology. 5) Collate, make available and where appropriate review any existing documentation and publications in forensic geology. 6) Produce a document endorsed by the Committee to be called 'A Guide to Forensic Geology'. IUGS-IFG achieves the aim and objectives by various activities. These include for example the delivery of knowledge transfer, training and outreach events throughout the world, by publications and by the provision of information on the IUGS-IFG web site (3).

5. Australia

Shari Forbes in Australia, in addition to establishing the AFTER facility, has focused on developing improved search techniques (85 to 105). This group has worked with the NSW Police Force (Dog Unit, Homicide Investigation Unit, Criminal Investigations Unit, Expert Referral Team), the Queensland Police Services (Homicide Investigation Unit), the South Australia Police (Major Crime Investigation Branch), the Australian Federal Police (Imagery and Geomatics Unit), the US Department of Homeland Security (Laboratories and Scientific Services Directorate, Customs and Border Protection), the Belgium Federal Police (Institut National de Criminologie et de Criminologie), and the Brazilian Federal Police. Many of the above events were held in conjunction with the International Union of Geological Sciences, Initiative on Forensic Geology (IUGS-IFG).

Research has been carried out by Rob Fitzpatrick and his team (229, 230, 234), at the Centre for Australian Forensic Soil Science (CAFSS), and by Brenda Woods (AFP) (21, 22 and 23) developing an approach for use by criminalistics experts for the first line examination of soils. The key purpose of the work by AFP has been to raise awareness of the potential for soil examination, and to have a working protocol for the initial examination and triage of soils to make better use of subject specialists down the line. This work is now being applied in the AFP forensic laboratories, the suggested protocol having been validated for Australia through their quality system.

6. Europe

In Europe, forensic geoscience is a very vital area of application in both search and in the provision of evidence.

Listed below in chronological order are examples of forensic geoscience events and activities. Only those with sessions on forensic geoscience are listed and it is not meant to be fully comprehensive.

Search training, for the Serious Organised Crime Agency, SOCA, was held in Leicestershire, UK, March 2013. The objective for the conference was to raise awareness regarding the support and guidance available from experts operating within a range of academic disciplines, in the search for missing persons and 'no-body' murder victims. This event was intended for UK Police Search Adviser (PoISA) and the search community. It provided specific information regarding the contribution that forensic geology, enviro-archaeological profiling, archaeology, anthropology and biology, can make in the development of search strategies and tactics. Additional expert advice and display materials were available to describe the application and use of geophysical search equipment, victim recovery dogs, and the support available from the National Missing Person Bureau and National Missing Person Adviser. (See 224, 226 and 229).

In 2013, the International School Science Fair (ISSF) came to the United Kingdom (UK) for the first time, being hosted at Camborne Science and International Academy, Cornwall. A workshop was held which comprised three main events: (a) the World Soil Map 2013 project; (b) a simulated crime scene and geological trace evidence recovery and analysis exercise and (c) a geological ground search for burials, such as weapons and items commonly used in crime, using geophysics.

Forensic geoscientists participated in the European Geosciences Union (EGU) General Assembly meeting in Vienna (Austria, April 2013), and at the 1st Heavy Mineral School, University of Milan, Italy, May 2013. On 1st and 2nd May 2013, Skip Palenik, USA, visited the University of Milan and delivered a series of training lectures and practical sessions on the subject of forensic geology. An MSc course was established in Forensic Geology in Messina, Sicily in 2015, and ran in 2016, supported by the IUGS-IFG.

From 15th-18th May 2013, the Ministry of Justice of the Russian Federation organized a session devoted to the way forensic science is presented in different countries. The session was chaired by the Director of the Russian Federal Centre for Forensic Science, and was attended by the Ministry of the Interior of Russia, the Police Service, Investigative Committee of Russia, Federal Drug Control Service of Russia and the Federal Security Service of Russia. Delegates were invited from Armenia, Australia, Azerbaijan, Belarus, Israel, Italy, Kazakhstan, Kyrgyzstan, Latvia, Netherlands, Northern Ireland, UK, Ukraine and USA.

The International Forensic Laboratory Managers meeting is held every three years, and in 2013 it took place from the 8th-10th October 2013 in Lyon France (1). This important forum brings together laboratory managers from INTERPOL countries across the world. At this meeting, Ritsuko Sugita presented an overview of forensic geoscience around the world as part of an international review on forensic science. There was also a keynote talk on 'State of the Art Forensics' in forensic science, featuring forensic geology and forensic soil science by Lorna Dawson. Both members of IUGS IFG outlined the extensive work of the IUGS-IFG. Discussion with crime scene managers was held and an exchange of methods applied in different countries was carried out.

As part of the European Researchers' Night, an event was held on 27th September 2013, at University of Roma Tre, on forensic geology 'The scientific investigation at crime scene: from CSI to forensic geosciences'. On 16th to 19th September 2013, the University of Pavia organized a workshop 'Improving the death scene investigation: advanced multidisciplinary approaches and their use in court' which included a presentation on geological materials as useful tools for search and localization. In November 2013, a meeting was held at the Servicio de Criminalística of Guardia Civil, Madrid, where there was a session on soil as evidence at the Seminary 'Análisis Criminalístico de Suelos II'.

Forensic geosciences was represented at the 'International Workshop on Forensic Science and Archaeology' held on 22nd and 23rd November 2013 at the American University of Rome by Alastair Ruffell and Rosa Maria Di Maggio. In Italy in February, 2014, the Italian Society of Environmental Geology (SIGeA), the Register of Chartered Geologist of Molise and the Register of Chartered Attorney of Campobasso, organized a conference where the first Italian book on forensic geology, written by Rosa Maria Di Maggio, Pier Matteo Barone and others was published (230).

ENFSI (European Network of Forensic Science Institutes) Animal, Plant and Soil Traces Working Group is a European working group, founded in 2010, to look at Animals, Plants, & Soils Traces in forensic science. It was established from 13 ENFSI-member institutes (from

9 different countries and 5 associate members). A meeting is held every year and Irene Kuiper (Netherlands Forensic Institute) is currently the group chair. It covers the areas of non-human DNA and includes members of forensic science laboratories. On 2nd and 4th April 2014 the Dipartimento of Earth Science of University Sapienza, of Rome, in collaboration with Forensic Science Dept. of Carabinieri organized the 3rd Working Group Meeting of ENFSI Animal, Plant and Soil Traces. The James Hutton Institute, in association with ENFSI-APST, MiSAFE: (http://cordis.europa.eu/project/rcn/108678_en.html) organized a '2014/15 Inter-Lab: Soil Forensic Intelligence and Evidence Quality Assurance Test'. The collaborative exercise was organized in order to build up a picture of the strengths of various soil forensic analytical capabilities across a wide range of participating European laboratories and to compare approaches in intelligence in the provision to investigating authorities. ENFSI-APST is now working to produce some basic guidelines to develop common standards within European laboratories.

A one-day conference focussed on 'communication', entitled 'Communicating Contested Geoscience, New Strategies for Public Engagement', was held at the Geological Society of London on 20th June 2014. Communicating forensic geology to fellow geologists, the public, media, other forensic scientists and the media can be challenging. This event provided examples of how clear and effective communication takes place between forensic geologists and the police. On 19th – 21st June 2014 the Italian Geological Society held its IV National Congress, where a session on forensic geology was organised by the Section Young Geologists in cooperation with Rosa Maria Di Maggio. The event had the aim of showing young freelance geologists the application of earth science to forensics and the related job opportunities. In May 2014, Roberta Somma, Rosa Maria Di Maggio, Laurance Donnelly and Lorna Dawson developed the 'First Level Postgraduate Master Specialisation Course in Forensic Geology'. This was the first course of its kind in Italy and has the support of the Italian Police/Carabinieri. This course was endorsed and supported by IUGS-IFG. On 28th October 2014, the Department of Science and Technology of University of Sannio, organised the workshop 'Mineralogical and petrographic techniques applied to judicial investigations' in cooperation with Rosa Maria Di Maggio and 'Team Geo Forense'. The event was aimed at introducing the application of forensic geology to academics and students.

On 2nd December 2014, The Geological Society of London, Near Surface Geophysics Group, Forensic Geosciences Group, in association with English Heritage, and supported by IUGS-IFG, held a one day conference on the applications of near surface geophysical techniques. Near surface geophysical techniques have become increasingly established in archaeological research and forensic geology and are now routinely applied in archaeological and geoforensic investigations. This multidisciplinary conference entitled, 'Future Horizons' captured shared interests between; the geological, environmental science, forensic science, geophysics, engineering, geotechnical, mining and archaeological communities for the assessment of forensic geoscience in the future. The sessions included: quality assurance in forensic geoscience, geoforensic applications in serious crime and terrorism investigations, techniques at crime scenes, environmental crime, and the issues of interpretation of geological forensic evidence.

On 19th February 2015, IUGS-IFG took part in the Geological Society of London themed events, 'The Year of Mud'. Two sessions were held entitled, 'How can Volcanoes Help Solve Crime? The venue was the Ulster Museum in Belfast. This attracted 75 attendees who participated in soil examination and footwear. A Forensic Geology session was held at the Northern Ireland Science Festival. This event was organized by the Ulster Museum in collaboration with Queen's University Belfast (QUB), Geological Survey Northern Ireland (GSNI) and Northern Ireland Regional Group of the Geological Society of London. This included an overview of, 'What is forensic geology and how has geology helped to solve

several high profile criminal cases in the UK and internationally?’ For example, how can soil found on a shoe help to solve a crime? A series of interactive talks was followed by a treasure hunt for forensic geological clues using exhibition material in the Ulster Museum which revealed how a forensic geologist thinks and shows how an understanding of geological processes and environments can assist the crime.

The 4th ENFSI annual meeting was held on 16th to 17th April 2015, at Riga, in Latvia. There were three sessions on; ‘Wildlife and Non-human DNA’, ‘Soil’ and ‘Morphology’. In addition, there was a ‘bring an unsolved case to the group’ session, which was useful for sharing innovative ideas and best practice. On 22nd to 27th May 2015, a training course was delivered in Namur, Belgium, organized from the ENFSI (ASPT). The course included pollen and mineral extraction and characterization, focusing on microscopy when samples are very small.

On 10th to 11th June 2015 the University of Messina, Sicily, hosted a new MSc course in Forensic Geology, the first of its kind in Italy. The Master is a specialization, higher educational, cyclical, and long-term course with a final First Level Postgraduate Master degree, after a final exam. The course aimed to provide the delegates with training in Forensic Geology and a basic knowledge in the legal field and a focus on technical and scientific matters, to deal with professional reports and consultations, as ex officio technical consultant in civil trials, or as expert witness/consultant in criminal trials, having a certified training and education. On 25th and 27th June, 2015 the Italian Geological Society held its National Congress, where a session on forensic geology was organized by the Section Young Geologists in cooperation with Rosa Maria Di Maggio. The event Forensic geology: application had the aim to show to young freelance geologists the application of earth science to forensic and the related professional perspectives.

On 16th - 17th September 2015, the University of Messina, the Master course in Forensic Geology and the Master course on Forensic Science, with the patronage of RIS of Messina, the IUGS International Union of Geological Sciences – Initiative on Forensic Geology (IFG), the National Council of Chartered Geologist and the Register of Chartered Attorney of Messina organised the two events ‘Days on Forensic Geology’ and ‘Field training on forensic geology for the search of buried objects in test sites’. The course was attended by approximately 50 delegates, and the content included a series of formal lectures followed by knowledge transfer events and field training.

The 4th European Meeting on Forensic Archaeology (EMFA) was held on 28th to 29th August, 2015 in Pontoise, France. EMFA is a joint venture between the Forensic Science Institute of the French Gendarmerie (IRCGN) and the ENFSI project group Forensic Archaeology. The theme of the conference was, ‘Crime Scene: Role of the Forensic Archaeologist in a Multidisciplinary Team’ and has a stated purpose of discussing the present and future state of forensic archaeology in European countries, to present relevant case studies, research and development. The 7th European Academy of Forensic Science Conference, Prague, Czech Republic, was held on 6th to 11th September 2015. The conference focused on, ‘Pushing Boundaries and Working Beyond Borders.’

Laurance Donnelly and Martin Grime (former British Police and FBI canine trainer), with support from Alastair Ruffell and Mark Harrison explored research opportunities aimed at better understanding the deployment of victim detector dogs to locate homicide graves and to explain the generation of false-positive indications.

In June 2016 the Institute of Geologists of Ireland and Geological Survey of Ireland organized a seminar on the Applications of Geology to Crime, Police and Law Enforcement

where Alastair Ruffell, Lorna Dawson, Duncan Pirrie, Jennifer McKinley, and Laurance Donnelly, held a knowledge transfer session with the Irish Police (Garda).

In Russia, forensic soil science is used regularly in casework. The Russian Federal Centre of Forensic Science (RFCFS) of the Ministry of Justice of the Russian Federation, - Moscow, has a laboratory dedicated to examination of soil and biological trace forensics , with Olga Gradusova – the head of the laboratory, and Ekaterina Nesterina – leading state soil forensic expert (235, 236 and 237). They carry out around 20-30 soil forensic examinations annually, with almost all cases of a criminal nature.

7. Latin America

In Argentina and Brazil several meetings were held (e.g. 238), driven by Carlos Molina. Scientific papers were produced (126, 143, 244) and interviews were given (e.g. to the Discovery Planet (September 12th, 2013)) and popular press articles were published (e.g. 238, 239, 240, 241, 242, 243).

8. United States

The Scientific Working Group for Geological Materials (SWGGE0) chaired by Bill Schneck, was started in 2011. In 2014, the Scientific Working Group for Geological Materials (SWGGE0) was dissolved, along with most of the other scientific working groups (SWGs) in the United States. A new organization was established under the National Institute of Standards and Technology (NIST) called the Organization of Scientific Area Committees (OSAC). This new organization largely replaced the various forensic science SWGs with a single organization. The result is that the many autonomous groups that used to operate independently of each other, and with very different processes and standards, now operate under a single umbrella organization with much greater consistency. One of subcommittees in the OSAC is the Geological Materials Subcommittee, and this group is continuing much of the work that was started by SWGGE0. The new subcommittee has met in person on two occasions, in January 2015 and January 2016. Additional meetings are planned for late 2016 and April 2017.

In its first year, the subcommittee created a draft guideline on collection of soil in the field (for use at crime scenes, for example). There are two workshops scheduled to take place late in 2016 to test the fitness for purpose of this draft document. Based on feedback from the workshops, the subcommittee intends to modify the guideline and then submit it to ASTM International, a standards development organization. The subcommittee is currently in the early stages of preparing new standards and guidelines related to forensic soil examination in the laboratory. Information about the OSAC, the Geological Materials Subcommittee can be found on NIST's website (<http://www.nist.gov/forensics/osac/sub-geo.cfm>).

The American Academy of Forensic Sciences (AAFS) hosted a two-day workshop 'What Did You Just Step In?! Use Forensic Soil Examinations to Find Out' and the Geological Society of America (GSA) Annual Meeting had a 'Progress in Forensic Geochemistry' session with 12 presentations. The GAC-MAC-AGC-AMC Joint Annual Meeting held a Forensic Geology session with 7 presentations.

10. Activities in other countries

'Evidence from the Earth' the seminal book by Ray Murray was translated into Chinese by the Ministry of State Security, PRC (245). Murray was then invited to China to present on the subject of the forensic geosciences. It was then arranged for GuoHongling, a forensic

geologist with the Institute for Forensic Science, China, to make a presentation at the IUGS-IFG meeting in Cape Town and become a member of this international body.

In addition, forensic geoscience is still active in Japan, largely through Ritsuko Sugita, (1), who held a session on forensic geoscience on 11th to 13th September 2015 in Japan, and with interest now growing in the middle East (Table 1, 15th to 18th November 2015).

11. The Future

As mentioned in the introduction, the development of forensic geology is advancing very rapidly, and it is likely that this situation will continue with great examples of the effectiveness of the approaches developed in this field for intelligence, investigation and as evidence in court. It is pleasing to see so many organizations with an international cooperation focus, and a drive to improve the quality standards globally.

However, geological setting, soil type, availability of equipment and/or availability and resolution of the available databases in each nation are different, and therefore optimization of developed techniques and the application of that knowledge depend on that individual national context. Increasingly collaboration between nations is vitally important, as crimes do not recognize national boundaries. Forensic geoscientists, working with partners, such as the wider forensic disciplines, legal practitioners, police officers and wider research groupings, along with continued international cooperation should help deliver safer outcomes in the fight for justice.

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Criminalistics

Gunshot residue, 2013-2016

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1. Introduction

This review paper covers advances in scientific methods applied to Gunshot Residues reported since the 16th Interpol Forensic Science Symposium in October 2013.

A literature search was conducted covering articles published in the main forensic journals in 2013, 2014 and 2015.

During discharge of a firearm, primer and gunpowder residues as well as metal particles from the projectile and the cartridge case are expelled from the muzzle and from other openings of the firearm. These residues are referred as primer residues, firearm discharge residues or gunshot residues (GSR). During the last three years, a review has been published on this topic by Chang et al [1]. The basic principles and fundamentals are discussed in this article: ammunition, GSR, evidential value, inorganic and organic detection, estimation of the firing distance, bullet hole identification, estimation of the time after discharge and determination of ammunition type.

Scanning electron microscopy coupled to energy dispersive X-ray microanalysis (SEM-EDX) still is the method of choice for the identification of inorganic GSR on samples. This technique is well suited for the detection of small particles (down to 0.5 μm) containing heavy metals such as lead (Pb), barium (Ba) and antimony (Sb) originating from primers with a classic composition (e.g. sinoxid primers). Moreover, it allows for the determination of the correlation between the morphology and the chemical composition of individual particles, composed of Pb, Ba and Sb, considered as characteristic of GSR. However techniques such as GF-AAS, ICP-OES, ICP-AAS and ICP-MS are still used in some forensic laboratories, because of their high sensitivity, their speed and their ease of use, despite the fact that morphological information of the particles is absent. Vanini et al. published two articles about the use of ICP-OES to detect GSR on samples taken from hands [2, 3]. They showed that the concentration of the elements is correlated to the number of shots, and in some favourable circumstances, it should be possible to predict the number of shots with a good accuracy.

2. Inorganic GSR

2.1 Fundamentals of GSR formation

Using particle-induced X-ray emission, Duarte et al. examined the heavy metal elemental ratios found in primers and compared these ratios to the ratios obtained from large particles (50 to 150 μm) recovered on targets [4]. These authors showed that these ratios do not correlate with each other, especially for Ba/Pb. This study confirms results in a previous

publication [5]: GSR particles found in cartridge cases do not always correlate with GSR particles coming from other samples (hands, target).

Melo et al. characterized submicron GSR particles by transmission electron microscopy [6]. For the brand examined in this study (CBC ammunition), they showed that the majority of the particles produced consist of crystallite agglomerates. Moreover, the diffraction patterns indicate the presence of metals and metallic oxides of lead and antimony. Following these observations, the authors assumed that a cloud of metallic atoms is immediately generated after the shot; these atoms condensate into metals and metallic oxide crystals, which then agglomerate around each other. Based on these observations and according to the authors, crystallinity seems to be a characteristic feature for most of the nanoparticles, and, as a consequence, GSR particles are not entirely amorphous, as claimed before.

2.2. Non-GSR sources of GSR-like particles

Since the beginning of GSR-expertise, concern has been expressed about GSR-like particles originating from a non-ballistic origin, which could lead to false-positive interpretation of the results at the source level; these particles are similar to GSR but do not originate from the use of primers. Numerous publications have already described particles produced by environmental and industrial sources. Especially, many researches have focused until now on potential sources of Pb, Ba and Sb from the environment. However, with the introduction of ammunition containing metal free primers, research nowadays has to be conducted to determine which are the potential sources of particles having the same composition as the new types of primers and presenting a typical GSR-morphology. Concerning this topic, Brozek-Mucha examined particles coming from welding fumes [7], for which one can expect a typical GSR-morphology. These particles indeed look similar to GSR particles and according to the author, the distinction is not trivial and can only be done by trained analysts. However, these particles are registered with a great number of particles of iron and iron oxides; taking these particles into account can prevent false positives in real cases.

As written before, GF-AAS is still used in some forensic laboratories, even if this technique does not offer any morphological nor any chemical information of individual particles; this leads to a greater risk of false positives. Therefore it is crucial to monitor levels of Pb, Ba and Sb in different environments, in order to set a threshold value of environmental contamination. In this way, and because there was a need in some real cases, Aksoy et al. [8] examined the level of Sb in different fabrics used to cover vehicle seats. They showed that polyester covers are a potential source of Sb. The presence of this element, uniformly distributed in the fibers, is due to the use of Sb compounds as catalysts during the production process. The authors conclude that in cases for which polyester material is used to cover the seats, GF-AAS should not be used to detect the presence of GSR in vehicles.

2.3. Interpretation of analysis results and the application of Bayesian principles

Applying the well-known case-by-case approach previously suggested by Romolo and Margot previously [9] in her daily work, Brozek-Mucha published a review [10] of selected problems dealing with the chemical and morphological properties of populations of GSR as a function of different factors, such as the type of ammunition, the distance from the muzzle to the target, the type of substrate the particles sediment on and the time between shooting and collection. Using this approach, the author demonstrated the possibility to get additional information on the modus operandi, by analysing the chemical and morphological properties of the particles of interest in details, and by performing reference shootings. This leads to an accurate characterization of the type of particles produced during the shooting incident.

In 2009 and 2011, Biedermann et al. proposed the implementation of the evaluative approaches and the Bayesian networks in the GSR field [11, 12]. The networks were based on statistical models developed by Cardinetti et al. previously [13]. They demonstrated how Bayesian networks may be used to model numerous factors involved in the calculation of the likelihood ratio. Based on own experimental GSR persistence data, Gauriot et al. developed a more complex Bayesian network [14]. According to them, this network was able to take into account more accurately the different scenarios and conditions related to a shooting incident. However, using two examples from real cases, they showed that even by applying this approach, the likelihood ratio strongly depends on various factors, such as the level of background of contamination. Since these factors can be assessed quite differently depending on the expert's personal opinion, the authors conclude that serious caution should be exercised when statistical approach is used, in particular Bayesian approach, is used for the interpretation of GSR results. The opportunity to comment this article was taken by Gallidabino et al. [15]. According to them, the weakness of the interpretation is due to the nature of the evidence (i.e. GSR) and not to the models proposed. These models only point out the complexity of interpretation in forensic science, and particularly in GSR.

Applying an evaluative approach in their routine casework, Hannigan et al. examined the background levels of GSR on clothing from arrested suspects who were not related to firearms cases [16]. These evidences are optimally suited for background level estimation since these suspects, unlike other people, follow the same path than suspects related to firearms (arrest, body search, transfer to police facilities etc...). Out of 100 garments analysed, 98 did not show any GSR particle; one garment had 1 GSR particle, another one had 2. These results give an excellent indication on the contamination risk of arrested suspects, and can be used as valuable data to calculate likelihood ratios. The frequency of occurrence of various cartridge case compositions from real cases was also reported in this article.

As for other trace particulates, secondary transfer of GSR is an important issue, since an innocent person could falsely be accused to have been present in a shooting environment, or even to have been the shooter because of the presence of GSR on him. French et al. [17] studied three scenarios: sampling a shooter, sampling an individual having shaken the hands of a shooter, and sampling an individual having handled a pistol that had recently been used. However, one should note that in this study, no (or very short) delays were observed between the shots, the actions and the sampling; this is not like in real cases. If the first scenario led to the detection of the highest quantity of GSR, the two other scenarios showed that relatively large numbers of GSR particles were transferred during the secondary transfer process. Moreover, no significant differences were observed in terms of size: secondary transfers of small and large particles were observed in both scenarios. This study confirms the risk of misinterpretation of results, and possibly misidentification of the shooter. In a second paper [18], French and Morgan examined two additional scenarios: sampling an individual at the end of a chain of two handshakes with a shooter, exploring the potential for GSR to undergo tertiary transfer; and sampling the bystander of a shooter. Significant GSR particles have also been recovered for both scenarios. Moreover, the efficiency of the transfer was found to be as high for the third transfer (40-50%), as for the second one (5-10%). This study highlights the need to prevent unwanted transfer during collection phase; some recommendations are given to prevent this risk.

Brozek-Mucha [19] examined the prevalence of GSR in selected populations. For 100 people who were not related to firearms, only one GSR particle was identified on one individual. For people coming into contact with firearms, the number of particles identified strongly correlates with the time interval between last shooting and sampling. And not surprisingly, frequent shooters (on a daily basis) are highly contaminated. Similar to studies

conducted by French et al., Brozek-Mucha also examined different scenarios involving primary and secondary transfer of GSR. A particular case of secondary transfer was also examined, i.e. the potential contamination of hunter's relatives. For this population, living in the same environment as a shooter, this situation does not represent a great source of contamination. In her conclusion, the author claims that the number of GSR particles found on a suspect should always be put in relation to the circumstances/context of the case.

Charles et al. examined the collection efficiency of GSR as a function of the type of fabric (cotton, leather and wool) [20]. They showed that the sheddability of the fabrics plays a crucial role in the collection efficiency of GSR, since the efficiency is about five-fold higher for leather (low sheddability) compared to wool (high sheddability); the main reason explaining this difference being the rate of saturation of the stubs.

2.4. Quality

In the domain of GSR analysis, the reference norm is the ASTM 1588 which was recently revised (March 2016) [21]. Compared to the previous version, the novelty is the introduction of the definition of major, minor and trace elements in particles of interest. Beside this norm, two guides exist: the ENFSI guide (more or less the same as the ASTM norm) [22] and the SWGGSR guide (more detailed, in terms of interpretation) [23].

Proficiency tests are conducted every year. They are organised by commercial provider QuoData (Germany) in collaboration with the ENFSI Expert Working Group "Firearms and GSR", and consist in the detection by SEM-EDX of 150 to 200 three-element particles (lead, barium and antimony) distributed in 4 size classes (0.5 to 2.5 μm). Three proficiency tests were conducted during the period of interest (GSR2013, GSR2014 and GSR2015).

White from the Texas DPS Crime Laboratory Service reported a specific aspect of Quality Insurance, i.e. negative and positive controls [24]. Because his laboratory uses three different SEM/EDX systems, he used the youngest to adjust the different parameters of these systems, in order to obtain the same analytical performances. For these positive controls, one can use the ENFSI proficiency tests described earlier. However these standards are quite expensive and according to the author, can suffer from beam degradation over time. Therefore he prefers to use his own in-house positive samples, obtained from shooter's hand.

However, because of potential cross-contamination, reservations can be expressed to introduce positive controls into the laboratory. Therefore Hearn et al. introduced ytterbium-tagged positive controls [25]. Tagging positive controls can indeed monitor potential cross-contamination of pieces of evidence. The method of tagging consisted in spiking sinoxid based primers (containing Pb, Ba and Sb) with ytterbium prior to discharge. With this procedure, the majority of the PbBaSb particles produced were successfully tagged with the marker. Ytterbium was chosen because its oxide should have little effect on the combustion properties of the primer. Moreover, none of its X-ray peaks overlap with any of the elements Pb, Ba or Sb.

Following the acquisition of a new field-emission SEM/EDX, Izraeli et al. optimized several parameters of the device in order to detect a maximum of particles within a minimum analysis time [26]. The samples used for the optimization process were taken from real cases. The ENFSI proficiency test was also used at the end of the validation. The parameters optimized were the accelerating voltage, the backscattered electron emission acquisition time, the backscattered electron image threshold, the magnification and the number of pixels per backscattered electron image. According to the authors, 20 kV is the

best accelerating voltage. They suggest to use the highest magnification possible together with the lowest image resolution possible.

Using a multivariate optimisation, Vanini et al. established the best operating conditions for detecting Pb, Ba and Sb by ICP-OES [27]. Three variables were studied: radio frequency power, nebuliser gas flow and aspiration rate. They showed that nebuliser gas flow was the most important parameter in the optimization of the signal intensities.

3. Instrumentation and methods

A number of new instrumental methods were introduced, which consisted mostly of adaptations of existing element analysis.

In [28] Goudsmits et al. present a comprehensive review of the recent trends in the analysis of organic gunshot residue. They cover inorganic as well as organic GSR analysis, sampling techniques, GSR markers and give a short overview of more recent developments using techniques such as Raman/FT-IR and electrochemical analysis.

In [29] Cid et al. report on a new technique they have developed to improve the limit of detection for tin in aqueous solutions with Flame Furnace Atomic Absorption Spectrometry. They describe the use of a subcritical fluid nebulizer that operates with liquid CO₂ to pre-concentrate the tin-ADPC complex in solutions, which yields a 240-fold increase in sensitivity and 325-fold improvement in detection limits. Such marked improvements can be well used in the detection of tin on hand samples from shooters, where tin, lead, barium and antimony are still used as indicators for the presence of GSR in cases where the analysts have only access to AAS techniques for their investigation. The authors illustrate the application of their method by using test shots with four types of 9mm ammunition.

3.1 Use of Raman Spectroscopic techniques in GSR analysis

There is a marked increase of publications in which Raman spectroscopic techniques are used, sometimes in combination with other instrumental methods.

In [30] Abrego et al. introduce their method combining Scanning Laser Ablation-ICPMS and Raman Spectroscopy for the detection and characterization of GSR particles from lead-free ammunitions. They describe their development of an adapted sampling stub, based on the commercially available carbon adhesive-covered SEM stubs, which they have covered partly by a polytetrafluoroethylene (PTFE) layer. This PTFE layer is used as a background for the Raman spectroscopy of organic GSR particles, while the adhesive layer is used for the search for inorganic particles by the SLA-ICPMS system. Using this very sensitive and quantitative elemental analysis system, they can automatically scan and characterize the composition of the inorganic GSR particles based on 20 elements commonly found in lead-free GSR and compare this to cartridge information in a case-by-case approach (since the inorganic GSR particles expelled by lead-free ammunition are ill-defined). The search for N-DPA and other known organic GSR particles is, on the other hand, still based on a manual search. In their future developments the authors will work to further automate the Raman search process, yielding instrument throughput times under two hours per stub.

In [31] Bueno and Lednev describe how they use data from complementary Raman and FT-IR analysis to improve the statistical discrimination of GSR. Using particles from .38 and 9mm ammunition they acquired both FT-IR and Raman spectra. Both types of spectra were shifted a small amount, indicating that they probe different vibrational modes of the molecules. By using statistical data analysis on combined spectra of only 46 particles, the authors show that it is possible to discriminate GSR particles as coming from one or the other firearm/ammunition combination. This information could be used by investigators to

assign or rule out a weapon/munition combination to have discharged a crime scene GSR sample.

Lopez-Lopez et al report in [32] on their study of a memory effect when using two types of ammunition (one containing DPA and the other ethyl centralite) through the study of the Raman spectra of the macroscopic particles. They do remark that the intra-batch variability (that is the variation between the spectra of particles within one batch) can be rather large. Moreover, the variability of the memory effect could vary depending on the sampling location. Their findings should therefore be limited to the analysis of macroscopic particles found on tan-coloured targets, where they can be easily visualized.

In [33] Zeng et al. discuss the fundamental work they have performed to elucidate the vibrational landscape of ethyl and methyl centralite visible in Raman scattering spectroscopy using Density Functional theory (DFT) simulations. They show that there is a good agreement between the theoretical predictions and the spectra they acquired on real samples using a home-built Line-scan Raman (LSRM) unit. The authors claim that their work is a step forward for the detection of GSR containing centralites using Surface Enhanced Raman Scattering (SERS) technique.

In [34] Bueno and Lednev report on their experiments to acquire automated chemical mappings of Raman data from adhesive tape samples of organic and inorganic GSR. The laser beam of the Raman microscope was scanned over the surface area of the samples, collecting Raman spectra from each point. The chemical mappings corresponding to the previously acquired GSR standards particles were used in a chemometric treatment (PLS-DA) to reveal the presence of particles which were subsequently automatically classified as organic or inorganic GSR. The authors claim that, when fully developed, their method will be able to find GSR particles and classify them without expert intervention. The method works irrespective of the composition of the GSR (classic or heavy-metal free) and detects particles larger than $3.4 \mu\text{m}$. Work is still underway to improve on the size limit for detected particles, to establish precise vibrational mode assignments of the particles and to speed up the scanning process.

3.2 Use of ATR-FT-IR imaging in GSR analysis

Also ATR attachments to FT-IR instruments have been used for the characterization of individual particles as GSR. In [35] and [36] J. Bueno and I. Lednev describe their use of Attenuated Total Reflectance(ATR) imaging and FT-IR spectroscopy in the application of finding and characterizing GSR particles on a cloth substrate. Both organic and inorganic GSR particles show a characteristic vibrational fingerprint, allowing them to be identified on a sample. Using statistical analysis methods, the authors were able to discern between GSR particles expelled by several types of ammunition. In order to determine the applicability of the method to detect both organic and inorganic GSR, the authors fired two weapons at a cloth and collected the GSR particles on adhesive tapes. Analysis of these samples shows that particles of sizes down to about $5 \mu\text{m}$ can be positively identified as GSR, based on their vibrational fingerprint. As this was a proof of concept study, the authors will expand their future experiments to include more types of firearms-ammunitions combinations in order to obtain a larger collection of vibrational fingerprint spectra and so minimize the risks of false positive assignments.

3.3 Use of Mass-spectrometric analysis techniques in GSR analysis

Mass spectrometry continues to gain momentum in the analysis of GSR, both for inorganic and organic types of particles. A lot of this effort is guided by the need to find alternative analysis procedures for the lead-free and metal-free primers which are starting to appear in general casework. In [37] Thomas et al. report on their study of unburned smokeless

powders by using UPLC/MS/MS techniques. Various powder samples of single and double base composition were analyzed to determine the relative composition of their additives. With this information it was easy to make a distinction between different brands and even production lots per brand. Further testing is necessary to validate this method for application on recovered GSR samples after firing or post blast explosives residues.

In [38] Szykowska et al. report on their work on detecting and characterizing GSR particles using Time of Flight - Secondary Ion Mass Spectrometry (ToF-SIMS). Using fired cartridge cases of shotgun shells, they were able to sample, visualize and analyze the characteristic GSR particles - even from different types of surfaces (eg wood, metal, plastic) after a secondary transfer had been performed by touching them with contaminated fingers. The particles were lifted from the secondary transfer surface using different types of adhesive tapes. Moreover, by correlating the images of Na⁺ and K⁺ ions, naturally found in the excretion of sweat, and with the elements of the characteristic particles, they could show that the GSR particles were transferred to the surfaces on the fingerprint ridges of the contaminating person. The composition of the GSR particles was finally confirmed by classic SEM/EDX analysis of particles from the cartridge cases.

The usability and advantages of Ion Beam Analysis for GSR forensics was tested by Romolo et al. in [39] and compared to the widely accepted technique of choice SEM/EDX. Because of a number of fundamental effects, Proton Induced X-ray Emission (PIXE) and Proton Induced Gamma-ray Emission (PIGE), offer a number of advantages in the analysis of ammunition types that contain no heavy elements. These advantages enable the detection and quantitative characterization of GSR particles containing only nonmetals, light metals and other typical lead-free primer elements such as Sr and Zr. Notably, authors successfully detect B in a lead-free primer with PIGE, through the use of a proton-induced nuclear reaction. Finally, the PIXE technique allows distinguishing between overlapping Ti and Ba lines, which often cause problems in correctly classifying potential GSR particles. Although the authors acknowledge the fact that the prohibitive cost of the IBA equipment make it impossible for forensic labs to invest in this technology on an individual basis, they point out that ample research facilities in Europe exist which can provide analytical services through international cooperation in selected high-impact cases.

In [40] the same research group takes the principle of IBA a step closer to the forensic practice by developing and testing a protocol to manually relocate particles on a previously analyzed SEM/EDX sample and performing multivariate statistical analysis on the PIXE data acquired from these GSR particles. The authors show that the protocol can successfully relocate the particles larger than 1 μm and quantitatively characterize them. Radiation stress tests show that the particles are not altered by the analysis so that a subsequent analysis is possible. Multivariate data analysis shows that samples of classic lead-primer ammunition of different brands can be discerned and ordered into groups. The characterization and grouping was performed on samples from both cartridge cases and shooters' hands. Furthermore, the known variability of composition between cartridge case particles and hand samples, nor possible memory effects on hand samples had a significant influence on the classification and grouping of the particles in distinct brands.

In [41] Taudte et al. present an extensive review of the work already performed in the analysis of inorganic and organic GSR using mass spectrometric techniques.

In [42] Benito et al. propose a rapid and sensitive method to sample and analyze 18 different species of organic compounds found in organic GSR by LC-QToF. In order to enable also the analysis of inorganic GSR components, a home-modified tape lifting technique was developed and compared to the classic swab sampling method. In their studies, both "lead-

free” and “heavy-metal free” ammunition types from Fiocchi and Remington were used. Their results show that, for most of the tested analytes, the modified tape lifter equipped with a partial PTFE layer outperforms the classic methanol swabs, while it still enables for the simultaneous analysis of inorganic GSR particles by SEM/EDX. The authors state therefore that their proposed method is an ideal analytical combination in case lead-free ammunition is used in a firing incident.

In [43] Tarifa and Almirall demonstrate yet another technique for the detection and characterization of both organic and inorganic GSR on the hands of a suspect. As the current method of choice for GSR detection relies on the time-consuming SEM/EDX technique, which only detects the inorganic components of GSR, a novel and fast method is proposed by the authors, which can detect both the inorganic elemental components and the organic compounds indicative for GSR from one single swab sample applied to the suspect’s hands. The proposed technique employs headspace analysis of volatile components present in the swabs by GC-MS - focusing on the N-DPA additives and powder compounds - and a Laser Induced Breakdown Spectroscopy (LIBS) analysis of the inorganic elements - specifically lead, barium and antimony. Although further work is necessary to confirm the results, the authors see advantages for this method as only one swab sample is required to quickly provide both organic and inorganic data in the search for GSR components.

In [44] Taudte et al. report on the use of Artificial Neural Network (ANN) software to automatically optimize the parameters for the quantitative analysis of 32 components they defined in organic GSR by gradient UHPLC. The method is able to analyze a mixture of components in under 27 minutes with detection limits below 0.2 ng, yielding LODs lower than previously reported organic GSR concentrations in simulations. Further work now needs to focus on the sampling protocols and treatment of samples for GSR collection of both organic and inorganic components in real-life shooting situations.

3.4 Field testing and field-deployable equipment for GSR analysis

There is a clear tendency to try to introduce GSR analysis tests already at the crime scene, for use by the police forces in tactical investigations. An example of this is the visualisation of contact of suspects with the butt or other parts of a firearm. A few years ago the PDT technique was introduced by the Israel police to visualise Fe(II) on the hand palms of shooters in the field or police station. In [45] and [46] Bar-Or et al. discuss their efforts to improve the results of the use of the PDT reagent (commercialised under the name Ferrotrace™ and Ferroprint™) to detect the presence of Fe(II). The sensitivity of the method can be enhanced by increasing the skin humidity after the PDT has been sprayed. Two methods are tested for this purpose: holding the hand in the vapor of a boiling water kettle, and covering the subject’s hand by a plastic bag to induce sweating. The first method is proven to be superior over the plastic bag. Furthermore, the authors test the effectiveness of the PDT method on the hands of children, a test that has not before been undertaken but is forensically relevant, as children in the United States often come into contact with firearms in their home and are sometimes involved in accidental or juvenile shootings. In comparison with adult test subjects, the test tends to give better results with the children, probably due to the elevated acidity of their sweat when compared with that of adults.

Another example of tests that can be used by police forces is presented by Robberts et al. in [47]. They describe the use of p-dmac as a presumptive color test for nitroglycerin. This color test may therefore be used as a simple field test regarding the nature of unburned powder pellets as originating from single or double-based powder. The authors show that the color reagent works by confirmation studies using GC-MS as an independent lab analysis technique on twenty five different brands of smokeless double-base powders.

Although presumptive color tests have been used by police forces in the field for a number of years, there is now a recent tendency to introduce more complicated equipment in the crime scene or police station environment. In [48] Hondrogiannis et al. show the application of a man-portable Laser Induced Breakdown Spectroscopy (LIBS) instrument for the analysis and discrimination of inorganic GSR on the crime scene. They show that it is possible to analyse, discriminate and produce a clustering model for three wide-spread ammunition brands based on the elemental distribution of twelve elements found in the GSR recovered from cartridge cases on the scene. Using this small dataset it is possible to attribute an unknown sample to the correct munition brand with a 66% probability. The findings of this field-applicable instrument have subsequently been confirmed using a laboratory benchtop instrument. The lab tests revealed the presence of two more elements in the mix which may be used to improve the discriminator efficiency. Although the big advantage of the portable system is that it is deployable in the field, the sampling technique may be improved to yield more constant quality spectra, an issue which will be addressed in the future research efforts of the authors.

In [49] Yeager et al. test the use of Ion Mobility Spectrometers as field-operable presumptive test instruments. Two commercial instruments - one hand-held and a benchtop unit - were tested in positive ionization mode for detection of DMT, DPA, EC and MC on swab samples acquired from the hands of shooters. Various QA aspects were taken into account in order to avoid false positive/negative results and control charts were setup for both instruments to validate the measurements. The authors conclude that IMS is indeed a viable technique for on-site testing on suspects' skin for recent firing of a firearm, but careful QA validation and pattern-based data analysis procedures need to be adopted. Finally, sample storage times and conditions will need to be strictly controlled in order for reliable control measurements to be possible in the lab afterwards.

Toal et al. from RedXDefense LLC have developed a field testing named XCAT based on a modified sodium rhodizonate test in order to screen potential shooters [50]. According to the authors, this test can then be analyzed by SEM/EDX for confirmation in case of positive results. Due to the use of sodium rhodizonate in acidic medium, the authors observe a decrease of the amount of lead in the particles. This necessitates careful review of particles of interest automatically detected by the SEM/EDX, especially the BaSb particles. This article does not mention/study the problem of false-positives (presence of lead due to a source different than a shooting incident).

3.5 Separation and Identification of gunpowder and additives using electrochemical methods

Although mass spectrometry is a modern and new instrumental technique to be introduced at the crime scene, other analytical techniques exist that can now be successfully used outside the lab environment. A branch of analytical chemistry that lends itself well to miniaturization and field-use is electrochemistry. Although in former years the techniques required the use of chemicals in a controlled (lab) environment, developments in sensor technology have made it possible for non-specialists to use the instruments outside the lab. In [51] O'Mahony and Wang present a review of the literature on the detection of both inorganic and organic GSR components. After discussing the classic techniques historically developed and used in large centralized lab facilities, they go on discussing the historic development of electrochemical analysis tools and protocols for GSR components. The authors see a large potential and a bright future for small hand-held instruments that are operated in the field, offering very rapid, specific and portable analysis capabilities at very low cost.

In [52] Bandodkar et al. discuss and demonstrate the development of a wearable electroanalytical device or 'forensic finger' which is built into a nitrile glove. The device consists of two components: a screen-printed carbon electrode, worn on the index finger cot of the glove, and a solid ionogel dot, cured onto the thumb cot of the glove. By rubbing over a contaminated surface with the index finger, the GSR particles present will transfer to the electrode of the glove. By subsequently pressing the index finger and thumb together the ionogel dot and electrode will complete the electric circuit and a Square Wave Voltammetric Scan can be performed in-situ. As the electrochemical analyzer is coupled to a notebook computer, the results can readily be displayed on the screen. The authors demonstrate this technique to work both on GSR (Pb, Sb and Cu metals) and explosives residue (reduction of DNT nitro-groups) without modification of the gloves' sensor electrode. Authors furthermore demonstrate through some limited testing of the forensic finger with regards to shelf-life of the ionogel and mechanical stress tests exerted on the electrode finger, that the system is relatively robust and operable in realistic field conditions. The usability of the forensic finger in the field is demonstrated by some sampling tests inside and outside of a firing range. The authors conclude that this technology demonstrates to be promising as a fast, portable, field-deployable screening method for rapid detection of security threats or firearms contacts. Future developments will follow in the incorporation of this technology in other wearable instruments and wireless transmission of results to smartphones and centralized databases.

Square Wave Voltammetry is a rapid and selective technique to detect metals such as Pb, Sb and Cu on a surface. However, the signals of Sb and Cu partially overlap and Ba is difficult to detect, as it strips at very negative potentials which will also hydrolyse aqueous solutions in which the reaction takes place. The use of the standard SEM/EDX technique is, however, cumbersome, time consuming, expensive and can only be performed in a central laboratory by highly-trained staff. In [53] O'Mahony et al. focus on the combination of rapid electrochemical screening on-site and a subsequent confirmation by standard SEM/EDX analysis of the particles in the laboratory, based on the same sample taken on the crime scene. This combined procedure allows for a field-deployable screening test to be performed by a CSI police operator, which is then officially confirmed by a forensic expert in the central laboratory. In order to sample the particles and perform in-situ SWV analysis, carbon electrodes were screen printed on Ag/AgCl substrate-electrodes and mounted on double-sided adhesive tape. This sampler allows for the stubbing of surfaces and the shooter's hand, directly followed by an SWV scan for elevated Pb, Sb and Cu concentrations. The sampler can then be loaded into the SEM for standard SEM/EDX particle analysis without prior modification. Tests by the authors revealed that the two methods did not interfere with the sample: SWV was still possible after the sample had been analysed with SEM/EDX and vice-versa. Finally, tests using a bare Carbon Screen-Printed Electrode without adhesive tape revealed that the adhesive tape is indeed necessary to lift and contain the particles from the sampled surface, as no SWV signal nor GSR particles were detected from the bare electrode without adhesive tape.

4. Shooting distance estimation and bullet hole characterization

4.1 General considerations

The largest part of GSR produced by a shooting is projected on the target (object or victim), if this target is close enough to the shooter. The diameter and the density of the GSR particles deposition pattern will help to determine the firing distance. This deposition pattern is usually revealed by colour tests; the most popular colour tests being the Sodium Rhodizonate test (for lead and barium) and the Modified Griess test (for nitrites).

However, other analytical strategies exist to estimate shooting distances. As further development of previous researches, ICP-MS and ICP-AES techniques have recently been

reported as valuable techniques for shooting distance estimation and bullet hole characterization. For instance Santos et al [54] tried to estimate firing distance based on a mathematical model that was obtained after correlating the firing distance to the amounts of Pb, Ba and Sb (measured with the ICP-MS technique on radial distances). The best results were obtained at radial distances of 2 to 3 cm from the bullet hole. Using this approach, it was possible to accurately estimate the firing distance between 20 and 90 cm using Pb or Ba, with an accuracy of +/- 6 cm; the results being less accurate with Sb. In another study, Turillazzi et al. used ICP-AES for the elemental analysis of GSR deposited on skin samples [55]. They logically observed an evident decrease in quantity of GSR with increasing firing distance: the concentration of GSR at 0.2 and 5 cm is much higher than at 50, 100 and 150 cm, distances for which the differences in concentration of GSR become less clear, although still detectable.

4.2. Bullet hole characterization

The rotating bullet will usually produce a wipe ring around the entrance hole. The presence or absence of a wipe ring will help to determine the nature of the bullet hole (entrance or exit).

Usually a visual examination of the morphology of the holes can already give valuable information about their nature. However, Jason and Haag warn to be cautious when in absence of a wipe ring, visually determination of the entrance and exit holes on victims clothing is performed by examining the direction of the fibres [56]. They have shown with high speed video frame series that when fabric is shored with tissue simulant, the collapsed temporary cavity forces the fabric fibres to puff outwards instead of inward, as expected with an entrance hole.

Fais et al. have analysed by means of micro-computed tomography intermediate gunshot wounds produced on human skin under different experimental conditions (fresh, covered with textile, submerged, decomposed, charred) [57]. They have detected radiopaque particles only in entrance wounds, demonstrating the possibility to perform a differential diagnosis between entrance wounds and exit wounds in all tested conditions, as the 3D mapping of the GSR distribution clearly demonstrates.

Two components (fatty acid and its ethyl ester) of gun cleaning oil survive the elevated temperatures from the gunshot, and therefore can be detected in the bullet wipe using gas chromatography, even after 5 rounds [58]. According to the authors of this study, this approach could be particularly interesting in cases where multiple guns were used (e.g. in cases for which police officers are involved and fatalities cannot be linked to the guns): to determine which gun fired which projectile, the analysis of the nature of gun cleaning oil in the bullet wipes could be performed. In order to enhance the discrimination between different oils, the authors suggest the use of tagged oil by police forces for cleaning their weapon.

4.3 Robustness tests of shooting distance estimation

It is well known that shooting distance estimations are prone to many uncontrollable factors, forcing the use of large uncertainties when reporting distances. Logically, many researchers and experts are interested in determining the extent of the effects of various factors on the robustness of the techniques used.

For instance Vinokurov et al. have shown that an up to 50% reduced gunpowder load affects the visual appearance of the targets and the chemically visualized GSR patterns [59]. However, according to the authors, this effect is not big enough to influence considerably the estimated shooting distances.

In another study, the differences in smokeless propellant grain morphology on the distance determination was examined [60]. The two tested propellant grain types were the ball/flattened ball vs. the flake type, for which the production process differs significantly. The aim of this study was to determine whether differences in propellant type would have an influence on the GSR patterns (directly observed visually, and after chemical treatment with the Modified Griess test). In a previous study by Ron Nichols [61], it was shown that flattened ball propellant travels further than flake propellant, and makes patterns up to 150 cm, whereas flake propellant only makes patterns up to 90 cm. In the present study, two calibers have been used (.45 and 9mm). Compared to the flake type, shooting with the ball/flattened ball type of propellant resulted in more propellant (or partially unburnt) particles on the target. Moreover, one can also observe a larger pattern diameter after chemical treatment. These differences were more obvious using the .45 caliber, compared to the 9mm caliber.

Nowadays, field kits are frequently used by police forces to perform screening tests of subjects or objects present on a scene of crime. However one of the question is to know if the use of these kits do influences the results of analysis performed afterwards. Concerning this issue, a study [62] showed that the use of the field kit for bullet hole testing on crime scenes does not influence the outcome of AAS analysis performed on the same bullet holes.

4.4 Quality Aspects

For the first time, a Best Practice Manual for chemographic methods was published in 2015 by ENFSI [63]. It provides a framework of procedures, quality principles, training processes and approaches to the forensic examination in the domain of shooting distance estimation. However, this manual presumes prior knowledge of the discipline, and is not a standard operating procedure. There are requirements for the personnel such as the responsibilities, the competence requirements, training and maintenance of competence. One can point out the advice to record the colour reactions by digital scan or photography, in order to avoid potential colour fading. Special care has also to be taken to avoid contamination, and to make sure there is no deficiency in the packaging of the pieces of evidence which may compromise the value of the examination. A complete bibliography is given at the end of the manual; a table of chemographic methods is also provided, specifying the reagents to be used for the different elements to be detected.

In the field of Quality Assurance, as a follow-up of a previous validation study published in 2005 [64], the validation of several other inkjet photographic papers (for the use in the Modified Griess test) was performed [65]. The three papers which performed best were: Epson Premium Glossy Photo Paper, Staples High Gloss Photo Supreme and HP Advanced Glossy Photo Paper.

4.5 Non-destructive techniques

Beside the use of colour tests, it is also possible to estimate the shooting distance by using non-chemical techniques. The development of non-destructive techniques for direct observation of GSR patterns are regularly reported. For instance alternative light sources, widely used in forensic laboratories for questioned documents, fingerprints etc..., can also be used to visualize GSR patterns. López-López and García-Ruiz reviewed over the last 10 years these non-chemical approaches for shooting distance estimation on clothing and skin [66]. Two visual methods are discussed: the video spectral comparator (VSC) and the digital infrared imaging. Other techniques such as milli-XRF, atomic spectroscopy, FTIR, scanning microscopies and computed tomography were also examined. According to the authors, there is some progress with great potential for these techniques, but in most cases they require additional studies such as determining the effect of the use of different arms and

ammunitions, ascertaining the interferences of dirt and blood, or investigating the impact of medical procedures such as debridement in skin specimens. In their opinion, Raman spectroscopy and Attenuated Total Reflectance-FTIR may also be promising techniques, but these have not yet been studied for this purpose.

Concerning bloodstain pattern analysis (not covered by the review proposed by López-López and García-Ruiz), it should be possible to distinguish between close-range shooting and longer distance shooting, based on the presence or absence of blood on the shooting hand and gun: in a recent study [67], Kunz et al. showed that from a distance of more than 40 cm, there were no blood spatters detected anymore. They also tried to link the characteristic appearance of the bloodstains on the firearm and on the hand to the shooting distance. Fine micro spatter spray combined with round and elongated droplets were found on the gun and shooting hand at contact and at 2 cm distance shots. Within this range it was still possible to identify exclamation mark and comma shaped bloodstains. According to the authors, this technique would enable the reconstruction of the position of the gun, even if in their study, the shots were all performed in the same setup position and direction.

The ability to quickly and efficiently detect possible traces of GSR on dark-colored clothing in a non-destructive manner through the use of an alternative light source has recently been reported [68]. Alternative light source is meant to provide an efficient method of gaining preliminary data to aid in the establishment of an investigation. However, according to the authors, it is not meant to replace chemical testing. The light source used is the Spex Forensics Mini-Crimescope (model: MCS 400). The wavelength of 445 nm proved to be the most successful in visualizing GSR patterns with the largest amount of contrast and the least amount of background fluorescence. In this study, the particles from a full-load shot showed neon green on a black background, and appeared to be the unburnt gunpowder particles.

X-ray techniques may also be valuable methods for direct observation of GSR patterns, as described earlier [69]. Recently, additional investigations were reported by Knijnenberg et al. who used 2D-milli-XRF as a tool for visualising elemental distributions around holes produced by lead-free ammunitions [70]. Copper, potassium and strontium plots were produced successfully, giving a clear distribution pattern up to 50 cm.

5. Time since discharge

Estimation of the time since discharge of a weapon or cartridge case is a question that regularly pops up, but is not yet addressed in routine forensic work. Steps forward are being taken however to make this process a practical possibility in future. In [71] Chang et al. describe their study of the evaporation of five organic GSR compounds from 9mm caliber cartridges which were exposed to different environmental conditions. The organic GSR components are sampled from the cartridges using SPME fibers which were subsequently extracted and analyzed using GC-FID. The authors not only study the influence of time, the storage environment, atmospheric storage conditions (wind, surroundings, sunlight, temperature) but also the place where the cartridges have fallen after discharge. Notably, they remark no difference between the amount of recovered organic GSR between cartridges that have fallen on a hard ground (such as concrete) and have bounced, and those that have fallen on a soft support (like a curtain). They conclude that the ambient temperature is the most influencing environmental factor which limits the maximum time since discharge, ranging from several weeks to half a day.

6. Doped ammunition

Since a few years the ammunition in use by the German and Dutch police has been doped with identifying elements. Using these rare earth elements, the lead-free ammunition is nonetheless easily detected and classified by both SEM/EDX as color tests. This idea has been picked up for general application and a number of research groups are developing technology to put this idea into practice for other ammunition types as well. They also develop the detection technology to ensure that the dopant particles can be easily detected – even on the crime scene. In [72] Destefani et al. propose the use of a fluorescent dopant marker which is added to the gunpowder to show the firing of a firearm on the clothing of a shooter on-site. The marker they developed contains the Eu^+ ion in an organic complex and is easily rendered visible using UV irradiation on the barrel of the gun, the target and the arm of the shooter. The authors furthermore characterize the marker complex using FTIR spectroscopy, (differential) thermal analysis (TGA/DTA) and mass spectrometry.

In [73] Weber et al. describe the extensive tests they have carried out regarding the practical use of doped ammunition in different forensic applications of in-situ GSR detection. They used standard ammunition doped with the rare-earth complexes of Eu and Tb, yielding green and red luminescent particles when illuminated with a UV lamp, as typically used by crime scene investigating officers. Several realistic experimental setups included: the hands of shooters (with and without washing of the hands), the distribution of the particles in a shooting range and car, clothing and tissue of a victim. They also investigated the influence of the dopant addition in several concentrations on the ballistic characteristics of the ammunition such as the bullet speed and the misfire failure rate. The authors conclude that the doped particles are easily visible under UV irradiation on all the tested surfaces, showing a persistence on the hands of the shooters of around nine hours. The particles remain detectable after up to sixteen hand washings. As long as dopant levels are kept low (2% maximum), no influence on the ballistic characteristics of the ammunition are observed. As a conclusion, the dopant rare-earth markers can be easily used on the crime scene to detect GSR visually and the microscopic particles are furthermore readily found using the standard particle analysis tools in the lab such as SEM/EDX and Raman spectroscopy.

7. Modeling of organic GSR deposition and persistence

The interpretation of GSR analysis results is still very much on a case-by-case approach and on source level. In order to use organic GSR and progress the interpretation of its detection into activity-related levels, a lot of study is needed to model specifically the organic GSR particles' persistence and deposition/contamination behavior. Moran and Bell report in [74] on their study of the permeation of organic GSR components through skin, as well as the evaporation of these compounds. These two processes have a profound influence on the persistence of organic GSR on the hands of a shooter and thus should be taken into consideration when taking samples from suspect's hands and in the choice of the method of analysis. As organic GSR components are not present as solid particles on the skin, they don't show the tendency of inorganic GSR to be lost because of secondary transfer processes. As most organic GSR compounds are lipophilic, they tend to remain for a long time on the skin of a shooter, enabling the detection of GSR even after several hours have passed since the shooting incident. However, due to permeation of the organic GSR through the skin barrier into the bloodstream of the shooter and the evaporation of the more volatile components, a careful study is necessary to assess the rate at which these phenomena influence the sampling efficiency and analysis results of GSR studies. The authors model the behavior of several components often found in organic GSR such as EC, 2- and 4-NDPA, DPA and DMP using Franz diffusion cells to simulate permeation of lipophilic compounds through the skin and GC/MS and LC/MS as analytical techniques for high- and low-volatile compounds respectively. The conclusions of the authors is that organic GSR have the advantage over particulate GSR of being less easily lost due to secondary transfer

and that a good choice of organic compounds and analytical techniques yields a good detection even after nearly 24 hours after an incident. Ideally, by choosing the analytes and combining GC/MS and LC/MS techniques, it may be possible to estimate the likelihood that a person has discharged a weapon within a 12-24 h timeframe. In [75] these authors continue their study of Diphenyl Amine (DPA) permeation through PDMS simulant membranes, using this time Ion Mobility Spectrometry (IMS) as a detector. They show that they are able to monitor DPA permeation in a Franz Diffusion Cell using IMS and are able to model the transdermal permeation parameters and that the majority of the DPA permeates through the skin over the course of 3 to 4 hours.

In [76] Gallidabino et al. develop a logical simulation model based on likelihood ratio values calculated for a case-specific pair of prosecutor and defense hypotheses, using realistic measurements of naphthalene peak areas in chromatograms acquired from recently fired cartridges. They show that this probabilistic model can be used to estimate the time since discharge of the cartridge. The naphthalene peaks are obtained by 'Solid Phase Micro Extraction' (SPME) of the vapor in the cartridge, which is subsequently analyzed by GC. The authors show that, even with a limited number of comparison shots (a constraint often encountered in real casework), a regression model can be proposed to use for interpolations, so that different hypotheses about the time since discharge may successfully be tested without the need for extra data acquisition and testing. In [33] the same authors discuss the development of an enhanced technique to sample the evaporation of organic powder components from cartridges in order to estimate the time since discharge. In their 'HeadSpace Sorptive Extraction' (HSSE) method, the classic SPME approach is taken a step further in that the evaporated compounds are adsorbed on a magnetic stirrer, covered with a large volume of sorbent phase (some 200 times the amount of the SPME method), which is subsequently recovered through thermal desorption and analyzed by GC/MS. A total of 55 previously identified compounds found in the cartridges of typical handgun ammunition were used in these trials. Over a period of 31 hours after firing the evolution of the evaporated target compounds was monitored by using the HSSE sampling method. This test was performed for two .45 ACP munitions by Magtech (single-base powder) and Geco (double-base powder). Aging rates were found to be a function of the tested individual compounds and also slightly influenced by the type of cartridge. Slower decrease rates of evaporation were obtained when concentration ratios of similar compounds were used as an aging indicator, yielding much more reproducible and linear relationships of concentration ratios versus time since discharge. The authors conclude that their newly developed HSSE technique shows promising results for estimation of time since discharge of handgun ammunition, but more study on the influence of real-life circumstances such as storage conditions, the weather during and after discharge and the type and caliber of munition needs to be conducted in order to validate their technique for forensic field work.

In their final contribution [77] Galidabino et al. report on testing of nine small handgun ammunition types using the described HSSE/GC/MS method. They can identify 166 different organic compounds, 141 of which were found in the GSR of every cartridge tested. Despite this fact, the quantitative compositional characteristic of each residue is so different between munition types, that in many cases a GSR type could be attributed to a specific source (cartridge). Regarding the possibility of use for estimating time since discharge, most additives to propellants are unsuitable because of poor reproducibility and slow evaporation rates during the first 24 hours after discharge. However, naphthalene and 27 other compounds show promises of being interesting analytes for forensic dating purposes. In their future work the authors will focus on development of robust statistical tools to model the aging process and further optimize their analytical technique.

8. References

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Criminalistics

Marks, 2013-2016

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1. Towards more scientific and objective examination of marks

The traditional way of shoe- or toolmark examination includes subjectivity in the process. This may lead to variation in the conclusions of different examiners. In recent years, the demand for more scientifically based, objective approaches, which lead to less variability of the outcome of an examination, has increased. In the United States this was expressed in a report of the National Academy of Sciences (NAS), published in 2009 (2), that requests an extensive assessment of the 'variability, reliability and repeatability' of methods used in forensic examinations. In other works, there is a demand for a statistical underpinning of the used methods. In addition, the report states that 'forensic science examiners need to understand the principles, practices, and contexts of scientific methodology, as well as the distinctive features of their specialty. Ideally, training should move beyond apprentice-like transmittal of practices to education based on scientifically valid principles'.

In the aftermath of the NAS report, a substantial amount of money was provided by the US National Institute of Justice (NIJ) for funding research on more objective approaches for examination of forensic evidence (3). In addition, at the beginning of 2015, the Organization of Scientific Area Committees (OSAC) was initiated by the National Institute of Standards and Technology (NIST) and the Department of Justice, 'to strengthen forensic science in the United States'. 'The organization is a collaborative body of more than 500 forensic science practitioners and other experts who represent local, state, and federal agencies; academia; and industry. NIST has established OSAC to support the development and promulgation of forensic science consensus documentary standards and guidelines, and to ensure that a sufficient scientific basis exists for each discipline.' Previously, most scientific disciplines had an own Scientific Working Group (SWG), like the SWGGUN for firearms and toolmarks examiners (4), the SWGTREAD for shoe- and tiremark examiners (5) and the SWGMAT for material analysts (including tape) (6). These provided practical guidelines but also instructions for setting up research projects and they organized proficiency tests and regular meetings. The SWGs will be discontinued (although the websites were still on air at the time of writing in June 2016) and all the documentation available for each discipline will be reviewed, extended where necessary and subsequently made available for the community. The relevant OSACs for shoe-, tire- and toolmark examiners will be the OSAC Footwear and Tire Subcommittee (7) and the OSAC Firearms and Toolmarks Subcommittee (8), which are both part of the OSAC Physics/Pattern Interpretation (9) as well as the OSAC Materials (Trace) Subcommittee (10), which is part of the OSAC Chemistry/Instrumental Analysis (9). In principle everyone can participate in one of the subcommittees and by 2016, the OSAC Firearms and Toolmarks Subcommittee had 20 members with more than 50 % forensic firearms and/or toolmarks experts as well as academic and industrial members to make sure that both, theoretical and practical aspects are taken into account. The OSAC Firearms and

Toolmarks Subcommittee also has a newsletter, a statistics support group, a conclusion task group and provides meetings online.

To ensure the high standard of forensic examination of shoe-, tire- and toolmarks in Europe, the Expert Working Group Marks (EWGM) of the European Network of Forensic Science Institutes (ENFSI) (11) frequently organizes expert meetings to exchange information on best practices, organizes proficiency tests and provides standard procedures. Unfortunately, at the time of writing (June 2016), the web presentation, seems to be discontinued and it is not known to the author, if and when the page will be online again. To render forensic examinations more objective, also several countries in Europe invest systematically into research and contribute to the scientific underpinning of forensic sciences.

There are various ways to render a toolmark examination more objective, in each step of an examination. Some of the possibilities might be more complicated to realize and may require a substantial amount of financial investment for research or new data acquisition hardware, but some are relatively easy and straightforward to realize and this review contains many articles that describe possibilities to do so. In a recent review by Vorburger *et al.* (12) from the NIST, many aspects of an objective examination of striated and impression marks are described. The article mainly focuses on firearm marks, but the general principle also holds for shoe- and toolmarks.

The following examples are specific for toolmark examination, but also generally hold for examination of other types of marks. The most obvious way to increase objectivity is to set up, document and follow guidelines for each step and share this information with other examiners, as was done by the SWGs and will be done by the OSACs as well as the ENFSI working groups, to standardize an examination as much as possible. Also fixed protocols for data acquisition parameters and hardware (microscope) setting, light sources, data storage etc. can help to reduce variability. Another possibility is using a jig or an automated toolmark generation device, to generate highly reproducible experimental marks.

A measure that can lead to a significant improvement of the *objectivity of the data* is to rely on true 3D depth data, instead of using conventional 2D photography data, as it has been demonstrated (e.g. by Vorburger (13)) that using true 3D data yields more accurate results, when comparing marks. Many articles in this review, particularly those concerned with the examination of toolmarks, rely on 3D data and several techniques are used and documented.

With respect to the actual comparison of toolmarks, it is very important to know the *variability* of marks of a reference set and the *individuality of mark characteristics*, to be able to judge the amount of similarity or dissimilarity of two marks under investigation. If for example a relatively rare type of toolmark has to be assessed, small case related experiments should be performed to determine the statistical properties of the mark. Besides manual mark comparison, also automated methods can be employed. These do not only have the advantage of increasing the *objectivity of the comparison*, but also that they offer the opportunity of batch processing, like e.g. searching large databases. For shoemark examination for example, many different approaches were presented during the last years, all focusing on searching for the best matching candidate in large databases of shoes. For toolmark examination, the automated methods focus more on statistically quantifying the variability and individuality of characteristics of known matching and known non-matching marks at different conditions, with the goal to decide whether unknown marks stem from the same tool or stem from different tools. An advantage of not only automated methods but using analysis software in general is the additional functionality and information it can provide, that cannot be obtained with conventional methods. For example, using software it

is possible to apply certain filters for visualization purposes, to accentuate either only very fine geometrical details in a mark or only coarse geometrical details. Also models of tools can be acquired in combination with methods to predict which mark a tool would leave in a particular substrate material.

Finally, the evaluation of the comparison results can be rendered more objective and transparent. Within an identification framework for example, as it is used in the US among others, identification and exclusion error rates should be reported alongside the conclusions (an example is given in (14)). Some countries in Europe and Australia are working towards using a Bayesian framework to evaluate the evidence and aim at presenting likelihood ratio values (LRs), for example the probability of the found results of a comparison, given the samples are of common origin vs. the samples are of different origin. In the Netherlands, at this moment several disciplines are actively working on, or already implemented, using the LR in official reports, however still accompanied by verbal conclusion ranges. Several examples for how the LR can be used in casework are given in the literature (e.g. (15; 16; 17; 18)). For a more detailed explanation and discussion of the LR approach, please refer to the article 'Examination of Firearms' by Erwin Mattijssen, in this collection of reviews.

To be able to determine meaningful statistical properties of toolmarks, databases should be available, generally the larger the better. However, the presented automated methods in this review are typically tested on self-made, limited databases. The main reason for this is that there are no large databases publicly and centrally available for testing. This however will be crucial to compare the performance of different methods. In addition, meaningful and statistically founded evaluations should always be made based on extensive databases, whether marks are examined manually or automatically. In fact, for a manual comparison the expert uses a 'mental' database of cases seen so far. Thus, the larger the database, i.e. the larger the experience, the more reliable the outcome is expected to be. For firearms and toolmark data, recently a website was launched by the NIST (19), on which everybody can share his or her own data and contribute to building up extensive databases. Hopefully a similar initiative will also be started in the future for other disciplines.

Besides all the mentioned advantages, automation may also have some drawbacks and although the traditional approaches should be adjusted by including new technologies, this has to be done in the right way (20). In addition, new techniques require more dedicated training and understanding. However, the body of available literature is increasing rapidly, as demonstrated in this review. A good contribution is a book edited by Max Houck (21), a collection of articles of the editor as well as various other authors, including an introduction to forensic science, the classification and interpretation of evidence, the documentation of the results and their presentation in court. In addition, the book includes a chapter on health and safety and on measurement uncertainty. It is part of the Academic Press 'Advanced Forensic Science Series', which has the goal to fill the gap between general introductions to forensic sciences and contributions that are highly technically specific to a particular field.

2. Structure of this review article

The review is subdivided into sections covering tiremarks, shoemarks, physical matches and striated and impression toolmarks. The latter was further split into publications about conventional and invasive toolmarks respectively, as the latter is a fairly new but very exciting branch of examination. Although it is still in its infancy, it has great potential. Progress in the past was mainly hindered by the fact that invasive traumas are available to the forensic pathologist/anthropologist and a certified toolmark examiner is often not on the premises. Hopefully this will change in the future, as there might be much more information hidden in invasive traumas that are perceived to date and thus a more close collaboration

between examiners of different disciplines might yield very promising results. At the Netherlands Forensic Institute (NFI), the section Weapons and Tools enriched its portfolio in 2005 with 'Microanalyses of Invasive Traumas' or MIT, and in close collaboration with the forensic pathologists and anthropologists of the NFI as well as SEM (scanning electron microscopy) based material analysts, has worked on many cases (roughly 20 per year) in which the forensic toolmark experts contributed a major part to the outcome of the examination.

The subsections follow the steps of toolmark examination in practice and, as the strategy of assessing evidence is similar for different disciplines, the structure of the individual sections is roughly the same. As there are many contributions describing methods for automatically comparing shoe- and toolmarks, sections focusing on either of the two are given separately. Alongside the development of automated methods, several groups integrated their approaches into software in the form of graphical user interfaces (GUIs) during the last years, which enable examiners to test the methods with their own data. These are presented in a separate section as well.

3. Tiremarks

In the relevant period of this review, only one article could be found concerning tiremarks. In the article, two databases called the 'Tire Database' and the 'Car Database' are presented (23). The 'Tire Database' contains about 15.000 tires, which can be browsed using 7 different search criteria. The results of a query are a technical description of the tire, a figure showing the tread pattern, its classification and information about the history. The 'Car Database' is available in Microsoft Office Excel and contains information about the original set of tires a car is delivered with. In addition, conversion tables to find corresponding tire sizes are given. The database consists of approximately 12.000 passenger vehicles, SUVs and light trucks, of more than 300 manufacturers. Search criteria include the wheel base, front and rear track and the tire size of the original set of tires. The authors state, that the 'Tire Database' is also included in the 'TreadMate' database (24).

4. Shoemarks

Shoemarks are frequently found at crime scenes and often provide valuable evidence for an investigation (22) as they can be the link between a suspect and a scene or between different scenes. In addition, shoeprints can give information about the course of action at a crimescene.

4.1. Detection

An article by Clutter *et al.* describes an experiment to find out, whether bloody shoeprints can be recovered from a fire crime scene, as arson is often used to remove shoeprint evidence (25). The authors place bloody shoeprints on eight types of common floor material, blue rubber tiles, grey vinyl composite tiles, plywood, stone, porcelain tiles, pressboard, wood and linoleum, and exposed the samples to heat and soot from a burning fire. For cleaning, liquid latex was sprayed on the samples and removed after drying. Photographs were taken before and after cleaning and analyzed by a shoeprint examiner, who rated the samples after cleaning as being of better overall quality, yet not on the scale of individual characteristics, in six out of eight cases. The authors remark, that the examiner could clearly see which tiles were treated and which were not and that that could have influenced the outcome of the study.

Shor and colleagues present a study on the possibility to recover shoe prints from victims (26). To this end, the authors cleaned the soles of the shoes of a person and let him walking

outside of the lab for approximately 50 meters. Subsequently, he was asked to step on the lower leg of a live volunteer. The procedure was repeated several times and the prints were removed in between with three different lifting methods: an electrostatic lifter, a black gelatin lifter and a white adhesive lifter. The black gelatin lifter turned out to be the best method for lifting dry origin shoeprints from a body, because the electrostatic lifter couldn't be applied easily and may cause electrostatic shock and the adhesive lifters causes a background reaction that might conceal the print. The authors conclude that the experiments have to be repeated with cadavers, as the skin properties of a dead body compared to a live body may have an impact on the result.

4.2. Acquisition

The vast majority of publications are based on 2D scans or photographs of soles or marks, but typically all information that is available is acquired. A new low-cost scanner that may be used to determine relevant areas of a sole already during data acquisition was presented by Needham and Sharp (27). It consists of a single piece of glass (or acrylic), strips of ultra-bright LEDs and two web cams. It is based on 'frustrated total internal reflection imaging' and in contrast to conventional scanners, it only records the regions of the sole which in fact make contact with the surface. In order to accurately measure these pressure points, the person wearing the shoe is required to 'rock backwards and forwards, to mimic the action of walking'. The authors state that with this new scanner it is possible to determine the wear patterns of soles. It is also possible to acquired white light images of the soles, for reference.

4.3. Casting and Preservation

Battiest and colleagues published an article studying the effect of various fixatives for casting impression shoemarks in sand (28). They designed an experiment in which a working boot was manipulated to include eight easily visible unique characteristics, subsequently used to make six impressions in three different types of sand (play sand, beach sand and construction sand) and fixated with five different types of fixative (no fixative, aerosol hairspray, aerosol acrylic sand and dirt hardener, aerosol 'Workable Fixatif' and one pump-action hairspray). To ensure that good boot impressions could be obtained, the sand was mixed with water. The resulting ninety impressions were cast (Evi-Paq Traxtone casting kit) and examined by the same experienced footwear examiner for presence of the unique characteristics. The experiments yielded that the most characteristics could be recovered from play and construction sand if a pump-action hairspray was used and for the beach sand, if no fixative was used. The authors note that in a real case situation, the ratio between sand and water is difficult to duplicate and that more data may be needed to confirm the results, as statistical significance could not be obtained.

4.4. Variability of shoemark characteristics

To determine the brand and class of a shoe can be important as a first step (29), as it often can be done rapidly and can be important in cases where quick action is required. Several databases exist, that can assist the examiner in this, SoleMate by Foster & Freeman (30) and SoleSearcher by the Federal Bureau of Investigation (31). A forensic examination can typically only provide strong support in a case however, if it is based on individual characteristics.

4.4.1. Wear

Several publications were encountered, specifically focusing on the variability of marks depending on wear and of real crime scene prints. The first article by Sheets *et al.* (32) presents a study for which the authors took 11 pairs of Nike athletic shoes and created cut-marks, 1–3 mm deep, into the outsoles. During a seven-week period, the authors monitored the soles to check the potential loss of the cut-marks and appearance of new accidental characteristics. To this end, an automated feature detector was implemented that measures

the change in the structure of the sole quantitatively. The net rate of wear was determined to be 0.1 %, mainly in the heel and ball area. The authors conclude that 'accidental characteristics can reasonably be expected to persist over time', but also that 'it appears that the cut depths we used in creating the artificial characteristics were probably unrealistically deep, most accidental characteristics are probably shallower than this, and might not last as long'. An unexpected outcome of the experiment was the apparent lack of new accidental characteristics.

4.4.2. Substrate materials

A similar approach was presented by Richetelli (33), who studied the difference between high resolution scans of shoesoles and simulated crime-scene-like prints. To this end, 5 shoemark examiners were given 10 pairs of athletic shoes each and asked to walk 4 steps on clear acetate sheets (200 crime-scene-like quality prints). After lifting, the marks were scanned with high resolution. To assess the presence of randomly acquired characteristics (RACs) in the original high resolution scans and the scans of the crime-scene-like print, a method was developed to automatically detect circles, lines and curves, triangles and irregular shapes. These shapes were at a microscopic scale. The results show that 33 – 100 % (85 % in average) of the random characteristics were not present in the crime-scene-like prints. The loss of information can be drastic, but as demonstrated by Stone in 2006 in his theoretical statistical assessment of the presence of random characteristics on shoesoles, also relatively small numbers of random characteristics can have relatively high evidential strength (34).

The variability of RACs in seven types of sandy soils that are often encountered in Florida, USA, was presented by Snyder (35). The authors collected/purchased Astatula fine sand, Immokalee sand, Cassia fine sand, yellow builders sand, fill dirt, crushed coquina and top soil and created one impression mark in each soil type with two different athletic shoes. On the soles, 18 and 19 (microscopic) RACs were selected beforehand and checked for presence in dental stone casts of the impression marks. It appeared that the casts made from fill dirt and the yellow builders sand showed the most RACs (71.6 % and 77.0 % remaining) and the Astatula fine sand and the crushed coquina the least RACs (16.2 % and 13.5 %). The main focus of the article by Battiest *et al.* is on the effect of using fixatives before casting (please refer to section 4.3 for details), but the preserved RACs in untreated sand give an indication of their variability in play, beach and construction sand (60.4 %, 91.7 % and 18.8 %).

4.5. Individuality of shoemark characteristics

4.5.1. Automated analysis

A substantial part of literature during the last three years was focusing on automated approaches for shoemark examination and in particular, the retrieval of best matching marks from a database. Approaches were using global features of a sole, to classify whole shoemarks (36; 37; 33) and local features, to classify whole and partial shoemarks (38; 39). Besides testing their method using marks of similar quality, some authors also studied the effect of wear (39) and real crime scene circumstances like marks in blood and dust, gel lifters, digital and chemical enhancement methods and the substrates ceramic, vinyl and paper (33) on their performance. By now there are many different approaches published for automated shoemark classification and most authors tested their algorithms with their own database, which makes it difficult to compare the methods. Therefore, Almaadeed *et al.* (38) tested their and three more algorithms with the same two databases and Luostarinen *et al.* (40) implemented seven published methods and compared their performance with the same database (created by the authors), including real crime scene marks and partial marks. They conclude that while most methods work well with whole marks of equal quality, only few also

perform well for (partial) crime scene marks with varying quality. Finally, Almaadeed *et al.* (38) presents a prototype of a graphical user interface for their method.

In the following, more details are given on the specific methods. Two approaches that mainly focus on the retrieval of shoeprints from a database using global features are presented in Min and Qi (36) and Wei and Gwo (37). The first article uses a line and circle detector in the frequency domain, for the front and the hind part of a shoeprint separately. The authors mainly focus on lines and circles in the pattern in general. The method is tested against the performance of a human examiner, using an author generated database of 73 marks. The second article describes a database search algorithm based on binarized photography data of shoeprints. First the outer contours and based on that a core point of a shoeprint are determined. Subsequently the print is subdivided in circular regions and the Zernike moments serve as features for the automated retrieval of potential matches in the database. The approach is tested using an author generated database with 5 prints each of 246 shoes, hence in total 1230 shoeprints. In her MSc thesis, Richetelli (33) presents an automated shoemark comparison method based on Phase Only Correlation in the frequency domain, earlier presented by Cervelli *et al.* (41). The main focus of the thesis was to study the reduction in the number of RACs for crime scene prints (see section 4.4.2) and the performance of the automated shoemark comparison, in case of degradation of shoeprints as a result of blood and dust prints on the substrates ceramic, vinyl and paper. Experiments were conducted by 6 analysts who each made 3 blood and 3 dust prints on the substrates. In addition, gel lifters as well as digital and chemical enhancement methods were examined. The results show that in most scenarios the performance of the automated classification is getting significantly worse. Based on these results the author calls attention to the necessity of using real crime scene data to get a realistic idea of how well automated shoeprint classification methods work in practice.

A method that is based on local features is presented by Almaadeed *et al.* (38). The article describes a system for automated comparison of possibly partial shoeprints with a database. Their approach uses a multiscale Harris and Hessian corner and blob detector and the scale invariant feature transform (SIFT) descriptor with a RANSAC feature vector matching, to render the comparison robust with respect to rotation, scaling as well as moderate shear, noise levels and wear. They test their method with hundreds of prints from the SoleMate database (30) as well as the LSF database (42), which includes marks showing 'wear, tear and other real world degradations'. Using the latter, the probability of returning the correct match within the first 15 was just above 80 % and 75 % for full and partial marks respectively. In addition, they compare their approach with previously published methods, applied to the same databases, and conclude that their method performs best. The authors also present a graphical user interface to study shoeprints, select regions of interest and automatically compare the prints with the algorithm they developed.

In his PhD thesis, Jones (39) focuses on automatically comparing shoeprints, in case of degradation by wear. The author compares two different approaches, based on SIFT descriptors in the spatial domain and on a ridge detection algorithm in the frequency domain. To test the approaches, he builds up a database of 2 types of running shoes, by repeatedly acquiring their prints over a period of 5 months in which they are used frequently. The SIFT based approach seems to be not robust enough with respect to varying quality of the captured images and the pattern variation over time, while the ridge detection algorithm seems to work better for this. In the thesis, also a conceptual framework for integrating the automated method into casework is described, by developing a shoe wear model for a specific shoe or an individual person. With sufficient data available, shoe wear at any point in time can be estimated and LR values be calculated.

Luostarinen and Lehmuusola (40) compared seven different automatic classification algorithms with global and local support, including matching based on the Power Spectral Density (PSD), Hu's moment invariants, Mahalanobis maps, Gabor transform, local interest points with RANSAC, spectral correspondence of local interest points and Fourier-Mellin transform (FMT). Three image sets with different quality impressions, including 499 pairs of high resolution scans ('the good'), photographs instead of scans of 367 prints in 'the good' set at different times ('the bad') and roughly 20 real crime scene marks, partial marks and rotated prints ('the ugly'). The authors observed that the algorithms based on FMT and local interest points with RANSAC worked particularly well for the 'good' and the 'bad' set and reasonably well for the 'ugly' set. In case of missing parts of the prints however, the FMT, being a global method, did not perform well and the strength of relying on local features became more evident. Altogether however, the authors conclude that low-quality shoeprints are still a challenge for existing methods and that further improvements are required to tackle those difficult cases.

4.6. Conclusion ranges

To study the variability in the conclusions of shoemark examiners, Raymond and Sheldon set up two rounds of shoemark comparison exercises, each including 6 comparisons, which were sent out to shoemark examiners in Australia and New Zealand. In the first round, the examiners were asked to conclude using the current range of conclusions in their jurisdiction. In the second round, the examiners were asked to conclude according to the range of conclusions suggested by the SWGTREAD in the United States (5). A total of 11 and 17 responses respectively were received and the authors state that the conclusions in the second round were more clear and that the variability decreased. They thus suggest introducing the range of conclusions suggested by the SWGTREAD in Australia and New Zealand.

4.7. Weighing the evidence

The knowledge of the frequency of occurrence of a type of shoe in a region can be useful information for a court, to estimate the weight that should be assigned to the result of a shoemark examination. This is particularly relevant in cases, in which the examination result is only based on class characteristics. Benedict *et al.* (43) collected 1,511 shoeprints from students in New Zealand and 500 shoeprints from students in Australia, and determined the 'pattern group' of each shoe (more general than class characteristics, as 'the exact number and placement of basic elements may differ between different moulds'). The aim of the study was to determine the geographical and temporal variation of shoeprint class characteristics. The results show that irrespective of time and location, a large variation of different patterns exists. Even the most common patterns (Converse All Stars and Vans Canvas Era) only comprised a small fraction of the total and many patterns occurred only once. Note that in case an LR was provided to the court as the result of the forensic examination, the frequency of occurrence of a type of shoe can be used to weigh the examination result *quantitatively*.

5. Physical Match

In the relevant period, only one article was found including a physical match. Finkelstein *et al.* (44) describe a case in which they could show that a metallic chip of dimensions 14.3 mm × 1.3 mm × 0.9 mm, found on a bolt cutter of a suspect, originated from the cut shackle padlock at the scene. The match was established by comparing the chip's microscopic edge and the fracture line of the padlock's shackle. As otherwise only class characteristics were encountered, the physical match of the metallic chip was crucial for the case.

6. Striated and impression toolmarks

6.1. Detection

For restoring obliterated marks on metal surfaces with chemical etching techniques, it is preferable to keep the affected layer as thin as possible. Therefore, Song (45) compared different chemical etching reagents, made by different combinations of nitric acid and hydrofluoric acid, with glacial acetic acid or acetone as a solvent, to recover engraved marks from motorcycle and car frames. To this end, the author took two metal plates from two vehicles, stamped numbers and simulated removal of the engraved numbers by filing and manual grinding. Subsequently, three etching reagents were tested on the samples and it turned out, that the combination 10 ml nitric acid (65 – 68 %), 0.5 ml hydrofluoric acid (40 %) and 10 ml acetone yielded the best qualitative results.

6.2. Occurrence

Two articles study the presence of marks that might be useful for casework on vehicle keys and label maker cut marks. Elek (46) published an introduction to the forensic mark examination of vehicle keys. The article discusses manufacturing techniques, key 'shabbiness' by usage and mark creation by duplication of the key. Besides conventional 2D light microscopy, scanning electron microscopy (SEM) is suggested as an alternative. Weber (47) presented a case study with a label maker, which included a cutter to separate consecutive labels. Two suspect labels were examined for characteristics that can be used for identification and it turned out that the cutter leaves striated marks on the edge of the labels, which are about 100 μm thick. The author concludes that based on the striated marks, as well as class characteristics like label color, width and thickness, the labels in question could be identified as created with the label maker.

6.3. Acquisition

Conventional optical 2D and stereo microscopy are frequently used in research and case studies (45; 46; 48; 49; 50; 51; 52; 53) but in general, small microscopic marks might be difficult to see because of the limited depth of field at high resolutions. In the last years, many authors chose for imaging modalities that provide a better depth of field at large magnifications like SEM (46; 54) and digital microscopy (47) or the true 3D measurement techniques focus variation (55; 56; 57; 58; 59; 60; 61; 62; 63), confocal microscopy (64; 65; 66; 67; 68), structured light imaging (69) and photometric stereo (70). Scanning electron microscopy was discussed for 3D surface acquisition as well by Tafti *et al.* (71). Though, the sample size for 3D SEM imaging is typically under a millimeter (72). Stylus instruments are also sometimes used (68).

6.4. Casting and Preservation

A best practice for bolt cutter casting was presented by Piper (49). Besides giving general advantages of casts like easy manipulation and suitability for microscopic examinations, the author notes that in particular for those bolt cutters 'where there is an area of metal within the cut surfaces that can be ascribed to the leading edge of the bolt cutter blade', casting can be advantageous for comparison. Wang (53) took two common materials for casting toolmarks used in China, Elite H-D + Light body dental impression material (Zhermack SPA, Rovigo, Italy) and L001 special elastomeric impression material (Beijing fenge science limited company) and compared them qualitatively for dimensional accuracy, also over time (up to 1 month), air bubbles, ease of use as well as 'sharpness and quality of the individual characteristics present on the casts'. The casts were made of hammer toolmarks in natural and painted wood, lead, iron and aluminum. The conclusions of the study are that both materials are suitable to copy individual characteristics, but that the dental impression material is superior in practice as it produces less air bubbles and stays more stable over time (about 0.25 % compared to about 1.75 % after one month).

6.5. Variability of toolmark characteristics

The variability of marks of tools like screwdrivers or chisel may be very high, as it is dependent on many parameters like the angle of attack, the substrate, the axial rotation angle as well as the depth of the mark. Several authors studied the effect on some of these parameters on toolmarks, namely the angle of attack (55; 56; 57; 63; 73), the substrate material (56), the toolmark depth (56), the direction of tool movement (pushing vs. pulling) (56) and tool wear (56). All experimental results in this section are based on automated methods.

6.5.1. Angle of attack

Baiker *et al.* (55) present an experiment including 50 new flat head screwdrivers of the same brand and type. These were used to make marks at five different angles of attack (measured with respect to a plane perpendicular to the substrate), 15 °, 30 °, 45 °, 60 ° and 75 °, in Cavex dental wax slabs (74). To reduce mark variability, an in-house developed motorized mark generator was used. Subsequently, 3D surface data of toolmark casts was acquired, the toolmarks were aligned automatically and compared using cross-correlation as the similarity metric. Known match (KM) distributions of marks at 0 °, 15 ° and 30 ° difference in the angle of attack were determined, as well as known non-match (KNM) distributions and it could be shown, that the KM and KNM distributions are very well separated for 0 ° (FPR = 0.00 %, FNR: 0.00 %), well separated for 15 ° (FPR = 3.00 %, FNR: 0.78 %) and still moderately separated for 30 ° (FPR = 36.67 %, FNR: 11.51 %) difference in the angle of attack. The experiment was repeated using 2D photographs of the marks, made with a subset of the tools. The results were slightly worse compared to the results obtained with the 3D surfaces. Larger differences are expected with used tools. Marks of another type of flat head screwdriver were compared as well, again yielding similar results. Using the KM and KNM distributions, a strategy is presented to calculate likelihood ratios (LR) to determine the strength of the evidence for the result of a comparison of striated toolmarks.

The influence of the angle of attack was also studied by Lock and Morris (63; 73). The authors modified the automated method for comparing striated toolmarks, previously published by Chumbley and Morris (75), by explicitly modeling the difference in the angle of attack between marks. To test the extended method, 6 screwdrivers from a batch of 50 sequentially manufactured screwdrivers were taken to create four marks in lead each for different angles of attack (measured with respect to the substrate) 30 °, 45 °, 60 °, 75 ° and 85 °. During mark creation, the tool was fixed in a jig to reduce angle of attack variation. Toolmark profiles for each of the marks were acquired using a profilometer and statistically analyzed. The results suggest that including the difference in the angle of attack in the model does have a positive effect on the ability to obtain separate KM and KNM distributions and in addition allows to estimate the approximate angle of attack, as long as the two marks are created at a difference in the angle of attack of < 10 °. For angle of attack differences of > 10 °, KM profiles comparison results were similar to those of KNM profiles.

6.5.2. Substrate material and toolmark depth

Baiker *et al.* also assessed the influence of the substrate materials wax and lead, also over time, the depth of a toolmark and the direction of tool movement (pushing vs. pulling) on toolmarks (56). Recently, also toolmarks made at various angles in the substrates brass, polyvinyl chloride (PVC) and aluminum were studied (76). The methods and experimental setup were roughly the same as described above. The results show that the toolmarks in lead are slightly less variable with respect to wax, and contain reliable details down to about 5 µm. For larger details, the differences were only marginal. Marks are most reliable at small angles of attack and as a consequence, pushing produced better marks for angles > 45 ° and pulling for angles < 45 °. Shallow marks are more reliable than deep marks.

6.6. Individuality of toolmark characteristics

Several articles compare toolmarks manually to assess the individuality of characteristics. Tools include lock pickers (51), bolt cutters (52), consecutively manufactured screwdrivers (48), letter stamps (50) and diagonal cutters (66). In addition, a large body of literature is available for automated comparison of marks made by screwdrivers (55; 57; 75; 63; 60), crowbars (76), slip joint pliers (59; 62), chisels (61; 68; 76), punches (68), bronze age hand tools (69) and lock picking tools (64; 65).

An article that may be interesting for judging the expected variation in a ground tool surface as a result of wear of the grinding tool in the factory, is presented by Lipiński *et al.* (77). The authors are not focusing on a forensic context but on presenting an automated system for measuring and analyzing the grinding tool surface over time. However, the results shown in the article might be of interest, as they indicate that abrasive wear of a grinding tools surface occurs if grains become blunt or if grains are missing, and that smearing can lead to variation of the surface quality and the surface topography of the product. In addition, particularly shaped dents can be present on the grinding tool surface over time, which may manifest themselves on the surface of the product. As a result, tools made at a later stage might e.g. contain relatively larger details, though being manufactured with the same grinding tool.

6.6.1. Manual analysis

The Mul-T-Lock is a high security lock cylinder and very difficult to open with a traditional picking tool (51). However, there exists a specific tool for picking this lock, the H&M Mul-T-Lock picking tool. To study whether this tool leaves class characteristics on the cylinder pins Volkov *et al.* set up an experiment with 15 new Mul-T-Locks, which were picked with 5 H&M Mul-T-Lock picking tools, 5 with another lock picking tool and 5 were opened with a key. Afterwards the locks were dismantled and compared qualitatively with a comparison microscope. The authors conclude that the H&M Mul-T-Lock picking tool does leave specific class characteristics. In a second article, the same authors assess whether a bolt cutter leaves specific class characteristics, as this would allow performing a quick preliminary examination with possibly excluding a tool without further time consuming detailed examination. To this end, 10 bolt cutters of various sizes and brands (2 × HIT, 4 × MCC, RECORD, SEIYO, TRUPER and one without a brand) were used to create cuts in a padlock, a steel bar and a chain ring. Subsequently, a comparison microscope was employed to measure the bolt cutter size and blade thickness and to compare it with the surface widths of the marks in the cut specimen. Based on the assumption, that all tools of the same batch share the same class characteristics, the authors conclude that the examiner can exclude a tool, 'if clear border lines are detected on the cut object and the width measurement between these lines does not match the blade's thickness (by a tolerance of 0.1 mm)'.

Consecutively manufactured tools are the worst case scenario for determining individual characteristics. King (48) presents an experiment with 10 consecutively manufactured flat head screwdrivers, which were ground automatically during manufacturing by a computer numerical control (CNC) machine. In total, 20 experimental marks were created manually at about 15 ° with each screwdriver in weathered lead and subsequently sets of 20 marks were sent to 10 firearms and toolmark examiners for a blind test. Results of 7 examiners were returned and showed correct 'identification' in 62 out of the 70 cases, and 8 'inconclusives'. No false positives were encountered. Thompson (50) studied the characteristics of consecutively manufactured letter stamps. Several sets of stamps were used to create two impression marks of each stamp in lead and were cast. Subsequently the casts were compared with a comparison microscope. For one set, sub-class characteristics were found,

another showed individual characteristics and yet another was 'practically devoid of meaningful individual characteristics'.

Heikkinen *et al.* (66) used 'three diagonal cutters from the same production batch' and produced 1050 cuts in copper wire (diameter 2 ± 0.1 mm), which were then acquired in 2D using a comparison microscope and in 3D using white light interferometry and confocal microscopy. Marks created with different areas of the cutter, marks created at different points in time and marks of different tools were subsequently compared using the consecutive matching striae (CMS) criterion (78). How the 3D surfaces are converted to a striated pattern is not specified in the article. The CMS matching resulted in 74 out of 80 being correctly identified. No false positives were observed. Different areas on the same cutter turned out to be as different as marks from another tool and the marks did only marginally vary over time (1000 cuts in between).

6.6.2. Automated analysis

Screwdriver marks were studied for individuality by Lock and Morris (63) and Baiker *et al.* (55) and both studies revealed that toolmarks made in the same substrate material of known matching tools could clearly be distinguished from marks of known non-matching tools. The experimental details of these studies are presented in section 6.5. For the technical details of the methods, please refer to the original articles. Baiker *et al.* (76) also compared marks of a set of 20 different flat head screwdrivers and 10 crowbars and it could be shown that for all tools the KM and KNM distributions were clearly separated.

Spotts *et al.* (61) and Grieve *et al.* (59) used the automated algorithm by Chumbley and Morris (75) to compare striated marks, and assessed the individuality of shear cut marks ('quasi striated') from slip joint pliers. This type of marks contains discontinuous groups of striations and therefore is more challenging than regular striated marks. Therefore, several parameters of the original method were determined empirically to be able to handle the new type of mark. For the experiments, 50 sequentially manufactured pliers were used to create 1000 cuts in copper and lead wires, with a diameter of 4.11 mm and 4.76 mm respectively. Subsequently, 3D surface data was acquired and profiles determined on two locations on the mark. The authors conclude that 'a high degree of separation in the data was observed although sufficient statistical separation was not achieved' and that 'more work is needed to increase the robustness of the identification process.'

Another contribution from Spotts and Chumbley (61) focuses on the individuality of striated patterns in impression marks created by chisels. Again the method by Chumbley and Morris (75) was used, after empirical determination of the parameters. The experiments consisted of creating 10 marks each with 50 sequentially manufactured chisels. Surface data was acquired and profiles determined on two planes, totaling 1000 profiles. The automated comparison yielded that complete separation between KM and KNM distributions could be achieved, in case outliers are not taken into account.

Zheng *et al.* (68) also focused on chisels, however on striated marks created with the tools and not on impression marks as discussed above. Toolmarks were made with 20 consecutively manufactured chisels in copper plates, two each, at a 90° angle (with respect to the substrate), controlled by a motorized toolmark rig. Profiles were determined using a stylus instrument. For comparison of profiles, the cross correlation function was employed. Twenty known profiles were compared with twenty unknown profiles and assigning the highest cross correlation value between profiles to the most probable match resulted in 100 % correct identifications. The cross correlation distributions of KMs and KNMs were clearly separated. A similar experiment was done with 20 consecutively manufactured punches. As punch marks are impression marks and cannot be easily captured with a stylus,

3D data was acquired using confocal microscopy. For comparison, the two-dimensional cross correlation function, or areal cross correlation function, was used. The highest value indicated the most probable match. As for the chisel profiles, the punch impressions could be identified 100 % correctly and again, the KM and KNM distributions were clearly separated.

An unusual application of toolmark identification was presented by Kovács and Hanke (69), who set up an experiment to automatically distinguish between replicas of 3 adzes, which are bronze age hand tools. An experimental archaeologist made 10 marks with each replica in wood and subsequently surface data of the tools and the marks were acquired with structured light scanning. The distinction between different tools was made based on a set of surface geometrical characteristics (slope and width values) at the working edge of a tool using a Geographic Information System (GIS). The analysis revealed that different bronze age woodworking tools show specific toolmark class characteristics.

How marks on locking cylinder pins can be segmented and analyzed automatically is presented by Clausing and Vielhauer (64; 65). The goal of their studies was to find the best way to distinguish between different methods of lock-picking: single pin picking, raking, pick gun as well as normal key usage, by testing the performance of 15 classification methods. To find out which classifier performs best, a 3D test set of 20 key pins from 4 locking cylinders, all opened with one of the four methods, was acquired using confocal microscopy. Each pin was subsequently pre-processed automatically to separate regions that include toolmarks from regions that do not contain toolmarks (64) and to separate marks by usage from production marks (65). Based on the results of the experiments, the tree based classifiers like e.g. 'Random Forest' and 'Rotation Forest' perform best, with true positive rates above 80 % and true negative rates above 70 % for almost all of the different methods of lock-picking.

6.7. Virtual and simulated toolmarks

To determine statistically meaningful properties of toolmarks given certain conditions, e.g. different angles of attack, the dataset should be as large as possible. However, creating and acquiring a large set of experimental toolmarks is very time consuming. Several authors therefore presented virtual toolmark generators that predict toolmarks using a 3D model of the tool working surface (58; 57) and/or analyze a set of real marks, determine a deterministic component and a stochastic component, and modify the latter in order to simulate new toolmarks (79; 73; 57). In this way it is possible to generate a large number of marks, for example at many different angles of attack, which can subsequently be compared with a suspect mark. This can be done manually or by the computer, which can automatically find the angle of attack producing the profile that best matches a suspect mark (57; 60). This saves time and has the additional advantage, that the state of the tool is not altered, as no experimental toolmarks have to be created. After the computer determined the optimum angle of attack, one experimental toolmark can be generated at the given angle for verification.

6.8. Software for automated analysis

Following the development of automated comparison methods for striated toolmarks (75; 55) and breech faces (70), several groups by now also developed graphical user interfaces that can be used for manually studying data, but also to test the methods that were developed by the groups. All these packages were presented at the AFTE (The Association of Firearm and Tool Mark Examiners) training seminar 2016 in New Orleans and are briefly demonstrated here. MANTIS is short for Mark ANd Tool Inspection Suite (80), and was developed by the group of Scott Chumbley from the Iowa State University (for the details on the method, please refer to Chumbley and Morris (75)). The software allows visualizing and manually

navigating through the data and automatically determine statistical measures that give an indication of whether different toolmarks are a match or a non-match. In addition, it is possible to load a surface model of a tool and calculate virtual marks. A system called the Cadre Forensics Virtual Microscopy Viewer (81), was presented by Ryan Lilien from Cadre Research Labs (82). As the name indicates, the software is simulating a conventional comparison microscope and basically consists of a split screen viewer with which two pieces of evidence can be compared. Full 3D manipulation like shifting, rotation and scaling are available, as well as additional features like filtering and lighting options. In addition, the two split screens can be linked together, for simultaneous data navigation. Finally, a graphical user interfaces called 'Scratch' (83), developed at the Netherlands Forensic Institute (84), was presented by Martin Baiker. It can be used to study and automatically compare striated marks of tools and firearms (e.g. land engraved areas or LEAs and primer shear marks), also with multiple LEAs simultaneously. In addition, the software can load surface models of tools, determine virtual toolmarks and compare these to real toolmarks. In addition, the software can automatically determine the actual angle of attack that leads to the best match between virtual and real toolmark. Finally, a likelihood ratio for the current comparison result can be determined, based on a known match and known non-match database.

7. Invasive striated and impression toolmarks

7.1. Acquisition

Most authors used conventional 2D microscopy or stereomicroscopy (85; 86; 87; 88; 89; 90; 91; 92; 93), but also scanning electron microscopy or SEM (90) to examine invasive toolmarks. Epifluorescence macroscopy was presented by Capuani *et al.* (94), arguing that the auto-fluorescent properties of bone can be exploited to improve the details of invasive toolmarks. Generally, if using 2D techniques small microscopic marks might be difficult to see, as a result of the reduction in the depth of field, particularly at very high resolutions. In addition, 2D images do not allow exact geometrical (objective) measurements. Therefore, some authors used digital microscopy (95; 96), for an increased depth of field and true 3D approaches including focus variation microscopy (97) and photogrammetry (98). Erickson *et al.* (99) discuss, how 3D datasets based on computed tomography (CT), magnetic resonance imaging (MRI) and surface scanning can be used to better visualize invasive traumas for the courtroom and to provide exact spatial measurements of traumas. González *et al.* (98) present a low cost solution for recording cut marks in bone using photogrammetry. They manually create 15 cut marks in 3 lamb bones, acquired the bone surfaces, automatically detect and align the marks and determine a number of real depth profiles perpendicular to the cuts. Eight characteristics relevant for the morphology, depth, width and angle are subsequently measured based on these profiles and the authors conclude, that these could be measured accurately.

7.2. Casting and Preservation

In case of invasive traumas, bone and cartilage are the most frequently examined materials. As bone might still emit fat until long after the victim passed away and cartilage can be slightly transparent, sometimes traumas are cast and/or replicas are made. Besides being more convenient to handle, casts can facilitate the detection of marks that are very hard to see otherwise. Casts also offer a solution in cases, when samples are too big to be measured conveniently. Clow and Lançon published an article on collection, preservation and examination of sharp force injuries (87). The article is mainly intended for forensic pathologists but also contains useful information for the toolmark examiner like suggestions for a casting material (red-brown MicroSil, no further details available) and creating experimental stabbing marks ('plastic/rubbery material'). Two articles (100; 101) describe using alginate and silicone for casting and type 4 plaster for creating replicas of sharp and blunt force traumas on skulls. It was noted that silicone casts better preserve fine details but

that both were sufficient for the case at hand. One of the articles (101) in addition presents a low-cost method to create a substrate for experimental stabbing marks, using a mixture of gelatin, sodium benzoate (food preservative) and acetic acid (vinegar). Dittmar *et al.* (102) present a comparison of three different materials, Xantopren L blue, MicroSil and Alec Tiranti RTV putty silicone, for casting toolmarks on fresh (sheep femora) and well preserved archaeological skeletal remains (animal bones). In total, 45 casts were analyzed with a macroscope and with a SEM and all three materials turned out to accurately copy the toolmarks. However, it was found that only the Alec Tiranti RTV putty silicone could be removed without leaving color stains and residue on the specimen. Despite the very long setting time of 45–60 min (vs. several minutes for the others), the authors recommend using this casting material.

As shrinkage of bone samples can occur if these are stored for a longer period, Bailey and Bailey (85) studied 14 antimicrobial solutions for their potential to prevent bone shrinkage. Toolmarks were created in 14 fresh porcine rib bones using a hack saw and stored for 6 months. Based on microscopic images taken prior to and after the storage period, the authors then decided qualitatively, whether shrinkage occurred or not. They conclude that buffered 10 % formalin, buffered 10 % formalin for 12 days with subsequent transfer to ethyl alcohol, 70 % isopropyl alcohol, 93 % ethyl alcohol, 5 % and 10 % iodine solution as well as 6 % sodium hypochlorite did prevent bone shrinkage but that they prefer solutions without formalin. Crystal and fungi growth respectively ruled out acetic acid and sodium chloride solutions as a suitable means for storage.

7.3. Variability of invasive toolmark characteristics

Two articles were found that study the variability of marks, one depending on burning (90) and the other depending on decomposition (92). Kooi and Fairgrieve (90) took 5 racks of domestic pig side ribs and inflicted more than 10 wounds on the dorsal and ventral part respectively with two single edged knives, one smooth, one serrated. Subsequently the rib racks were split and one part burned in an open fire pit for about 1 hour. Afterwards, the toolmarks on the fresh and the burnt part were examined for differences with a stereomicroscope and a SEM. The made observations include that linear cuts, V-shaped cross sections and hinge fractures were all observable in fresh and burnt samples, but that features such as mounding and wastage were often destroyed during burning. The overall prevalence of features was estimated to be about 40 %. These results are based on relatively coarse structures, as fine striations could not be observed on any of the samples. Oblique faulting and bone lifts were only possible to see with the SEM.

Spagnoli *et al.* (92) studied the effect of decomposition in air on cuts and blunt force marks on costal cartilage over a period of 4 months. The tools used were three different knives with smooth edges, a scalpel, a cutter and a flick-knife, three knives with a serrated-edges blade, a bread knife, a bowie knife and a steak knife, and a hammer. In addition, a force was applied to some of the samples by manual bending. In total, 52 samples of human costal cartilage were used to produce the marks and were compared frequently with a stereomicroscope with casts from the fresh samples to check for signs of decomposition. It turned out that after only one week of decomposition, the detection rate for striations fell from 44–88 % to 17–33 % for non-serrated blades and from 77–88 % to 28–39 % for serrated blades. Blunt force marks didn't show specific characteristics. The samples did not contain any striations after 1, 2 and 4 months.

7.4. Individuality of invasive toolmark characteristics

Many articles in the last years were published comparing various tools for differences in the characteristics of the marks they leave in bone (97; 94; 86; 95; 88; 96; 91), cartilage (95) as well as aortic tissue, kidney, skin, liver, cardiac and skeletal muscle (89). Tools of interest

were knives (97; 86; 95; 89), usually comparing serrated and non-serrated models, heavy bladed instruments like axe and machete (88) as well as hacksaws (94; 96) and chainsaws (91). One author presents automated approaches to distinguish between different tools (97) and three authors evaluate the performance of their approach quantitatively (97; 95; 96).

7.4.1. Manual analysis

A study involving serrated, semi-serrated and non-serrated knives was presented by Crowder *et al.* (95). Three examiners with varying degrees of experience in sharp force trauma analysis were studying the differences in knife class, knife edge bevel (left, right and even), direct vs. indirect (using casts) mark assessment and using a standard dissection microscope vs. using a digital microscope. To this end, in total 28 cuts were made in wax, 14 in deer bone and 14 in porcine cartilage and the marks were categorized into fine (microscopic) and coarse (visible to the naked eye), as well as a combination of both and none. The results show that generally marks from serrated knives could be distinguished from non-serrated blades, but distinction between serrated and semi-serrated blades turned out to be more difficult. If serrated and semi-serrated blades were grouped, classification accuracy could be increased from 79 % to 96 %. Classification accuracy for edge bevel was 65 %. Casting the marks as well as using different microscopes did not have an influence on the results, but the amount of experience in sharp force trauma examination did.

Jacques *et al.* (89) created experimental cutting marks with a serrated and a non-serrated knife in a variety of human tissues, aorta, skin, liver, kidney as well as cardiac and skeletal muscle and asked three forensic pathologists to assess the marks for the presence of striations. The study yielded that only the aorta and skin marks made with the serrated knife showed striations, which were labeled by the authors as class characteristics. All other tissues as well as the non-serrated knife did not leave discernible marks.

In an attempt to relate the geometrical characteristics of a cut mark to the used tool, Cerutti and colleagues (86) created 11 specifically forged 'knives' with varying properties such as width and symmetry of the blade as well as shape and highness of the bevel. With these tools, cuts in porcine femora were created, 110 perpendicular to the bone (10 cuts for each tool) and 110 with an inclination, and analyzed using a stereomicroscope by two examiners. The results show a large variation in the measured geometrical characteristics of the marks and it was not possible to use them for relating a mark to a specific weapon. The authors conclude that the physical properties of a bone like the geometry and the strength as well as the speed of the hit during mark creation (which was not constant), might have too large of an influence on the resulting mark to enable accurate cut mark classification. Inter-observer variation was small.

In her MSc thesis, Highsmith (88) studied the possibility to classify toolmarks caused by heavy bladed instruments. Two machetes, one serrated, and one axe were used by 4 volunteers to dismember 32 limbs of 8 wild hogs. In total, 141 discernible impact sites were found for the machetes (grouped) and 121 for the axe. The author then macroscopically assessed the traumas for kerf width and depth, frequency of kerves, incisive marks, lateral fractures, shattering, cluttering, crushing and bone cut through. Based on the measurements, statistically significant differences between the instrument types were found for the kerf width and depth as well as the frequency for shattering, crushing and bone cut through.

The individual characteristics of hacksaw marks were studied by Capuani *et al.* (94) and Love *et al.* (96). In the first article, two hacksaws with different tooth appearances (pointed, sharp and smooth, blunt) were selected to generate for each 10 false starts and 10 complete sections on porcine bone. These marks were imaged with epifluorescence macroscopy and

analyzed for 9 variables in case of a false start and 16 variables in case of a complete section. Comparing the results for the two saws revealed that size, raker set and ripcut shape were common to both and that it was possible to reconstruct the trauma. More specific characteristics were related to the shape and profile of the kerf, the consistency of cut and type of wall striations. While class and sub-class characteristics could be used to determine the saw type, individualization was not possible. The second article (96) compared the marks of four types of hacksaw, one electric, with alternating, wavy and raker tooth sets based on a set of 15 variables. In total, 58 experimental marks were made in human femurs, imaged using a digital microscope (Keyence VHX-1000 (103)) and studied for differences by 3 doctoral anthropologists. Based on the analysis, decision trees were implemented to test the discriminatory value of each variable using a subset of 4 variables, floor and wall shape, minimum kerf width and average tooth hop and another one using 3 variables, wall shape, minimum kerf width, average tooth hop. Minimum kerf width, floor and wall shape as well as average tooth hop turned out to be important variables and based on these, the accuracy in saw type determination ranged between 83 % and 91 %

A contribution studying the individuality of various types of chains used for chainsaws was found in the MSc thesis of Moore (91). Five types of chain (chisel and standard tooth with standard skip, chisel tooth with full skip as well as semi-chisel tooth with semi- and full skip respectively) were mounted on the same chainsaw and used to produce 20 complete sections and 10 false starts each in deer long bones. The results show that chainsaws marks can be distinguished from other types of saws based on kerf width, severity of fragmentation and pitting in the kerf wall, but a clear distinction between the various chains could not be reached in general. However, some trends were observed comparing the size of exit chipping, the size of the breakaway notch in complete sections, the angling of the kerf floor in false starts and the mass of bone wastage that might enable to distinguish between standard teeth and chisel-/semi-chisel teeth chains.

7.4.2. Automated analysis

Bonney (97) published a study that aimed at automated classification of cut marks in bone made by a serrated knife (steak knife), a non-serrated carving knife and a non-serrated knife with a bamboo blade. In total, 10 cuts were made in the dorsal and ventral part of porcine rib specimens respectively with each of the knives. Subsequently, 3D surface data was acquired using a focus variation technique (Alicona IFM (104)). For data acquisition, casts were made from the samples as they were too big for scanning and provided better contrast. From the surfaces, profiles were taken perpendicular to the direction of the cuts and 8 geometrical features were measured. An automated discriminant analysis based on these features then served to classify 86.7 % of the blades correctly (note that the same dataset was used for building and testing the classifier). A sample set including cuts from unknown origin was also analyzed.

7.5. Case studies for comparing invasive toolmarks

Three case studies, in which a suspect tool could be identified with high confidence as being used in a murder, were presented by Weber *et al.* (93). In two cases, knife marks were found in costal cartilage and in one case, hack marks were found in bone. The article describes the steps that were taken during the examination including the generation of experimental marks and casting and presents images with comparison results. The authors conclude that the presented cases are encouraging, because they demonstrate that marks in human bone and cartilage can be useful for tool identification.

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Criminalistics

Paint and Glass, 2013-2016¹

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1. Introduction

This review paper covers advances in scientific methods applied to the forensic examination of glass evidence since the publication of the 16th International Forensic Science Symposium in October of 2010 (given that a review of the glass literature was not undertaken in 2013) and advances in the forensic examination of paint evidence since the publication of the 17th International Forensic Science Symposium in October of 2013. This chapter covers a review on both of the subjects (glass and paint) using the peer-reviewed literature, published reports, books and book chapters on the subjects as well as highlights of presentations and proceedings from forensic science meetings and symposia. Forensic examiners must also be aware of the publication of standard guidelines and test methods as well as the developments within the manufacturing industries including production volumes, production locations, and the current trends in the manufacture of these widely used materials.

Glass is defined as an inorganic product of fusion that has been cooled to a rigid condition without crystallization (1). This material is composed of a mixture of inorganic components that are responsible for its different physical properties. Glass has been identified as a “model trace evidence matrix” due to the following characteristics of this material (2):

1. It is a commonly encountered as evidence due to its fragile nature.
2. It is easily transferred from source to scene, victim, or suspect.
3. It does not degrade significantly over time, and once transferred, it can persist on objects after transfer.
4. Fragments of sufficient size and nature are recovered, which make them suitable for analysis by a number of different methods.
5. Standard methods have been developed to determine the optical and chemical properties of glass.
6. Suitable reference materials are available from a variety of sources with known ground truth property values that can be used for validation studies, calibration procedures and to determine the analytical figures-of-merit for the examinations including error rate determinations.
7. The optical and chemical properties are relatively homogeneous within a single pane or sheet of glass and the differences among manufacturing sources of glass are much greater than differences within a single manufacturing source.

¹ Glass Review: 2010-2016; Paint Review: 2013-2016

8. The physical-chemical measurements result in continuous, numerical values that can be subjected to statistical analysis methods to aid the interpretation of the results. These data can also be used to assess frequency of occurrence of RI and/or elemental composition, for a given population of glass.
9. The scientific literature describing glass analysis spans more than five (5) decades and now includes four (4) international standard methods providing the scientific foundation to the forensic examination of glass evidence.

2. Overview

The field of forensic glass examination has advanced considerably over the last six (6) years since the publication of the previous INTERPOL review on the subject. In particular, advances in the elemental analysis of glass have been reported in both the analytical chemistry and the forensic science literature. Of notable importance, three (3) international (ASTM) standards describing the elemental analysis of glass have been published and a fourth international (ASTM) standard related to the measurement of refractive index has been renewed. ENFSI-authored guidelines have also been developed and published.

2.1 Peer-reviewed literature

The main forensic science journals reviewed for this chapter were the *Journal of Forensic Sciences*, *Forensic Science International*, *Science and Justice*, the *Canadian Journal of Forensic Sciences*, the *Australian Journal of Forensic Sciences*, the *Journal of the American Society for Trace Evidence Examiners (ASTEE)*, the *European Paint and Glass (EPG) working group newsletter* and a new Elsevier journal initiated in 2016, *Forensic Chemistry*. In addition, more than ten (10) different analytical chemistry or other science journals have published peer-reviewed communications on the advances of forensic glass examination. In addition, the proceedings from several forensic and analytical chemistry conferences are briefly cited here and links to World Wide Web links and resources are also provided.

2.2 Additional publications

Several books include book chapters devoted to the forensic examination of glass and paint evidence. Of particular interest is the volume published in 2016 and edited by Jay Siegel, *Forensic Chemistry; Fundamentals and Applications* (2). This volume contains updated chapters on the subjects of “Analysis of Glass Evidence” by Almirall and Trejos and “Paint and Coatings Examinations” by Kirkbride. The updated chapters cover, not only the current and state-of-the-art examinations but also provide new information on the current state of the interpretation for both types of evidence. The analysis of glass and paint was covered within a recent overview of “Forensic Applications of Mass Spectrometry” chapter in the *Encyclopedia of Mass Spectrometry* by Almirall and Trejos (3). A chapter discussing materials analysis, including glass evidence using “Laser Ablation Inductively Coupled Plasma Mass Spectrometry in Forensic Science” was published in the *Encyclopedia of Analytical Chemistry* (4) by Trejos and Almirall. Paint and glass are also covered in an edited volume (102).

Of particular significance to forensic glass examiners that conduct measurements for comparing glass fragments, four (4) ASTM standards were published within the last six (6) years. The “Standard Test Method for the Automated Determination of Refractive Index of Glass Samples Using the Oil Immersion Method and a Phase Contrast Microscope” (E1967-11a) was renewed for the third time in 2011 (5). The standard test method for the “Determination of Trace Elements in Glass Samples Using Inductively Coupled Plasma Mass Spectrometry” (E2330-12) was renewed in 2012 (6). A new standard test method for the “Forensic Comparison of Glass Using Micro X-ray Fluorescence (μ -XRF) Spectrometry” (E2926-13) was published for the first time in 2013 (7) and a new standard

test method for the Determination of Trace Elements in Soda-Lime Glass Samples Using Laser Ablation Inductively Coupled Plasma Mass Spectrometry for Forensic Comparisons” (E2927-13) was also published for the first time in 2013 (8). In addition to these international documentary standards developed by ASTM, a “Guideline for Evaluative Reporting in Forensic Science” was published in 2015 (9) by the European Network of Forensic Science Institutes (ENFSI) under the “Strengthening the Evaluation of Forensic Results across Europe (STEOFRAE) initiative. This guideline provides “all reporting forensic practitioners with a recommended framework for formulating evaluative reports and related requirements for the case file”. The guideline is supportive of the use of likelihood ratios to report forensic results and provides case examples, including an example for a glass case comparison. The project core group involved with the creation of this ENFSI document included scientists from Forensic Science Ireland, the INCC in Belgium, the NFC in Sweden, the NFI in Netherlands, the Guardia Civil of Spain, the IRF of Poland, the University of Edinburgh in Scotland, LGC Forensics in the UK and the University of Lausanne in Switzerland. Interestingly, at the Annual Meeting of the Expert Working Group Paint & Glass that took place at the end of September 2013, the majority of EPG members (31 of 37 ENFSI institutes) rejected the proposed guideline for several reasons including the need to present alternative approaches to interpretation (Personal communication with Stefan Becker, BKA).

2.3 Conferences and Symposia

The following scientific conferences and symposia contained presentations on either glass or paint and coatings examinations and some of the following contained presentations on both topics. The list of conferences below is provided in chronological order and includes the name of the conference followed by the year the conference was held and then includes a brief description of the glass and paint/coatings presentations. Finally, the web site that contains the program and proceedings for the conference is also provided.

- American Academy of Forensic Sciences (2010-2016). Various poster and oral presentations focusing on the analysis of glass have been included in the American Academy of Forensic Sciences’ annual meeting. The link to each year’s proceedings is as follows: <http://www.aafs.org/resources/proceedings/>.
- SciX (2010-2015) The annual SciX meetings have included several sessions focusing on forensic science, including trace evidence analysis including a presentation on chemometric analysis of glass evidence in 2015. The final program for each year can be found in the archives: <https://www.scixconference.org/program/archive>.
- American Chemical Society (2010-2016) The annual American Chemical Society meetings have included presentations focusing on forensics within the Analytical Chemistry section*. The link to each year’s program is as follows: <https://www.acs.org/content/acs/en/meetings/nationalmeetings/programarchive.html>. Glass was not mentioned in any session, but paint was included.
- International Forensic Science Symposium (2010, 2013) Interpol hosts a Forensics Symposium every three (3) years. The 16th International Forensic Science Symposium in 2010 included a session focusing on glass and paint(<http://www.interpol.int/INTERPOL-expertise/Forensics/DVI-Pages/International-Forensic-Science-Symposium>). The 17th International Forensic Science Symposium in 2013 included topics on paint analysis (<http://www.interpol.int/INTERPOL-expertise/Forensics/Forensic-Symposium>).
- International Symposium on the Forensic Sciences (2010, 2012, 2014) The International Symposium on the Forensic Sciences is hosted by the Australian and New Zealand Forensic Science Society. The 22nd symposium in 2014 included presentations focusing on paint analysis; the agenda can be found in:<http://>

- www.aomevents.com/ANZFSS2014/Program_Workshops/Symposium_ProgramThe agenda for the 20th (2010) and 21st (2012) symposium could not be located online.
- European Academy of Forensic Science Conference (2012, 2015) The 7th symposium (2015) included poster presentations discussing glass and paint analysis (<http://www.eafs2015.eu/poster-sessions.htm>). The agenda for the 6th symposium (2012) could not be located online.
 - Trace Evidence Symposium 2011: Science, Significance and Impact (2011) The National Institute of Justice hosted a trace evidence symposium in which oral presentations and/or posters on both glass and paint were presented. <http://projects.nfstc.org/trace/2011/agenda.htm> and http://projects.nfstc.org/trace/2011/images/TES2011_Final_Agenda.pdf
 - Impression Pattern and Trace Evidence Symposium (2015) Hosted by the National Institute of Justice and the Forensic Technology Center of Excellence, this symposium included oral presentations and/or posters on both glass and paint. <https://rti.connectsolutions.com/p6psgg6hkpm/>
 - Trace Evidence Data Workshop: Improving Technology and Measurement in Forensic Science (2016) The National Institute of Standards and Technology hosted a workshop which included talks and discussions about the various databases, including those pertaining to glass and paint, that have been established throughout the world. (Link to website is not available at the time of this writing).
 - Annual IFRI Forensic Science Symposium (2012-2016) The International Forensic Science Research Institute hosts an annual symposium, which have included poster and oral presentations focusing on the analysis of glass and paint.
 - 2012 Annual IFRI Forensic Science Symposium Poster presentation on glass; <http://ifri.fiu.edu/news/2015/ifri-4th-annual-symposium-great-success/ifri-forensic-science-symposium-2012-program.pdf>
 - 2013 Second Annual IFRI Forensic Science Symposium Oral presentation on glass; <https://issuu.com/fiupublications/docs/ifrisymposium>
 - 2014 Third Annual IFRI Forensic Science Symposium Oral presentation on glass and paint; <http://ifri.fiu.edu/events/2014/third-annual-forensic-science-symposium/forensic-science-program-2014-1.pdf>
 - 2016 Fifth Annual IFRI Forensic Science Symposium Poster presentation on glass; http://ifri.fiu.edu/news/2016/ifri-symposium-proceedings-available/2016_ifri-symposium_program.pdf

2.4 Industry Information

The National Glass Association (www.glass.org) is an industry-supported clearinghouse of information on the manufacture of glass including safety, education, and other types of general communications. The official communication arm of the NGA is the online publication Glass Magazine (www.glassmagazine.com). Many of the production statistics quoted in this review are cited from this source. A recent (2014) IBISWorld report (10) provides a succinct summary of the glass manufacturing industry: “Companies in this industry produce a wide range of glass products by melting silica sand or cullet and fabricating purchased glass. The industry includes four segments: flat glass manufacturing, including laminated glass; pressed or blown glass and glassware; glass container manufacturing, including bottles and jars; and product manufacturing from purchased glass, which includes lighting, mirrors, architectural glass and electronic glassware” (10). The glass manufacturing industry is expected to grow 5.5% from 2016-2021 due to an increase in new building construction and a healthy rebound in the automotive industry, two main consumer segments of glass manufacturing supply. The market segmentation of products and services from US manufacturers, for example, is shown in figure 1 (source: IBISWorld) (10). According to Glass Magazine, the publication sponsored by the National Glass Association,

this future increase in production follows a sharp reduction in capacity in North American glass production.

In Canada and the USA, there were forty-four (44) working float glass lines in thirty-seven (37) plants in 2005, and in 2015, there were thirty-four (34) lines in twenty-five (25) plants in operation (11). The two (2) plants that were operating in Canada in 2005 were shut down and similar levels of capacity reduction took place in western Europe (11). The level of activity in Europe is “far below” 2008 levels with the glass industry not yet recovered from the global recession.

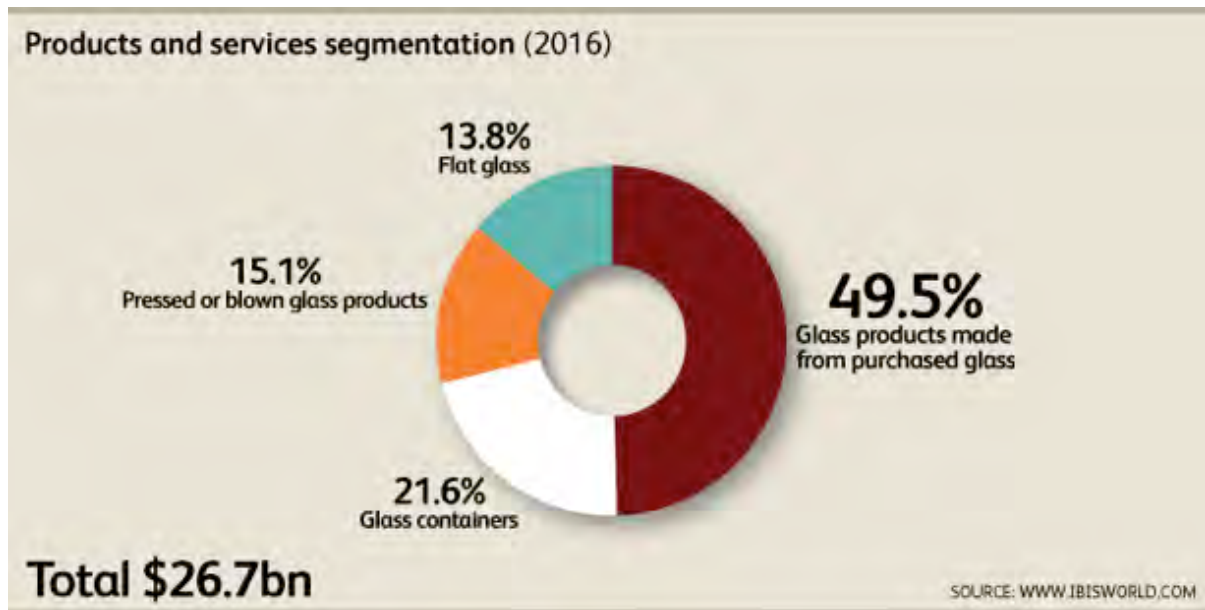


Figure 1. Market segments for glass manufacturing in the U.S.A. for 2016 (source: www.ibisworld.com)

During the same period, China and the other BRIC countries (Brazil, Russia, India, and China) experienced tremendous growth in the same decade (11). Chinese glass manufacturing grew from twenty-five (25) glass plants in 2005 to sixty-four (64) plants (125 lines) in 2015 and currently represents close to 50% of glass volume production globally. The other BRIC countries now have twenty-one (21) glass plants between them (up from eleven (11) in 2005) (11). Globally, there are ~400 float glass lines (210 float glass plants) currently operating and the total manufacturing output of all float glass plants in the world is on the order of 1 M tons every week (11). In China, plants that do not meet western manufacturing standards have closed recently so even though there has been consistent and significant growth in that market over the last ten (10) years there have also been some plant closures (12). An interactive map of all the current float lines including plant location and number of lines can be found at worldofglassmap.com (13).

This information is relevant and important to glass examiners because glass composition varies with the source of the raw materials as well as the formulations for end use. Some of the unintended contaminants (trace elements) present within the glass melt as a result of the geology from the raw materials and that do not affect the physical and optical properties of glass are detectable. Standard test methods (see above) have been developed to determine the chemical composition with sensitive instrumentation to provide a way of differentiating between glass sources. The larger the variety of geological sources of contamination producing trace signatures in glass, the greater the differences that can potentially be

detected among the different manufacturing sources. Given that products that contain glass such as vehicles are distributed and sold on a global market, there is an expected large variability of glass composition in these globally distributed products.

This review covers three main sections within each evidence type; 1) examinations, 2) transfer, persistence and databases and 3) interpretation.

3. Glass Examinations

A new automated procedure for the fast classification of glass fragments using differential interference contrast developed by Buchholz et al was reported to reduce the time it takes to classify glass fragments by their optical properties (14).

Tulleners et al developed a method for the determination of unique fracture patterns in glass and glassy polymers as part of NIJ-sponsored research and the final report is published on the NIJ web site: <https://www.ncjrs.gov/pdffiles1/nij/grants/241445.pdf> (15). Similarly, Haag et al examined the fracture of glass fragments during projectile penetration and perforation of glass (16).

In 2011, Ryland reported the improved discrimination of flat glass samples measured by μ -XRF with similar refractive index (17). In 2012, Ernst, et al reported the advantages of XRF over SEM as being more sensitive (nominal LODs of ~ 100 ppm for XRF vs ~ 1000 ppm for SEM-EDS), especially for elements of higher atomic number. As a consequence, XRF is reported as more discriminating than SEM, allowing not only the classification of glasses into categories but also a better discrimination among glasses of the same type (18).

In 2014, Cheng et al (19) reported the use of portable X-ray Fluorescence (PXRF) for the qualitative and semi-quantitative elemental analysis of glass. The major elements Si, O, Ca, Al, and Na, as well as trace elements Sr, Rb, K, Fe, and Sn were measured in twenty-five (25) glass samples by PXRF. The amounts of some elements, such as Fe, K, Zr, and Sr were found to vary in different samples, while other elements, such as Th, were found to be consistent in most tested glass samples. Discrimination of 98.31 % of 7,500 pair-wise comparisons created from twenty-five (25) glass samples was found.

In 2013, the Elemental Analysis Working Group (EAWG) performed a series of inter-laboratory tests that compared the analytical performance of μ -XRF, ICP-MS and LA-ICP-MS for the analysis of glass fragments manufactured in the same plant at short time intervals (20). In the same year, the EAWG reported on the performance of different match criteria for comparing elemental composition comparisons using the same techniques (21). The foundational work described in these two (2) publications resulted in the drafting of ASTM E2926-13 method for the use of XRF in glass examinations by the members of the EAWG (7).

Cahoon and Almirall continued the evaluation of the utility of Laser Induced Breakdown Spectroscopy (LIBS) for application in glass analysis by determining the wavelength dependence of the irradiation laser on the forensic analysis of glass (22). These workers concluded that the UV 266 nm laser couples better with clear, colorless glass and therefore results in better analytical figures of merit than the IR 1064 nm laser. These results were not unexpected as similar results have been previously reported not only for LIBS analysis but also for laser ablation analysis.

Koch and Günther produced a general review of the state-of-the-art of laser ablation inductively coupled plasma mass spectrometry and included optimization of LA-ICP-MS parameters for materials, including clear and colorless materials (23). LA-ICP-MS has been

reported to have many advantages over the solution ICP methods (2) including minimum sample consumption during the analysis (approximately 300ng) and the analysis of very small fragments (as small as 0.2 to 0.4ug or ~0.1mm to 0.4mm can be analyzed in several replicates using this method (7, 20, 21).

Weis et al (24) reported the utility of LA-ICP-MS analysis for the analysis of glass including the reporting of figures of merit using “ground truth” reference materials and also establishing a match criterion in forensic comparison analysis of float glass. This work, elaborated at the BKA in Germany reported that the best performing match criteria is the use of a “comparison interval” whereby the interval is ± 4 s (standard deviation with a minimum 3% RSD) is used. These workers report a maximum error rate of 2 out of 1891 false inclusions (0.1% false inclusions). The BKA analysis incorporated sixty-two (62) samples with six (6) replicates from each sample of survey glass from different glass plants. The same match criteria (± 4 s with a minimum 3% RSD) was used by the EAWG (20, 21) in the USA for a collection of 104 samples (3 replicates) and a type 2 error rate (false inclusions) was reported at a rate of 0.3% (36/10712 comparisons) and the population of the glass samples was also survey samples from different manufacturing plants or samples from the same plant but manufactured at different times. Dorn et al (25) separately reported the discrimination of float glass by LA-ICP-MS and evaluated the match criteria using casework samples and also determined that the use of a ± 4 s (with a minimum 3% RSD) performed the best. This work is of particular importance because it reports the use of eighty-two (82) casework glass samples to generate the LA-ICP-MS data (nine (9) replicates). The estimate of the probability of false “matches”, or the Type II error rate was ~ 0.1% (or 7/6642 comparisons). The agreement between the three different research groups in Germany, Canada and the USA prompted the development of the ASTM E2927-13 method for glass analysis using LA-ICP-MS and including the recommendation of the use of the ± 4 s (with a minimum 3% RSD) as a match criteria when comparing glass data by LA-ICP-MS. It should be noted that the EAWG reported a detailed comparison of a variety of match criteria for comparing LA-ICP-MS and XRF data (21) with the ± 4 s (with a minimum 3% RSD) match criteria as resulting in the best performing match criteria. Table 1 summarizes the results from the three (3) different groups.

FIU Collection 104 samples, 3 replicates BKA Collection 62 samples, 6 replicates CFS Collection – 82 samples from casework, 9 replicates	Type 2 Error Rate (%) False Inclusion	Type 2 Error Rate (%) False Inclusion	Type 2 Error Rate (%) False Inclusion
	FIU²¹ Florida International University	BKA²⁴ Bundeskriminalamt	CFS²⁵ Centre of Forensic Science
Comparison Interval ± 4 * standard deviation (minimum 3% RSD)	0.3 (36/10712)	0.1 (2/1891)	0.1* (7/6642)
T-Test (Welch’s Modification) 95% confidence, Bonferroni correction	2.2 (117/5356)	--	--
T-Test (equal variance) 95% confidence, Bonferroni correction	0.5 (29/5356)	0 (0/1891)	--
Equivalence Test θ calculated with known	1.9 (206/10712)	--	--
Equivalence Test θ calculated with Cardinal glass	0.02 (2/10712)	--	--
Equivalence Test θ calculated with FIU Database	2.6 (277/10712)	--	--

Table 1. Error rates for the comparison of glass samples in collections using LA-ICP-MS data (Source: 24, 25 and Symposium of Trace Evidence Databases).

Jisonna, et al reported the use of particle induced X-ray emission (PIXE) in the forensic analysis of tempered sheet glass (26). The elemental concentrations of five (5) trace elements were determined using this technique. The trace element concentrations for Ca, Fe, Mn, Sr, and Ti were compared to those obtained by inductively-coupled plasma-atomic emission spectrometry (ICP-AES) following complete digestion by hydrofluoric acid with good agreement between both techniques. The limits of detection for trace elements are typically lower (better) for the ICP-AES method. However, these workers show that the concentrations of these five (5) elements can be accurately measured by the PIXE method and given the non-destructive nature of the analysis, PIXE can be used to complement ICP-OES for the analysis of glass, according to these authors.

DeYoung, et al also used the PIXE technique to compare glass fragments (27). These workers were able to “identify those glass fragments that originated from different sources based on their elemental analyses”. The developed protocol includes specific approaches to calculating uncertainties and handling measurements below the level of detection. The results indicate that this approach has increased sensitivity for several elements with higher atomic number compared with X-ray fluorescence methods. While not as sensitive as LA-ICP-MS or ICP-MS methods of dissolved samples, these workers describe a simpler sample preparation process that may be used to presort glass fragments for more comprehensive elemental analysis at a later time.

Schenk and Almirall described a comprehensive comparison for the analysis of glass between LA-ICP-OES (28) to LA-ICP-MS and μ XRF/EDS. The development of a method for the forensic analysis of glass coupling laser ablation to ICP-OES was presented for the first time. LA-ICP-OES has demonstrated comparable analytical performance to LA-ICP-MS based on the use of the element menu, Al (Al I 396.15 nm), Ba (Ba II 455.40 nm), Ca (Ca II 315.88 nm), Fe (Fe II 238.20 nm), Li (Li I 670.78 nm), Mg (Mg I 285.21 nm), Sr (Sr II 407.77 nm), Ti (Ti II 368.51 nm), and Zr (Zr II 343.82 nm). The relevant figures of merit, such as precision, accuracy and sensitivity, are presented and compared to LA-ICP-MS. A set of forty-one (41) glass samples was used to assess the discrimination power of the LA-ICP-OES method in comparison to other elemental analysis techniques. This sample set consisted of several vehicle glass samples that originated from the same source (inside and outside windshield panes) and several glass samples that originated from different vehicles. Different match criteria were used and compared to determine the potential for Type I and Type II errors. It was determined that broader match criteria is more applicable to the forensic comparison of glass analysis because it can reduce the affect that micro-heterogeneity inherent in the glass fragments and a less than ideal sampling strategy can have on the interpretation of the results. Based on the test set reported here, a plus or minus four standard deviation ($\pm 4 s$) match criterion yielded the lowest possibility of Type I and Type II errors. The developed LA-ICP-OES method has been shown to perform similarly to LA-ICP-MS in the discrimination among different sources of glass while offering the advantages of a lower cost of acquisition and operation of analytical instrumentation making ICP-OES a possible alternative elemental analysis method for the forensic laboratory.

Grainger et al (29) reported on a classification and discrimination study of 243 automobile glass samples in New Zealand using LA-ICP-MS. An intact side window (tempered pane) and an intact windscreen (laminated pane) were analyzed to investigate the spatial trend of elements in automotive glass. Most elements displayed no spatial variation over the panes. Pb had the largest variation in the windscreen and was also found to have a large variation in the database. Most samples were able to be classified into the vehicles' country of origin

using a multiclass classifier. However, this was not possible for all samples, due to the origin of glass differing from the origin of the vehicle in some cases. The elemental composition of Australian and Northern Hemisphere samples differed greatly making them easy to separate; however, there was little variation within the Australian samples, making it hard to discriminate between different samples. A three-step method, which combined the use of elemental composition, ΔRI and RI , was used to discriminate database samples. The method distinguished 84% (172/204) of samples in the database. When Australian samples were removed from the analysis, the discrimination increased to 95% (148/156).

In an Australian study using LIBS, El-Deftar et al (30) was able to discriminate 97% of comparison pairs using fourteen (14) laminated and six (6) non-laminated glass samples, but no attempt was made to determine the number of correct associations in a blind study as would normally be conducted in this type of study. The Type II error rate (false inclusions) was not determined.

Sjåstad et al (31) reported the use of Pb-isotopic ratios to discriminate glass samples. These workers developed a method for analysis of Pb-isotope ratios using a Multi-Collector ICP-MS instrument requiring digestion of the sample prior to solution analysis.

In a different study, Sjåstad et al (32) presented the analytical protocol for optimizing isotopic determination of Pb for comparison of glass by use of LA-ICP-MS to determine isotope ratios. In a third study the following year, Sjåstad et al (33) report the application of LA-multicollector-ICP-MS in the determination of Pb-isotope ratios in common glass for forensic purposes. Finally, the same research group reports in a paper by Martyna et al (34) the application of likelihood ratios (LRs) for the comparison of data collected Pb-isotope ratios. The assessment of the applied LR models performance was conducted by an Empirical Cross Entropy (ECE) approach. Thirty-five (35) glass samples were subjected to IRMS analysis and were described by Pb-isotope ratios: $208Pb/204Pb$, $207Pb/204Pb$, $206Pb/204Pb$, $208Pb/206Pb$, and $207Pb/206Pb$. Univariate and bivariate LR computations were performed, assuming normally distributed data subjected or not to a logarithmic transformation. Principal Component Analysis (PCA) was employed for creating orthogonal variables to propose an alternative LR model. It was found that the application of variable $208Pb/204Pb$ seems to be promising as it delivers one of the lowest percentages of false positive and false negative rates as well as being the only variable for which an ECE plot gave satisfactory results.

Lee et al (35) report a discrimination study of side-window glass of Korean automobiles using LA-ICP-MS. Thirty-five (35) samples from the side windows of cars produced and used in South Korea were collected from the official agencies of five (5) car manufacturers and from two (2) glassmakers. In addition, 120 samples from side mirrors were collected from the same suppliers as well as from small businesses. Their chemical compositions (including Pb-isotopes) were analyzed using LA-ICP-MS and linear discriminant analysis (LDA) was performed. The percentages of major elements (Si, Ca, and Fe) in side-window glass varied within narrow ranges ($30.0 \pm 2.36\%$, $5.93 \pm 0.52\%$, and $0.33 \pm 0.05\%$, respectively), while the differences among Pb-isotope ratios were not significant. The light rare earth elements (LREEs) were different from each glassmaker. From the LDA, the types of side-window glass were successfully discriminated according to car manufacturer, glassmaker, and even glass thickness. However, glass from side mirrors were not determined as good forensic identifiers.

Baidoo et al (36) used a radiochemical technique, an application of k_0 -method in instrumental neutron activation analysis for glass analysis using a low power nuclear research reactor. In this work k_0 -INAA was been applied on glass samples to determine

major, minor and trace element concentration. As many as fifty (50) elements were detected and quantified with 3–5 mg of 0.1 % AuAl comparator monitor (0.1 % gold–99.9 % Aluminum wire). The average concentrations of SiO₂, Na₂O, CaO, Al₂O₃ and MgO ranged between 76–96 %, 11.15–12.66 %, 5.26–10.71 %, 1.13–2.73 % and 3.51–6.23 % respectively. The relative concentrations of impurity elements; Cr, Fe, Mn, and Co determined from the glass samples were used to match the physical appearance (color) of the glass based on general knowledge of colored glass production. The analytical procedure was validated using SRM 610 (glass matrix) and SRM GBW07106 (rock matrix) both as control samples which indicated a relative uncertainty of 15 and 6 % respectively for SRM 610 and SRM GBW07106. The authors opined that the relative sensitivity at which some of the elements were detected in major, minor, and trace levels have indicated that the k₀-method in instrumental neutron activation analysis using low power research reactor could be a useful technique in glass analysis for forensic and archeological applications.

Rushton et al (37) have reported on the effect of annealing on the variation of glass refractive index values of non-tempered, float glass pane and a tempered, float glass pane. The two (2) panes of colorless, float glass were cut into 150 5 cm × 5 cm squares. The pre- and post-annealing RI values from three random areas from each square were measured. Bayesian statistical hierarchical modeling of the results showed that, for the non-tempered, float glass pane annealing increased the variability in RI by a factor of 1.29–1.58, with a mean of 1.43 (with 95% probability); and for the tempered, float pane of glass annealing decreased the variability in RI by a factor of 0.63–0.76, with a mean of 0.69 (with 95% probability). In addition, these workers found that although there were no systematic differences in Δ RI across either pane of glass, there were observable differences across both panes of glass. These results provide information regarding the expected RI variation over entire panes of both non-tempered and tempered float window glass for both pre- and post-annealing RI measurements.

Funatsuki et al reported (38) the forensic analysis of automobile glass from three different manufacturers in Japan based on the RI, XRF, and X-ray absorption fine structure. The samples were classified into the corresponding groups using XRF, which should be the first step for identification. Samples having different manufacturing times showed differences in the refractive index. Based on XAFS, the amplitude of the EXAFS spectra and the intensities of Fourier transforms differed between manufacturers. In the scheme for manufacturer identification proposed in this study, performing XRF and refractive index studies is the first step. The concentrations of CeO₂, MgO, Al₂O₃, and K₂O allowed these workers to distinguish among manufacturers. For samples containing Ce, discrimination between manufacturer based on the amplitude of the EXAFS spectra and the intensities of Fourier transforms was possible. The same group (Funatsuki et al), reported (39) the forensic identification of automobile window glass manufacturers based on Ce chemical states. To identify automobile window glass manufacturers, the chemical states of Ce in twenty-nine (29) types of glass were analyzed using XAFS. These workers found that the amplitude of EXAFS spectra and intensities of Fourier transforms differed between manufacturers. Although the manufacturers for seven (7) out of twenty-nine (29) samples could not be identified, twenty-two (22) out of twenty-nine (29) samples were identified correctly.

Eyring et al reported (40) the microspectral characterization of green glass fragments. This study, published in *Microscope*, was a continuation of efforts to assist criminalists in sorting glass fragments with similar colors that might have originated from different sources. Microspectrophotometry (MSP) was used to address this problem and assist with fragment color sorting. The irregular shapes of glass fragments and their large refractive index (RI) differences relative to air makes an immersion mounting technique necessary prior to MSP

analysis. Following this preparation, the MSP sorting technique was applied to a group of twenty-five (25) green bottles.

Munger et al reported (41) on RI variations within panes of vehicle windshield glass samples. Refractive indices of seven (7) double-paned vehicle windshields were measured to assess the variation across the pane of glass and to evaluate collection techniques for known glass standards by comparing false negative rates. Measurements were made using a Foster and Freeman GRIM3 instrument, and a minimum of 240 measurements collected per pane. The mean SD of the windshields was 0.00004 RI units. It was further determined that collecting a known sample from two (2) different sections of a shattered windshield gave the lowest rate of false negatives when using ± 2 standard deviations to estimate the RI variation of the known glass. Additionally, refractive indices often were highest in the center of the windshield and decreased when sampled toward the edges, according to these workers.

Alamilla et al reported (42) a validation of an analytical method for the refractive index measurement of glass fragments. The validation was performed by studying analytical features such as the working range, precision, robustness, and bias. Locke silicone oil type B, glass standards type B and a sodium D source (589.3 nm) were selected for a working RI range from 1.50225 to 1.52381, according to the typical RI values of glass samples of forensic interest. The method was applied to eight validation samples (six (6) glass fragments from different parts of an automobile, a glass container, and an architectural tinted window), which were differentiated through their RI values. Finally, the procedure was applied to interpret the origin of glass evidence taken from a hit-and-run incident. A common origin of recovered and control glass fragments was supposed on the basis of the RI determination of these samples, together with the application of a reported match criteria in forensic pairwise comparisons of glass fragments. The results were confirmed by LA-ICP-MS analysis.

Michalska et al recently reported (43) an optimized sample preparation procedure for glass fragments for analysis by SEM-EDS. These workers report quantitative analysis using SEM-EDS by requiring a flat and smooth sample surface. To meet these requirements, instead of the typical embedding procedure, which is not always practical for minute fragments, these workers used optical microscopy for selecting glass fragments that are smooth and flat as possible and directly placing them on a scanning electron microscopy stub. The results using two (2) SEM-EDSs were compared for embedded and nonembedded glass standards. No significant differences in accuracy, precision, reproducibility, and false answer rates were observed using likelihood ratio models suggesting that the reported method of sample preparation is suitable for forensic analysis.

4. Glass Transfer, Persistence and Databases

O'Sullivan, Geddes and Lovelock (44) reported the migration of glass fragments from the pockets to surfaces of clothing garments in the United Kingdom. The aim of this study was to investigate the possibility that fragments of glass migrate from a pocket of a garment to its surfaces during police and laboratory handling after a person is suspected of breaking glass. Sixty (60) fragments of glass were seeded into a pocket of a fleece jacket and a pair of denim jeans. Three (3) experiments were performed; one examined a searching, recovery and blanking procedure, another examined the pre-laboratory 'handling' process of an item in an evidence bag, and the third experiment looked at the removal of an object from a pocket laden with glass and subsequent removal and packaging of the garment. Up to two (2) fragments were recovered from the surfaces of the fleece jacket and the denim jeans via the searching, recovery and blanking procedure. Similar numbers were also recovered from

the insides of the evidence bags. Up to four (4) fragments were recovered from the surface of the fleece jacket and up to five (5) fragments were recovered from the surface of the denim jeans after pre-laboratory 'handling'. Similar numbers were recovered from the insides of the evidence bags. Comparable numbers to those from searching/recovery experiments were observed when garments were removed after taking an object from a pocket. Their findings show that some migration can occur.

Irwin (45) reported on the transfer of glass fragments when bottles and drinking glasses are broken. These workers carried out experiments to determine if and how many glass fragments are transferred onto upper garments following breakage of bottles and drinking glasses. In all instances used in the study, glass was transferred. These workers report the number of transferred fragments after a bottle is broken ranges between 3-25 fragments and the number of fragments transferred following the breaking of a drinking glass ranges between 3-125 fragments.

Cooper (46) reported on the *indirect* transfer of glass fragments to a jacket and their subsequent persistence. This author conducted experiments to investigate the indirect, perhaps innocent, transfer of glass evidence. The experiments involved the transfer of glass fragments from a surface scattered with broken glass to a hand, and then from the hand to the sleeve of a poorly retaining jacket. The persistence of the transferred fragments was studied by collecting the glass fragments as they fell off the jacket while the wearer was walking on the spot. The results showed that large numbers of glass fragments can be picked up on a hand from a suitable surface and transferred from the hand to the jacket. In seven (7) of nine (9) tests performed, ten (10) or more glass fragments were recovered from the jacket sixty (60) minutes after the original contact between the hand and the broken glass. More than twenty (20) fragments were recovered in three (3) of these tests. These results call attention to the need to avoid secondary (or indirect) transfer of glass evidence given the possibility that "matching" glass from the scene or actual known sample is sometimes available for transfer.

Seyfang et al report (47) on the characterization of glass fragments originating from portable electronic devices (PEDs). PED glass is reported as easily recognized using SEM-EDS and RI measurements and is easily distinguished from domestic and automotive soda-lime glass using these methods.

Jackson et al reported (48) on a survey of glass found on the headwear and head hair of a random population vs. people working with glass. The study investigated the prevalence of glass particles on the headwear and head hair of two (2) different population groups; the general public who do not work with glass, and from glaziers who are people that work with glass and have regular contact with broken glass. The 232 samples collected from the head hair and headwear from the random population resulted in the recovery of six (6) glass fragments in total on six (6) individuals (i.e. one fragment each). All of these fragments were from head hair samples with no multiple fragments recovered. The two (2) headwear samples that were taken revealed no glass fragments. The head hair and headwear of twenty-five (25) glaziers from the glass workers produced 138 glass fragments found in total on twenty-four (24) of the twenty-five (25) glaziers. The size and number of fragments found in each sample were also generally larger for the glaziers group. The results from this study indicate that the prevalence of glass on the head hair and head wear of the random population is very low in comparison to the head hair and headwear of those who have regular contact with breaking glass. The significance of this finding with respect to the interpretation of glass evidence was also discussed.

Almirall, Corzo and Hoffman recently reported at a NIST Trace Evidence Database Conference on the existing, public databases containing forensic glass data such as RI and elemental composition. A survey was emailed out to a large number of laboratories known to conduct glass analysis and (6) six laboratories reported maintaining and using databases containing RI, elemental analysis or both types of data. Table 2 below summarizes the list of the existing databases including the number of samples, the types of samples in each database, the data that is collected (RI or elemental) as well as how the database is used in forensic casework or for research purposes.

Table 2. Existing databases from six (6) different laboratories and their reported uses of the databases (Source: Invited presentation by Almirall during the NIST Sponsored Trace Evidence Databases Conference in Gaithersburg, July 2016).

5. Glass Interpretation

Zadora and Ramos (49) reported the use of likelihood ratios (LRs) and an “information-theoretical approach” to evaluate the forensic comparisons of glass samples. The paper presents the influence of database selection for the analysis of chemical profiles determined by SEM-EDS. The use of empirical cross-entropy (ECE) plots is discussed and these authors conclude that the oxides of the major elements Ca, Si, and Na provide good discrimination between samples.

Zadora and Neocleous (50) report LRs to compare refractive index data with SEM-EDS data for glass comparisons concluding that the RI data with SEM-EDS data is “appropriate” for database comparisons.

Zadora, Neocleous and Aitken (51) report the use of a “two-level model” for the evaluation of glass evidence in the presence of zeros. These workers report that LRs provide a natural way of computing the value of evidence under competing propositions and propose LR models for classification and comparison that extend the ideas of Aitken, Zadora, and Lucy and Aitken and Lucy to include consideration of zeros. These authors view the presence of zeros as informative and model it using Bernoulli distributions. The proposed models are used for both the evaluation of forensic glass (comparison and classification problem) and paint data (comparison problem). Two hundred and sixty-four glass samples were analyzed by SEM-EDS and thirty-six (36) acrylic topcoat paint samples were analyzed by pyrolysis gas chromatography mass spectrometry. The results for glass comparisons was reported as “highly satisfactory” and the comparison of paints resulted in 3.0% false positives and 2.8% false negatives.

Neocleous, Aitken and Zadora (52) report on the transformations for compositional data with zeros with an application to forensic evidence evaluation. The authors here used a two-level multivariate likelihood ratio model for comparison of forensic glass evidence in the form of elemental composition data under three data transformations: the logratio transformation, a complementary log–log type transformation and a hyperspherical transformation. The performances of the three transformations in the evaluation of evidence were assessed in simulation experiments through use of the proportions of false negatives and false positives.

Lucy and Zadora reported (53) on the mixed effects modeling for glass category estimation from glass refractive indices. For this study, 520 glass fragments were taken from 105 glass items (container, window, or automotive). Each of these three (3) classes were defined as glass categories. Refractive indexes were measured both before, and after a programme of re-annealing. Because the refractive index of each fragment could not in itself be observed before and after re-annealing, a model-based approach was used to estimate the change in

refractive index for each glass category. The change in refractive index was then used to calculate a measure of the evidential value for each item belonging to each glass category. The distributions of refractive index change were considered for each glass category, and it was found that, possibly due to small samples, members of the normal family would not adequately model the refractive index changes within two (2) of the use types considered. Two (2) alternative approaches to modeling the change in refractive index were used, one employed more established kernel density estimates, the other a newer approach called log-concave estimation. Either method when applied to the change in refractive index was found to give good estimates of glass category, however, on all performance metrics kernel density estimates were found to be slightly better than log-concave estimates. These results and implications of these two (2) methods of estimating probability densities for glass refractive indexes were also discussed.

Ramos and Zadora (54) used an information-theoretical feature selection using data obtained by SEM-EDS for classification of glass fragments. The database used for this work consisted of 278 glass objects (automobile and architectural windows and containers) for which seven (7) variables based on SEM-EDS data are available. A multivariate model was described for the computation of the likelihood ratios with an Empirical Cross-Entropy (ECE) objective function used for feature selection. The model is applied to all the sixty-three (63) possible univariate, bivariate and trivariate combinations taken from the seven (7) variables in the database, and its performance is ranked by its ECE. The results are reported as “nearly perfect” discrimination between glass sources.

Napier et al reported (55) a composite Bayesian hierarchical model of compositional data with zeros. These workers present an approach for modeling compositional data with large concentrations of zeros and several levels of variation, applied to a database of elemental compositions of forensic glass of various use types. The procedure consists of partitioning the data set in subsets characterized by the same pattern of presence/absence of chemical elements and then fitting a Bayesian hierarchical model to the transformed compositions in each data subset. The model is assessed using cross-validation, and is reported to perform well in both the classification and evidence evaluation tasks.

In a separate communication, Napier et al report (56) an easy-to-use and freely accessible online application for the analysis of forensic glass fragments. The application is browser based and takes as input .csv or .txt files containing measurements from glass fragments obtained using SEM-EDS. The application was developed to (i) classify glass fragments into use-type categories (classification), and (ii) compute the evidential strength of two (2) sets of fragments under competing propositions (evidence evaluation). Detailed examples of how to use the application for both tasks are described. The suitability of the statistical methods used by the application was validated using simulation studies, and improvements upon previous methods were found in both tasks, according to these authors.

Garvin and Koons (57) reported an evaluation of match criteria used for the comparison of refractive index of glass fragments. RI measurements from five (5) float glasses were used via resampling to assess the frequencies of false exclusion errors for eight (8) comparison criteria as functions of the number of measurements. The comparison criteria were based on ranges, fixed intervals, and multiples of standard deviations of the known source measurements. The observed error rates for the eight (8) tests studied are between zero and ~ 35%, depending upon the match criteria, the number of measurements, and the RI distribution for a glass source. The authors state that the results of this study can be used to predict the false exclusion rate for a test criterion under a given set of conditions or to select test criteria that result in a desired error rate for these typical sheet glasses.

Weise recently reported (58) a comparison between a frequentist and a Bayesian approach to interpret glass refractive index univariate data. The author compares the use of probabilities (in a frequentist approach) to the use of probability densities in the Bayesian approach, using a casework as a database source. This effort caused the author to conclude that “nothing substantial new is gained by calculating likelihood ratios at the source level”.

Newton recently reported (59) an investigation into the variability of the refractive index of glass focusing on the effect of debris contamination on the RI variability. Not surprisingly the results suggest that the variability of refractive index measurements is increased when debris contamination is present on glass fragments.

Finally, Howes et al reported (60) on the readability of expert reports for forensic glass comparisons for non-scientist report-users. The authors argue that scientific language contains features that may impede understanding for non-scientists. These workers assessed the readability of expert reports (n = 78) of forensic glass comparison from seven (7) Australian jurisdictions. Two (2) main audiences for reports were evaluated: police and the courts. Reports for police were presented either as a completed form or as a brief legal-style report. Reports for court were less brief and used either legal or scientific styles, with content and formatting features supporting these distinctions. Simple suggestions, based on theory and past research, are provided to assist scientists to enhance the readability of expert reports for non-scientists.

6. Paint and Coatings Examinations

Wright et al reported (61) on the analysis and discrimination of single-layer white architectural paint samples using a variety of instrumental methods. Fifty (50) single-layer white architectural paints were compared to determine the discrimination power using FTIR with 68 undifferentiated pairs resulting, yielding a discrimination of 94.45%. After adding stereomicroscopy, SEM-EDS) and backscatter electron (BSE) imaging, and Py-GC/MS, the overall discrimination was 99.35%. The blind verification replicates were also correctly associated demonstrating a high degree of discrimination of single-layer white architectural paints using methods of analysis often encountered in forensic science laboratories.

Roberts et al reported (62) on the use of paint evidence to investigate fires. ATR-IR measurements were used to study the degradation of paint samples upon heating. Five paint samples (one clay paint, two car paints, one metallic paint, and one matt emulsion) were characterized by a combination of ATR-IR, Raman, X-ray fluorescence spectroscopy and powder X-ray diffraction. The thermal decomposition of these paints was investigated by means of ATR-IR and thermal gravimetric analysis. Clear temperature markers were observed in the ATR-IR spectra namely: loss of $\nu(\text{C}=\text{O})$ band, $>300^\circ\text{C}$; appearance of water bands on cooling, $>500^\circ\text{C}$; alterations to $\nu(\text{Si}-\text{O})$ bands due to dehydration of silicate clays, $>700^\circ\text{C}$; diminution of $\nu(\text{CO}_3)$ and $\delta(\text{CO}_3)$ modes of CaCO_3 , $>950^\circ\text{C}$. The results from this study suggest the possible use of portable ATR-IR for nondestructive, in situ analysis of paints to gain information about the fire.

Zięba-Palus and Trzcińska reported (63) on the application of IR and Raman Spectroscopy in paint examinations. A micro-Raman spectrometer equipped with several excitation lasers was used for the identification of pigments. Three cases comparing car paint are discussed in detail. The comparison of Raman spectra of paint chips found on clothing of a victim or smears found on body of a damaged car to those of paint chips originating from the suspect car enabled the identification the car involved. The authors state that this method can be useful in establishing the color and make of the car even when no comparative material is available.

Hutaniu et al reported (64) on recent applications of mass spectrometry in paint analysis. A general review on several applications of MS in the analysis of paint, artist's paints, and powder coatings components recently reported in the literature were presented.

Defeyta et al recently reported (65) on the use of Micro-Raman spectroscopy and chemometrical analysis for the distinction of copper phthalocyanine polymorphs in paint layers. In art analysis, copper phthalocyanine (CuPc) is often identified as an important pigment (PB15) in 20th century artworks. Given that PB15 is used in different polymorphic forms, the identification of the polymorph could provide information on the production process of the pigment. Raman spectroscopy, combined with chemometrics, was used to discriminate between polymorphs of pigment crystals in art works. The results obtained by Linear Discriminant Analysis (LDA), using intensity ratios as variables, demonstrated the ability of this procedure to predict the crystalline structure of a PB15 pigment in unknown paint samples.

He et al reported (66) on the characterization of automotive coatings. FTIR and Raman were used to characterize the organic components and SEM-EDS and ICP-MS were used to characterize the inorganic components. Two four-layered samples from a case were compared layer by layer as an example. FTIR, Raman, SEM-EDS, and ICP-MS provided similar results on the two samples.

Lavine et al reported (67) on the simulation of ATR-FTIR using a correction algorithm to allow ATR spectra to be searched using IR transmission spectra of the paint data query (PDQ) automotive database was presented. The reported correction algorithm to convert transmission spectra from the PDQ library to ATR spectra is able to address distortion issues such as the relative intensities and broadening of the bands, and the introduction of wavelength shifts at lower frequencies, which prevent library searching of ATR spectra using archived IR transmission data according to these workers.

Maric et al reported (68) on the use of synchrotron infrared imaging to assess the extent of interlayer component migration within multilayer automotive paint samples, with a particular emphasis on the cross-linking additive melamine. Two-dimensional infrared chemical images revealed that melamine consistently diffuses in select paint samples from the underlying basecoat into the outermost clear coat layer. Pigments from the basecoat were also found to migrate into the adjoining layers. This is significant as the relative abundance of both melamine and pigments will vary greatly depending upon the region of the layer analyzed. This component migration will may impact the information gleaned from a questioned sample via library searching software or multivariate statistics.

In 2014, Maric et al reported (69) on the use of synchrotron FTIR for the characterization of automotive primer surfacer paint coatings. The chemical diversity of electrocoat primer, primer surfacer and basecoats of automotive paint samples from 75 vehicles of international origin were examined. Significant diversity was found in the synchrotron FTIR data from the primer surfacer coats. Fourteen (14) discrete groups associated by manufacturing country and specific manufacturers (and even individual plants) were differentiated. The model generated from the primer surfacer was significantly more discriminating than a previous model generated from FTIR analysis of clear coats of the same vehicles. Analyses of the primer surfacer also avoids issues of possible environmental degradation and component migration observed with FTIR of clear coats.

Trzcinska et al reported (70) on the examination of car paint samples using visible microspectrometry for more objective measurement of color. Sixteen (16) samples of solid

and metallic bright and dark red paints taken from different cars were examined. Raman spectra were also produced in order to detect the pigment composition of the samples. Different criteria were used to develop a discrimination strategy between the samples.

Lavine et al reported (71) on the use of search prefilters for mid-infrared absorbance spectra of clear coat automotive paint smears using stacked and linear classifiers. By using stacked partial least squares classifiers and genetic algorithms for feature selection and classification, it was demonstrated that search prefilters can be developed to provide information from clear coat paint smears. Search prefilters developed using specific wavelengths or wavelet coefficients outperformed search prefilters that utilized spectral regions. Clear coat paint spectra from the PDQ database may not be well suited for stacking as there are few spectral intervals that can reliably distinguish the different sample groups (i.e., assembly plants) in the data. These workers report that, the similarity of the IR spectra within a plant group and the noise present in the IR spectra may be obscuring information present in spectral intervals.

Lavine et al followed this work with two additional studies (72,73) on the use of search prefilters for infrared library searching. Clear coat paint smears were analyzed using IR transmission spectra collected on a Bio-Rad 40A or Bio-Rad 60 FTIR spectrometer and an approach based on instrumental line functions was used to transfer the classification model between different instruments. In the first study (72), 209 IR spectra of clear coat paint smears comprising the training set were collected using one manufacturer of an IR spectrometers, whereas the validation set consisted of 242 IR spectra of clear coats obtained using a second manufacturer. In the second study (73), pattern recognition methods were used to develop the search prefilters (i.e., principal component models) to differentiate between similar but non-identical IR spectra of clear coats on the basis of manufacturer (e.g., General Motors, Ford, Chrysler) or even by assembly plant. Search prefilters to identify assembly plants were successfully validated using 10 blind samples provided by the Royal Canadian Mounted Police (RCMP) as part of a study to populate PDQ to current production years, whereas the search prefilter to discriminate among automobile manufacturers was successfully validated using IR spectra obtained directly from the PDQ database.

Suzuki reported (74) on information gathered from the analysis of IR spectra of U.S. automobile original finishes (post – 1989). This work involved the *in-situ* identification of bismuth vanadate using extended range FT-IR, Raman Spectroscopy, and X-Ray Fluorescence Spectrometry. This worker reports that Chrome Yellow ($\text{PbCrO}_4 \cdot x\text{PbSO}_4$) was a common pigment in U.S. automobile OEM finishes for more than three decades but was discontinued in the early 1990s. Bismuth Vanadate ($\text{BiVO}_4 \cdot n\text{Bi}_2\text{MoO}_6$, $n = 0-2$) was introduced in 1985 as a replacement inorganic pigment which also produces a bright hue and has excellent outdoor durability. Some differentiation of commercial formulations of this pigment is possible based on far-infrared absorptions, Raman data, and elemental analysis. This worker reports that spectral differences arise from the presence or absence of molybdenum, the use of two crystal polymorphs of BiVO_4 , and differences in pigment stabilizers. Suzuki also noted that bismuth vanadate is not used by itself, typically found with Isoindoline Yellow, hydrous ferric oxide, rutile, Isoindolinone Yellow 3R, or various combinations of these pigments.

As a follow-up to that work, Suzuki published (75) the ninth in a series of papers on IR spectra of U.S. automobile original finishes (1998–2000). This report was focused on the identification of bismuth oxychloride (BiOCl) and silver/white mica pearlescent pigments, also using extended range FT-IR, XRF spectrometry, and SEM/EDS analysis. Suzuki reports that BiOCl was the first viable synthetic pearl pigment developed 50 years ago. It was only

used for a limited time period in automotive paint (model years 1998–2000), serving to produce luster for a single Chrysler black metallic color. Silver/white micas are primarily used in white pearl tri-coat systems. This article describes the identification of bismuth oxychloride and silver/white mica pearlescent pigments in automotive finishes using FT-IR spectroscopy, X-ray fluorescence (XRF) spectrometry, and SEM/EDS analysis. Data for some cadmium pigments, which were used in automotive paint several decades ago, are also presented as they produce infrared absorptions similar to that of bismuth oxychloride.

Zięba-Palus and Michalska reported (76) on the characterization of blue pigments used in automotive paints by Raman Spectroscopy. Sixty-six blue automotive paint samples (26 solid and 40 metallic) were examined in this study. The majority of the collected Raman spectra provided information about the pigments present. However, fluorescence precluded pigment identification in some cases. Laser excitation at longer wavelengths or pretreatment to effect photobleaching often resulted in reduced fluorescence, particularly for solid color samples, and allowed pigment identification. Pairwise comparisons resulted in 97% and 99% discrimination for solid paints and metallic paints, respectively. These workers followed this study with a second study on the use of photobleaching (77) to reduce the fluorescence background in Raman spectra of automotive paints. The method was applied to group of 20 blue solid and metallic paints. The process of bleaching was studied in detail based on two samples. According to these workers, the applied procedure satisfactorily quenched fluorescence in 90% of examined samples and made pigment identification possible.

Lambert et al reported (78) on the Raman analysis of multi-layer automotive paints focusing on measurement variability and depth profile. A microtome thin section analysis without sample preparation was used to evaluate an experimental design 'fractional full factorial' with seven factors, for a total of 32 experiments representing 160 measurements. Chemometric treatments (PCA) were applied to the resulting spectra and the findings suggest the importance of sample preparation, or more specifically, the surface roughness, on the variability of the measurements on the same sample. Moreover, the depth profile experiment highlighted the influence of the refractive index of the upper layer (clearcoat) when measuring through a transparent layer.

Palenik and Palenik reported (79) on some practical microscopy methods for pigment analyses. These workers used examples of pigments in paint, fibers and cosmetics to demonstrate practical sample preparation and imaging methods that permit detailed visualization and utilization of pigments as evidence in forensic and industrial examinations. These workers used smears, cross sections and the more sophisticated ion-polished cross sections sample preparations. For imaging, they used techniques to appreciate from millimeters to nanometers, which included polarized (PLM) and oil immersion light microscopy as well as scanning (SEM) and transmission electron microscopy (TEM). These workers demonstrate the ability to find true differences in the finest components of materials, which may be suggestive of a specific manufacturer, batch difference or quality issue. Finally, the resulting images provided a simple and visually compelling means by which to convey such similarities or differences to a lay audience or jury.

La Nasaa et al reported (80) on the effects of acetic acid vapor on the aging of alkyd paint layers in artwork including characterizing the acid degradation processes involved. VOCs deriving from wooden frames and museum furniture consist of several aldehydes, formic acid and a high abundance of acetic acid. The aim of this study was to evaluate the interactions between alkyd paints and acetic acid that take place during the curing process of the paint layers. A set of reference Winsor & Newton alkyd paint layers was exposed to acetic acid vapor for six months to model these interactions. In order to evaluate the main degradation pathways occurring during the artificial aging, a multi-analytical approach based

on chromatographic and spectroscopic techniques was used. The results described the main degradation processes of the organic and inorganic components used in the production of the alkyd resin paint.

Chaplin and Clark reported (81) the use of Raman microscopy of anachronistic pigments on a purported Chagall nude for art conservation. A painting attributed to the artist Marc Chagall was examined using Raman microscopy to determine authenticity. The presence of phthalocyanine pigments precludes the painting from being created prior to c.1938 hence allowing for a declaration of forgery without any doubt and resulting in the destruction of the painting, according to French Law.

Bower et al reported (82) on the determination of the age of 20th-century oil-binder ink prints using ATR FT-IR to study a postage stamp case. For this study, samples with known origination dates were used to calibrate the drying of oil binders in inks and paints. Py-GC-MS was also used as a validation technique. The age determination calibration was applied to a stamp to determine possible philatelic counterfeits from a World War II Jewish Ghetto in Occupied Poland, obtaining a date of 1946 ± 6 (1 s, n=9) for the genuine stamps, and 1963 ± 16 (1 s, n=19) for the various reproductions.

Thoonen et al reported (83) on the use of optical microscopy for automotive paint analysis. Color and texture information was extracted from a microscopic image of a recovered paint sample and this information was compared with the same features for a database of paint types, resulting in a shortlist of candidate paints. A test database was used and two retrieval experiments were performed with the results presented in the publication.

Zhang et al reported (84) on the use of optical coherence tomography (OCT) to obtain high-resolution and cross-sectional images of the automotive paints in a non-destructive, and high-speed manner. Eight (8) automotive paint samples of different brands were examined and the images of multi-layer structures provided by the OCT system with 5 μm depth resolution were consistent with those by SEM. Structural features from the images using peak analysis and optical attenuation fit was also used to distinguish samples. The important parameters identified were optical path length (OPL) of base coat, the optical attenuation coefficient (OAC) of base coat, the OPL of clear coat, the back-scattering ratio (BSR) of clear coat and base coat, the OPL of primer surfacer, and the BSR of base coat and primer. The authors also report the ability to conduct 3-D imaging using this technique.

Groves and Palenik reported (85) on the evaluation of a one-part blue light-curing acrylic resin for embedding trace evidence prior to the preparation of thin sections with a microtome. The results of this study show that blue light-curing acrylic resins provide the desired properties of an embedding medium, generate high-quality thin sections, and can significantly simplify the preparation of paint chips, fibers and a multitude of other types of microscopic samples in the forensic trace evidence laboratory.

Buzini and Suzuki reported (86) a review article on forensic applications of Raman spectroscopy for the in situ analyses of pigments and dyes in ink and paint evidence. A comprehensive review of the forensic applications of Raman spectroscopy for the characterization, differentiation, comparison, and identification of trace evidence and questioned documents, consisting of paint and ink, respectively, was presented.

Germinario et al reported (87) on the chemical characterisation of a large number of spray paints using Py/GC-MS, FTIR, and μ -Raman. Some pigments and extenders could be efficiently identified by examination of the FTIR spectra and pyrolysis products. However, for most samples, μ -Raman spectroscopy investigation was required in addition to the these

techniques, in order to achieve the complete chemical characterization of organic and inorganic pigments, extenders and fillers.

7. Paints and Coatings Transfer, Persistence and Databases

Muehlethaler et al reported (88) the results of an extensive collaborative survey study on batch-to-batch variation in spray paints. The survey was performed as a collaborative project of the ENFSI Paint and Glass Working Group (EPG) and involved 11 laboratories. Analysis of batches from different color groups (white, orange, red and black) with a wide range of analytical techniques revealed that batch samples are more likely to be differentiated since their pigment composition is more complex (pigment mixtures, added pigments) and therefore subject to variations. The techniques aimed at color/pigment(s) characterization (optical microscopy, microspectrophotometry (MSP), Raman spectroscopy) provided better discrimination than techniques aimed at the organic (binder) or inorganic composition FTIR or SEM-EDS and XRF. White samples contained TiO₂ as a pigment and the main differentiation was based on the binder composition (C-H stretches) detected either by FTIR or Raman. The inorganic composition provided some discrimination. The discrimination of samples when data was interpreted visually as compared to statistically using principal component analysis (PCA) yielded very similar results but the statistical data can be applied for interrogating large data sets and provides for more objective criteria for decision making.

Muehlethaler et al also reported (89) on the influence of the shaking time on the forensic analysis of FTIR and Raman spectra of spray paints. Infrared and Raman spectra were collected to study the homogeneity of the paint distribution after shaking a spray can for times of 0, 1, 2, 3, 4 and 5 min. Not surprisingly, the results confirm that differences arise in both the spectroscopic techniques used in this study. The authors do report that PCA of the replicates show that the spectra are reproducible after 3 min of shaking.

Lavine and Sandercock reported (90) on improving the PDQ database search strategies to enhance investigative lead information for automotive paints. These workers applied "low level" data fusion techniques to combine and extract information based on class membership information is extracted. Search prefilters were developed to determine the assembly plant of the vehicle from which an unknown paint sample originated. The development of search prefilters for the PDQ database to exploit multiple sources of IR data was needed to extract investigative lead information from clear coat and primer paint layer smears.

Schnegg et al reported (91) on the determination of the paint coatings of motorcycle helmets. Twenty-seven (27) helmet coatings from 15 different brands and 22 models were considered. One sample per helmet was collected and observed using optical microscopy and FTIR (7 replicate measurements per layer were carried out to study the variability of each coating system). PCA and Hierarchical Cluster Analysis (HCA) were also performed on the infrared spectra of the clearcoats and basecoats of the data set. The most common systems were composed of two or three layers, consistently involving a clearcoat and basecoat. The coating systems of helmets with composite shells systematically contained a minimum of three layers. Acrylic urethane and alkyd urethane were the most frequent binders used for clearcoats and basecoats. More than 95% of the coatings were differentiated just based on microscopic examinations. The chemical and physical characteristics of the coatings allowed the differentiation of all but one pair of helmets of the same brand, model and color. Chemometrics (PCA and HCA) corroborated classification based on visual comparisons of the spectra.

Olderiks et al reported (92) on the potential for the recovery of spray paint traces from clothing by beating the garment with a plastic rod. The efficiency of the method was evaluated by spray tests with fluorescent paint and the results show that beating is an efficient way to recover and concentrate paint particles but appeared to be less satisfactory for smooth woven fabric. Application of the method in casework was effective for graffiti paints as well as for flaked car paint.

Jost et al reported (93) a preliminary study on the weathering and aging of spray paints using optical, FTIR and Raman measurements. Six different spray paints were exposed to outdoor UV-radiation for a total period of three months and both FTIR and Raman measurements were taken systematically during this time. FTIR degradation curves were plotted using the photo-oxidation index (POI), and could be successfully approximated with a logarithmic fitting ($R^2 > 0.8$). The degradation can appear after the first few days of exposure and be important until 2 months, where it stabilizes. Raman results suggest that the pigments are much more stable and do not show any sign of degradation over the 2 months.

Jackson et al reported (94) on the results of surveys of vehicle color frequency and the potential for transfer of vehicle paints to stationary objects in Sydney, Australia. Two surveys investigated (i) the frequency of the color of vehicles observed on both a motorway and suburban roads in Western Sydney and (ii) the frequency of different vehicle paint colors transferred to car park pillars and walls from five different car parks within North West Sydney, Australia. The highest frequency of vehicle colors recorded was white, grey, black and blue. The four most commonly observed colors from the five car parks were blue, white, red, and silver.

8. Paints and Coatings Interpretation

Schossler et al reported (95) on an authenticity case study in Brazil involving important Brazilian and European artists such as Candido Portinari, Juan Gris, Camille Pissarro, and Umberto Boccioni, among others. In this investigation, modern synthetic painting materials were identified in all the ground layers of the suspected paintings. The use of diverse instrumental analytical techniques such as FTIR, PLM and PyGC-MS enabled this characterization. The results demonstrated the presence of titanium dioxide, calcium carbonate and kaolin as inorganic components of the paints, and polyvinyl acetate copolymerized with vinyl versatates or diisobutylphtalate as binding media in the ground layers of the paintings and used as chronological markers.

Direny has applied for a patent (96) for the use of microtagging automobile paint samples to identify and track the automobiles. These microscopic microtag particles are mixed into the vehicle paint and contain unique alphanumeric code sequences. If and when the microtag particles are recovered from a crime scene, a simple UV light test and a magnification instrument can be used to identify the unique alphanumeric code sequence within the microtag particles.

Michalska et al have reported (97) the application of a likelihood ratio approach for solving a comparison problem of Raman spectra recorded for blue automotive paints. The proposed LR models delivered low false positive and false negative rates ($< 10\%$), and the ECE plots confirmed that their performance was much better than visual comparison.

Lambert et al have reported (98) on combining spectroscopic data using multiblock technique as chemometric tool for differentiating paints after analysis with molecular spectroscopy tools. The concept of Multiblock, as a chemometric tool, is to combine data

from several different analytical techniques in order to visualize most of the information at once. IR and Raman spectroscopy were considered as “blocks” of data of the same dataset. One algorithm called common component and specific weight analysis (CCSWA) has been used in order to produce independent PCAs for each block, and the combined (common) information in a score plot. The results showed group patterns of the analyzed paints, related to both binder and pigment compositions in one single score plot.

Muehlethaler et al also reported (99) on the evaluation of FTIR analyses using a likelihood ratio approach for spray paint examinations. A continuous approach was developed to determine a likelihood ratio with the similarity measure of infrared spectra of spray paints based on distributions of sub-populations given by the color and composition of spray paint cans. The analysis takes into account the rarity of paint composition and also the “quality” of the analytical match.

Lavine et al reported (100) on the evidential significance of automotive paint trace evidence using a pattern recognition based infrared library search engine for the Paint Data Query Forensic Database. Search prefilters were developed from 1181 automotive paint systems spanning 3 manufacturers: General Motors, Chrysler, and Ford. The best match between each unknown and the spectra in the hit list generated by the search prefilters was identified using a cross-correlation library search algorithm that performed both a forward and backward search. The results obtained using the commercial library search algorithms for the top twenty hits were always greater than 99%.

Muehlethaler et al reported (101) on the evaluation of FTIR spectra for the determination of likelihood ratios (LRs) to evaluate spray paints. An example of a practical case is described.

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Criminalistics

Fibers and textiles, 2013-2016

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1 Introduction

This review is following the previous one produced by *Palmer* [1] in 2013. It catalogues relevant literature about research and development in the field of forensic examination of fibres and textiles between June 2013 and the end of June 2016. In addition it mentions research and other activities reported by the proceedings of the meetings of the European Textile and Hair Group (ETHG) of the European Network of Forensic Science Institutes (ENFSI) during the same period. It also contains references from other sources.

2 General

Year after year the ETHG chairperson pointed out the decreasing number of volunteers for presentations during the meetings. Surprisingly, the number of participants was constantly increasing. Considering that each participant – a forensic fibre practitioner - is at least able to share casework experience, this may raise several questions:

- is there a lack of time to prepare presentations?
- is there a lack of time for research?
- is there a lack of confidence/appreciation in sharing their own work?

The first action taken by the ETHG committee in 2008 was to encourage discussion within small groups by systematically organizing 'Bring your own case' sessions during the meetings. Each participant was invited to bring either a case example or an analytical question or some interesting findings. A committee member was assigned to each small group and was in charge to report major points of discussion in front of the whole audience at the end of the session. These sessions were very appreciated among participants.

Another, more recent, action taken by the ETHG committee was to set-up an 'Advanced Training Workshop' every two years instead of a regular meeting. This kind of training programme was a specific request from the participants due to a lack of technical and/or industrial presentations during the meetings. Two major topics were especially requested:

- textile production/finishing: back to school for one day in 2014 at the Faculty of Textile and Clothing Technology (Niederrhein University of Applied Science, Germany) with various courses about fibres, textile production and finishing and textile properties testing.

- microspectrophotometry (MSP): back to the basics of the MSP technique in 2016 and on the influence of measurements parameters for optimizing the spectral quality.

Besides these training workshops, technical lecturers (Dystar, Procter & Gamble) were also invited to present interesting data for forensic practitioners during regular meetings.

Attending more general meetings such as the annual one of the European Association of Forensic Science (EAFS) was also interesting to gather innovating research on forensic fibre examination. Indeed, forensic research and development is not the prerogative of forensic science institutes anymore and is now popular and widespread in universities. For example, several UK universities developed research on forensic fibre analysis or persistence since the closure of the Forensic Science Services (FSS) which was well-known for its European leadership in forensic research. Sadly, 2015 also saw the closure of 'Contact Traces', another UK laboratory specialized in microtrace evidence.

Funding forensic fibre research remains thus an important mean to maintain the use and the development of this type of evidence as well as other non-profitable evidence (paint, glass, ...). In these times of austerity, isolated countries may encounter difficulties in obtaining funding for research. In Europe, a solution was already found in networking (through the ENFSI) on common research interests and in applying for European funding. A recent benefit of these subsidies was the set-up of a 'Reference Fibre Database' containing the microscopic and spectroscopic data of textile fibres available on the market.

3 Case Reports

Jochem [2] presented the case of a woman found in her apartment with multiple stabbing wounds. The knife was lying beside the victim. Fibre traces were recovered with a 1:1 taping on the victim and with tape lifts on seats in the living room. The suspect explained that she found the victim dead but accused her brother of the murder. The suspect's garments among which pink gloves were seized. Hundreds of matching pink polyester fibres were observed on the 1:1 taping and also about one hundred on one of the living room seats. This led to the conclusion that the suspect was wearing her gloves during her stay nearby the victim. In consequence, the suspect changed her declarations: 'I was eye-witness of the crime, a hooded man killed her, I tried to help and to pull away the victim from the offender'. Transfer experiments with the pink gloves to test the three following scenarios resulted in:

- a transfer of less than 10 fibres per taping in case of weak contact
- a transfer of less than 20 fibres per taping in case of intense contact
- a transfer of more than 50 fibres per taping by grabbing and pulling

The fibre findings on the 1:1 taping (around 20 fibres per taping) gave no support for the suspect's statement of 'pulling away the victim from the offender'.

Gannicliffe [3] reported a cold case from 1977. Two young women were found murdered and (partially) naked in two different isolated locations. They had disappeared earlier in the city centre about 20 km away from both crime scenes. A car was obviously used for the transportation. In 1977 biological traces and fibres were retrieved from clothing, nail scrapings and head/pubic hair and were preserved pending future advances in forensic science. In the 1997-2004 period DNA analyses allowed to identify one suspect (who died in 1996) through the National DNA Database and his brother (by Y-STR DNA analysis). In 2005 Police enquiries established that the suspect's car in 1977 was a motor caravan which had been scrapped in 1992. The suspect's motor caravan was a regular van converted to be a motorhome by a specialized company and the Police traced another vehicle converted

around the same time. The fabric (seating, curtain) used to customize this vehicle were said to be 'identical' to those of the suspect's scrapped motor caravan (by witnessing of its 4 successive new owners between 1977 and 1992). Two types of brown viscose fibres (seating fabric) and one type of yellow printed viscose fibres (curtain fabric) were analysed as reference material. Those types of fibres were searched on the tapings made on one victim's coat in 1977. About fifty brown viscose and 5 yellow printed viscose were found corresponding (microscopy/MSP-Vis/TLC). In 2007 the suspect was acquitted in Court arguing sex with the victims was consensual and accusing his dead brother for the murders. After the 'Double Jeopardy' Act in 2011 the offence was re-prosecuted and forensic examiners were asked to find new evidence. Additional DNA work on victim's ligatures detected profiles corresponding to both suspect's. A limited number of target viscose fibres was additionally recovered from tapings on one victim's underwear, on remains of nail scrapings after DNA analysis and in DNA extraction tubes. However, differences in the UV region between reference material and traces were observed in new analyses with MSP UV-Vis. The fibre examiner concluded that the MSP differences (batch variation or effect of sunlight) could not necessarily exclude these viscose fibres as having come from the suspect's car. This cold case is perfectly illustrating the importance of systematic trace recovery (tapings) at the crime scene and to preserve any packing/object involved in the evidence analyses.

De Wael et al. [4] made an extensive review of fibre examination of 1:1 taping illustrated by murder case examples. In ten years of use (2002-2012) in Belgium 36 cases concerning 39 victims were treated among which 23 cases started in an investigative way (no comparison material available) and the remaining ones directly in a comparative way. Investigative cases often led to the highlight of fibre collectives whose possible source material was subsequently seized during house search, except in only two cases where no collectives were found on the 1:1 taping. In case of comparison (investigative and comparative cases) 75% of cases had a 'positive outcome' (correspondence found with a reference material). This review also reported plenty of advantages for the use of the 1:1 technique: the ease of visualizing the fibre distribution (target fibre mapping for reporting, witnessing in court), the ease of examination (less background), the detection of fibre type combination, the detection of secondary transfer and last but not least the verification of modus operandi. Indeed the most fundamental advantage is the possibility to discriminate between crime related contact and legitimate contacts, in cases where suspect and victim were acquaintances. The major drawbacks of the 1:1 taping were identified as being the extensive efforts that have to be made both at the crime scene (time-consuming recovery) and in the fibre lab (if the whole 1:1 taping had to be examined). Several points of interest were finally discussed about the frequently found target fibres, the extending of the 1:1 taping around the victim or the use of a semi 1:1 taping and the usefulness in case of wet, soaked or dirt covered victims.

4 Textile / Fibre Damage

Was-Gubala [5] reviewed different types of damage or degradation of textiles and fibres:

- mechanical damage (stabbing and shooting incidents);
- thermal damage (arson, hit-and-run, terrorist cases);
- environmental and laundering effects (important for comparisons with control samples);
- changes caused by micro-organisms (exhumation cases).

4.1 Mechanical damage

Wells et al. [6] examined the effect of laundering of garments on the severance caused by sharp force impact. A kitchen knife and a Phillips screwdriver were used to stab twill weaves (jeans fabrics) and single jersey knits (t-shirt fabrics). The fresh damage was documented by

photography before and after one washing cycle. The appearance of the fabrics was different after laundering.

Kemp et al. [7] examined the severance morphology after stabbing new and laundered fabrics after 6 and 60 cycles (jeans and t-shirt) with a kitchen knife. Laundering did not significantly alter the severance morphology at low power magnifications. The variability of fibre ends viewed with SEM was higher in degraded fabrics.

Cowper et al. [8] studied the stabbing variables that affect severance appearance. No significant effect was seen for fabric extension during stabbing. Severance length was affected by several parameters such as the participant gender, fabric type laundering (age of fabric) and knife type. The severance appearance was found to be highly dependent on the participant's stabbing and knife withdrawal technique. Similar observations were made for severances caused by a trained sharp-weapon user [9].

Wightman et al. [10] carried out a study about damage caused by air weapon pellets to clothing and underlying tissue. These authors used an air rifle with 4 types of pellets (pointed, hollow point, flat and round) and investigated the effect of different types of garments covering ballistic gel (skin simulant). The differences in pellet forms were reflected in the damage to the textiles. A denim jacket effectively stopped pellets shot at 9 metres. At 18 metres only one out of five pellets penetrated the gel.

Carr et al. [11] investigated the damage caused by hand-gun bullets to clothing and the underlying tissue. Different types of ammunition induced other damage. Soft point flat nose Remington ammunition caused stellate fabric damage and little fibres with mushroom endings, while full metal jacket ammunition resulted in punch-out damage and mushrooming being more common. The entry wound size for the 2 types of ammunition was similar, while the exit wound was much larger for the Remington ammunition. Apparel layers did not change the amount of bony debris but did have an effect on the size of the wound.

Cail et al. [12] investigated the damage caused by shotgun shells with no. 8 lead pellets fired with a standard 12-gauge shotgun from several distances (37, 41, 46 and 50 m). More pellets penetrated the ballistic gel at shorter distances. Only sweatshirt and denim were able to stop all pellets at 50 m.

A laboratory test method was developed to recreate knicker ripping [13]. Laundering (once a week for a year) did not affect the force or energy required to initiate tearing. The method allowed to measure the force required to rip the thongs at different test speeds. Test speed affected the measured mechanical properties and the amount of damage. This may allow to comment on the level of applied force required to rip knickers off an alleged victim.

4.2 Degradation of textile

Geisenberger et al. [14] reported on a yellow discoloration of garments of blunt trauma victims. This yellow staining was pronounced in light-coloured textiles. These stains were suspected to originate from body fat coming from contused adipose tissue and this was confirmed using GC-MS.

Lowe et al. [15] examined the effect of soil texture on the degradation of a cotton t-shirt and cotton/polyester briefs without and in contact with pig carcasses. The results indicated that cotton fabrics in contact with a decomposing body will be preserved longer when compared to the same textile buried in soil, but not in contact with a decomposing body. The soil texture had no apparent impact on the degree of degradation. The cotton/polyester fabric was still preserved after 14 months burial, regardless of soil texture or contact with remains.

Ueland et al. [16] studied the degradation of cotton garments on a decaying body. Their experiments with clothed pig carcasses deposited on a soil surface for up to one year indicated that the decomposition fluids delayed textile degradation. Whereas cotton garments not associated with remains degraded markedly, the cotton samples exposed to the decomposition fluids remained relatively intact.

5 Significance of Evidence

5.1 Transfer and persistence studies

Roux and Robertson [17] reviewed the different factors affecting fibre transfer and its mechanism and highlighted that reconstructing experiments of an alleged incident will rarely be simple and will need a lot of information.

A significant number of fibres from 50 up to more than 1500 were transferred onto knife blades after the simulation of a single stabbing [18]. The highest density of fibres was generally found at the limit of penetration and in the cutting edge areas. Simulating consecutive stab events showed that fibres originating from the first damaged garment were still recovered from the blade. In simulations where two garments were stabbed in sequence, fibres of both garments were recovered from the blade. However, the amount of each type of fibres depended more on the garment itself (i.e., shedding and textile structure) than on the order of the consecutive stabbings. Conversely, the presence of fibre traces (from the first garment involved) near and/or inside the second stab damage (or inside injuries) could be an important indicator of the stabbing sequence.

The significance of fibre traces on buried bodies was investigated [19]. Fluorescent wool and cotton fibres were transferred onto the skin of porcine carcasses that were subsequently placed in four burial sites and left underground for 14 days. The excavation process was initiated using a stratigraphic approach and the carcasses were carefully brushed due to soil that had adhered to the porcine skin. This routine would also need to be carried out at crime scenes for fibre recovery. No total loss of fibres was observed even if persistence was low: less than 5% of cotton fibres and around 10-15% of wool remained. On one hand, decomposition products acting like glue may have increased the adherence of fibres and on the other hand, brushing may have been responsible for a higher loss, especially for smaller and more volatile cotton fibres.

Hong et al. [20] compared fibre persistence on the hands of living subjects that had washed their hands with standing water and with running tap water and dried them with a towel. The washing never showed a total loss (persistence around 5%), but surprisingly, no significant difference was noticed between the use of standing water or running tap water.

Fibre persistence on immersed garments was studied regarding several factors of influence. The influence of the recipient garment [21] was first tested during an immersion/emersion process in standing water, as the common step to all casework involving underwater conditions. A smooth garment led to weak persistence of around 20%, while higher values of 80-90% were obtained for various garments (cotton T-shirts, fleece and acrylic pullovers) offering more structure and texture. The amount of protruding fibres and the density of the rough fibrous network at the surface of the recipient garment were identified as key factors for increasing persistence. Afterwards, the influence of the water flow and the stay (from 1h up to 7h) in running water [22] was studied using a gentle water flow (running tap water in laboratory) and a medium water flow (2 m³/s) waterway (including boat activity). A gentle water flow slightly affected the fibre persistence which remained more or less constant over

time. No rapid loss was observed when increasing the water flow and the fibre persistence linearly decreased over hours. The loss of fibres during the immersion step was highlighted as an important factor that increased when using a higher water flow. Moreover, the gentle deposition method used in this study undervalued the possible loss in real casework.

Another study [23] compared wool, acrylic, cotton and polyester fibre persistence following submersion in both standing water (reserve pond) and flowing water (river with 2 m³/s mean flow rate) for 2 hours, 48 hours and 1 week. The different fibre types depicted a similar behaviour. A greater initial loss (after 2 hours) of fibres was observed in flowing water and fibres were still found after one week in both water environments.

All these fibre persistence studies suffered from a high range of variability in persistence values which is often the case in persistence studies, especially those involving real actions or real environments.

Another aspect concerning underwater environments is the possible intake of extraneous traces from water onto victim, but no study exists at the moment. However, a recent work [24] analysed microfibrils in marine sediments using a forensic science approach. Indeed, fibres were reported as a large proportion of the microplastics recovered from sediment, ice and subsurface waters. The material type found in greatest proportions was polyester. Most fibres had unique combinations of characteristics (colour, material type, cross-sectional shape, etc.). From a forensic point of view, this may suggest that no fibre collectives could be brought by water onto a victim.

5.2 Other fibre studies

Two target fibre studies have been performed. In the first one [25], two target fibre types – a black acrylic fibre and a teal coloured cashmere fibre – were compared to unknown fibres tape lifted from dressing rooms in three local clothing stores. No correspondence to the black acrylic target was found using light microscopy but the comparison produced two potential matches with the teal cashmere target which were further eliminated using microspectrophotometry. In the second one [26], *Palmer et al.* studied the random prevalence of two commonly encountered synthetic fibre types: black acrylic and blue polyester. Surface debris tapings were collected on bus seats, pub seats and cinema seats. No matches were found with either of the target fibres using high power comparison microscopy and UV-Vis microspectrophotometry. These findings showed that the probability of an ‘adventitious’ match with a particular fibre type/colour combination is extremely low and that current techniques employed by operational forensic laboratories are fit for purpose.

A discrimination study of black and dark coloured fleece garments was performed using common forensic instrumental methods [27]. Almost all fleece fabrics were solely composed of polyester fibres among which the most encountered cross-section type was polygonal. In some cases the sheddability tests also revealed a few thicker fibres originating from the inner ‘base layer’ of the fleece fabric and thus potentially providing a second fibre type transferred during crime related contacts. Most of the black fleece fabrics could be differentiated and the discriminatory power was 0.9985. However, most of the absorption spectra showed similar features in the visible range, typically two absorption bands at 450 nm and 600 nm.

6 Evidence Collection

The new edition of the Encyclopedia of Forensic Sciences [28] reviewed various well-known methods for fibre recovery and discussed how to prevent issues in terms of choice of recovery method, documentation, packaging and contamination.

The recovery efficiency of tape lifting using different tapes was recently studied [29] together with the ability of the fibre examiner to detect selected target fibres on tape lifts with a various background of other fibres. All tapes recovered more than 90% of the target fibre traces insensitively to the donor material, to the recipient material and to the strength of the adhesive tape. Concerning scanning tapes for target fibres, there was no indication that some examiners consistently performed better or worse than others. Difficulties were observed for target fibres with pale colour which were overcome by using various illumination modes. In case of difficult background on the tape lifts examiners selected a higher number of false-positives. This required extra-work during later comparison but without any influence on the eventual conclusion.

Whilst tape lifting remains the recommended method for fibre recovery, *van der Weerd* [30] presented the use of stubs for combined fibres and DNA recovery on garments. This method allowed to properly collect DNA traces in localised area on the questioned garment but was not optimal for fibre recovery. Furthermore, the stub should ideally be inspected first to prevent any loss of fibre traces during DNA isolation steps.

Another unconventional method using polystyrene rods was investigated for fibre recovery [31]. Indeed, the use of tape lifts on paper, plastic bags or any items with possible fingerprints can be problematic and fibre recovery using forceps under a low power microscope may be considered time consuming. The average recovery rate from all tested substrates was at least 99% for various natural and man-made fibre types.

Bowen et al. [32] proposed a new procedure for removing very small particles adhering to trilobal nylon carpet fibres and preparing them for SEM/EDS analysis. Observing similar particles would increase the probative value of a correspondence between questioned and known fibres. However, this procedure needed to maintain stringent anticontamination precautions at each analysis step and to use control blanks. Also, the procedure is not compatible with the use of tape lifting as a recovery method [33,34].

7 Instrumental Methods

In forensic fibre comparisons, a high degree of discrimination is obtained, using the combination of traditional methods; these include:

- Microscopy (bright field, polarization and fluorescence)
- Microspectrophotometry (MSP in the visible and UV range)
- Infrared spectroscopy (FTIR)
- Thin layer chromatography (TLC)
- Additional information on the dyes can be obtained using:
 - Raman spectroscopy
 - High-Performance Liquid Chromatography (HPLC)

A review covering instrumental methods that can be used in fibre comparisons is provided in [35]. The authors covered microscopic, spectroscopic and chromatographic methods. These authors also investigated 7 selected sample pairs of polyester fibres with a highly similar morphology [36]. These samples were examined using a series of analytical methods of which the determination of molecular weight by gel permeation chromatography seems promising. Although the method is destructive, it can be applied on a single fibre as only a sample of 1 μg is needed. Another review focused on spectroscopic techniques [37].

7.1 Microspectrophotometry (MSP)

MSP is a well-established method in fibre comparisons. The measurement of the UV-range can be used in order to account for metameric fibre samples, which present the same morphology and absorption spectrum in the visible range but can be distinguished with their spectra in UV-range [38].

Two publications concerned the discrimination of reactively-dyed cotton fibres using MSP UV-Vis. The first one [39] examined blue and red cotton samples, while the second one [40] compared the discrimination for black, blue and red cotton samples using both MSP UV-Vis and thin layer chromatography, preceded by enzymatic digestion. Both methods were comparable for the discrimination between black cotton samples. MSP UV-Vis was better in distinguishing between blue cotton samples, while TLC led to a higher discrimination for the red samples.

Was-Gubala and *Starczak* [41] found discrimination with MSP UV-Vis and Raman for cotton and polyester fibres to be similar. Raman spectroscopy was found to enable the measurement of the major components of the dye mixture and best results were obtained using a NIR laser source. For polyester fibres, MSP UV-Vis measurements can be conducted above 310 nm. It is of limited use for slightly dyed polyester fibres. The discrimination was also reduced for cotton fibres where the contribution of minor components is four times less than that of the major dye. The same authors [42] also studied the behaviour of analogously dyed wool and polyamide fibres with MSP UV-Vis. A strong absorption of UV radiation below 320 nm ('keratin' effect) was visible in the shape of the absorption spectra of all dyed wool fibres. This effect decreased with increasing dye concentrations in the wool fibre while polyamide fibres were never affected.

Mujumdar et al. [43] performed some work on the fluorescence emission spectra of optical brighteners using MSP. They were able to distinguish washed from unwashed cotton and nylon fibres by using principle component analysis on the fluorescence spectra, while this was not possible for acrylic fibres.

Excitation-emission fluorescence studies [44,45] of dyed fibre samples showing very similar absorption spectra, were performed. The discrimination between the samples was improved by subjecting the multidimensional data (excitation-emission matrices) to a chemometric analysis.

7.2 Raman spectroscopy

A review of Raman spectroscopy in forensic science can be found in [46].

The use of polarized Raman spectroscopy was described by several authors [47–49]. For example, the polarization ratio using only one Raman band at 1614 cm^{-1} for polyester fibres provided the best discrimination between different manufacturers, while very similar ratios were obtained for fibres originating from different parts of the same garment [48]. The discrimination potential of this method for polyester fibres was 0.838.

Using Raman spectroscopy cotton and viscose fibres dyed with direct and reactive dyes were studied and discriminated [50]. Spectra obtained with different laser wavelengths were similar because mostly influenced by the dye signal. The changes were mainly connected to the intensity of the bands and, in case of lower dye concentration, to the contribution of the polymer (especially with the NIR laser).

Buzzini and *Massonet* [51] examined black, blue and red acrylic, cotton and wool fibre samples using Raman spectroscopy. They found the discriminating ability to depend on the fibre type, colour and laser wavelength. These authors [52] also compared Raman

spectroscopy with the traditional methods for fibre examination (microscopy, UV-Vis MSP, TLC). They reported that Raman spectroscopy was a good complementary method to be used after microscopy and MSP UV-Vis.

Raman spectroscopy followed by multivariate data analysis was used to analyse cotton fibres dyed using similar formulations and submitted to different aging conditions [53]. Discriminant analysis allowed to correctly classify the aged fibres versus new fibres.

Massonnet et al. [54] reported on the application of surface enhanced Raman spectroscopy (SERS) for dissolved dyes (in methanol), dyed fibres, extracted dyes and eluted dye components on TLC plates. SERS generally enhanced the Raman signal for dissolved and extracted dyes and allowed identification of components in the dye mixture for 55% of the components if only a single fibre was extracted.

7.3 Chromatography

A recent survey [55] of dye extraction publications revealed that pyridine:water (4:3) is among the most commonly cited extraction solvent. Hence, this solvent was evaluated for the extraction of dyes from 172 commercially prevalent North American textile dyes and indicated that approximately 80% of the dyestuffs (various dye classes and fibre types) were extractable. Pyridine:water could thus be considered as a 'universal' solvent for extracting unknown dyes from questioned fibres.

A micro-extraction system (microfluidic device) was developed that can be used to extract dyes from single fibres in less than 10 minutes [56].

A very promising method making use of high pressure liquid chromatography-diode array detection-mass spectrometry (HPLC-DAD-MS) to identify dyes on single fibres (of 1 mm length) was described by *Carey et al.* [57]. Although the repeatability of retention times was low, the method has the advantage of recording both absorption and mass spectra, that allow for identification of dye components. The method was validated for acid, basic, reactive, direct and disperse dye classes and is currently implemented into routine case work at the fibre laboratory.

Research was also performed by *Hoy* [58] on the development of a method using ultra high pressure liquid chromatography using both diode array detection and tandem mass spectrometry (UPLC-DAD-MS/MS). The focus of this work was on optimizing micro-extraction of 1 mm length fibres for different dye classes and developing a chromatographic method with suitable resolution and sensitivity. The dye classes consisted of acid, basic, disperse, vat, direct and reactive dyes. For the latter dye class chemical digestion was performed.

Morgan [59] issued a report on the characterization of dyes extracted from millimetre-length fibres, using UPLC and UV-Vis (DAD) and MS/MS detection.

Kato et al. [60] published results on the analysis of disperse dyes using liquid chromatography in trap mass spectrometry (LC/LIT-MSⁿ). The method allowed for reliable identification of coexisting dyes, that could not be separated by LC or detected by DAD. The authors also reported on the possibility to discriminate between dyestuffs coming from different manufacturers, based on the identification of by-products from dye synthesis.

7.4 Emerging instrumental methods

Some new instrumental methods have emerged in the forensic science literature and some of these have been applied on textile materials. While these methods are very promising, for

the time being, most of them are only applicable to larger samples and not in the analysis of single fibre fragments, except the two following references.

An interesting research was done by *van Oijen* and *van der Weerd* [61] on the spectrometric imaging of polarization colours. This emerging method aimed to obtain information about fibre morphology, colour as well as its generic class directly from fibres on tape liftings. This new method could eventually improve the effectiveness of future fibre finders.

Cochran et al. [62] reported on the direct analysis of dyed textiles using infrared matrix-assisted laser desorption electrospray ionization (IR-MALDESI). A variety of dyes belonging to several dye classes (and fibre polymers in some cases) were analysed from various fabrics with little sample preparation. Further research [63] was successively performed on single fibres and tested directly from the surface of a tape lift of the fibre with a background of extraneous fibres.

A review of the application of laser-ablation inductively-coupled plasma mass spectrometry (LA-ICP-MS) can be found in [64]. The application of laser induced breakdown spectroscopy (LIBS) in forensic science was described in [65].

Ionas et al. [66] studied the distribution of flame retardants in textile furnishing using mass spectrometry and energy dispersive micro-XRF. They found no evidence of traditional flame retardants such as polybrominated diphenyl ethers or organophosphates, but instead the elemental composition of the samples suggested the presence of aluminium trihydroxide and antimony trioxide.

7.5 Identification of fibres

A solubility test for the discrimination between acrylic and modacrylic fibres was reported [67]. Using dimethylformamide of a solvent, some acrylics cannot be discriminated. Using a mixture of this solvent and ethanol (90:10 v/v) 15 out of 16 modacrylic fibres dissolved, while none of the 43 acrylic fibre samples dissolved.

A PCR-based DNA method for the identification of satoosh fibres (fine down hair of the Tibetan antelope) was described in [68]. The method is very sensitive and can be used in investigations of illegal trade. Although guard hairs can be recognized using microscopy, no obvious morphological differences were noted between down hairs of the Tibetan antelope and those of the cashmere goat.

Paolella et al. [69] described a method to differentiate unequivocally between yak and cashmere fibres. The keratin from these animal hairs was first digested by trypsin and specific peptide markers were analysed using liquid chromatography coupled with electrospray ionisation mass spectrometry (LC/ESI-MS).

8 Quality aspects

Tridico et al. [70] reported on myths and misconceptions in the morphological identification of animal hairs. The examples in this work underlined the importance of proper training of forensic practitioners in microscopic techniques and the need for reference collections to compare samples with authenticated specimens.

Hess [71] pointed out some difficulties the participants of the ETHG collaborative exercise of 2015 encountered with the identification of vegetable fibres. The key in the discrimination of commonly used bast fibres such as flax and hemp was a correct use of the modified Herzog

test. This simple microscopic test makes use of polarization microscopy to determine the twist direction of bast fibres due to differences in the fibrillar orientation. Only 62% of the participants correctly identified flax (S-twist) and 76% correctly identified hemp (Z-twist). The modified Herzog test was revisited in [72]. The authors explained the origin and the limitations of the observed Z and S-twist effects in bast fibres.

The ETHG collaborative exercise of 2014 [73] revealed difficulties with microspectrophotometry for the measurement and interpretation of absorption spectra of fibres. The survey performed in 2015 also indicated some technical issues. Therefore, a MSP workshop was organized in 2016 to optimize instrument performance, methodology and interpretation.

Morgan [74] issued a report on statistical measures for fibre comparisons and the possibility to exchange data between different laboratories. Interlaboratory experiments were set up for UV/Vis and IR and the variability of the measurements was examined. The use of multivariate statistical treatment of data compared between the three laboratories. Although the chemometric methods provided a more objective way to compare absorption spectra of known and questioned fibres within one laboratory (classification with an accuracy of 98%), it was shown that intra-laboratory exchange of data lowered the accuracy (88%).

9 Textile Industry/New Fibres

Over the past twenty years [75], worldwide production of manufactured fibres has increased of 166% (from 25 million metric tons in 1994 to 65 million tons in 2014). In the meantime (1987-2012) [76] the domination of cotton in the world production of textile fibres has been exceeded by a spectacular increase of synthetic fibre production (+270%). This latter (14 million tons in 1987 vs. 51 million in 2012) has almost doubled the quite stable cotton production (18 million tons in 1987 vs. 26 million in 2012). Among manufactured fibres [75], polyester has maintained a substantial lead in the worldwide production which has moved from 47% in 1994 to 76% in 2014. Other synthetic fibre types (olefin, acrylic, nylon) have declined over the same period. Growth in manufactured fibre production resulted in a major shift in production from North America and Europe to Asia (occupying 86% of the world production in 2014 vs. 50% in 1994). Besides these leading fibre types, a noticeable increase [76] was also observed for certain noble/animal fibres/hairs: Mulberry silk (+170%), cashmere (+62%) and especially yak (+3500%). This huge amount (thousands of tons) of yak hair (16.5-21 μm) was not found in textile goods until these years and was probably used as a substitute for cashmere (14-19 μm). Around 25-35% of the pure cashmere products on the world market were estimated to be wrongly labelled (substantial amount of non-declared other animal hairs). Last round trials from the Cashmere and Camel Hairs Manufacturers Institute highlighted a lack of ability in correctly identifying animal hair samples (even for certified testing laboratories).

The Dystar company (a joint venture between Hoechst, Bayer and BASF dye producers) is active in denim production and dyeing since years [77]. Denim were traditionally dyed with the indigo dye whose production continued to increase over the world (60% in China and 20% in South Asia). 80% of Dystar denim products were blue indigo-dyed denim and nearly 20% were black (Sulphur Black dyes) besides which various dyestuffs classes were also used to produce other unusual denim colours. Particular shades of blue were obtained using an additional dye (e.g. a yellow sulphur dye). Special finishing was mainly washdowning or damaging/tearing effects but occasionally consisted in coating application, (local) overdyeing, spraying, ... depending on designer's request. Approximately 20% of blue denim

produced annually should differ from standard indigo denim, but it is highly dependent on fashion whose actual trend is to come back to the roots with pure indigo denim.

A recent review [78] of the academic dye research over the past 15 years was focused on new insights into substrate structure (cellulosic fibres), into dye interactions in aqueous solution and on substrates, and into dye degradation and products. For example, it was shown that the photofading mechanism of reactive dyes on cotton is caused by both visible and UV light, with visible light being the dominant factor for azo dyes and UV for phthalocyanins. Although the field of dyes is one of the oldest in chemistry, much is still left to be discovered.

The Procter & Gamble company produces detergents whose formulation needs to fulfil multiple functions [79]. Over 50 ingredients were typically used in modern formulations. Detergents may indeed cause dye leaching, fabric yellowing in the sunlight or colour bleaching. Some detergents may contain 'shading dyes' (absorb yellow and emit blue) and most contain optical brighteners (absorb UV and emit blue). Most of new white garments already include optical brighteners among which most widely used are Br15 and Br49.

Artificial suede material was investigated as the possible source of some bundles easily detected on tape-lifts [80]. 60% of the studied reference materials were polyamide fibres and another 25% was polyester. They mainly consisted of microfibres held together with polyurethane (PU) as a bonding agent. Material characterization with infrared spectroscopy was hindered by the contribution of the bonding agent, of the dyeing and of some finishing or fibre surface oiling.

General information regarding man-made fibres can be found at different internet sites [81,82] as well as general news about textile industry [83].

Kiekens [84] suggested the following aspects in the evolution of textile industry:

- New fibre types like bamboo and PLA fibres are now on the market but their success may be considered as overrated.
- Ecology plays an increasing part in the research and development of new sustainable fibre types such as those based on chitin or succinate.
- Another ecological aspect is the decreasing use of water during dyeing: using less water is effective in textile industry but dyeing without water is still on research for twenty years (e.g. CO₂ dyeing is technically possible but needs very complex equipment).
- Research also pays more and more attention to natural dyes. However, their quality and their availability are limited.
- Smart textiles stay confined in an emerging phase since the 90's.
- Research and development are extensive in Europe while Asia produces mass textiles but also more and more high quality textiles. The future of Europe is niche products associated with intensive research.

10 Knot and rope analysis

Chisnall [85] proposed a fully illustrated standardized terminology for forensic knot analysis.

11 Evidence interpretation

Essential to forensic science is a correct and balanced reporting of examination results. The ENFSI has committed itself in issuing a guideline for evaluative reporting in forensic science [86]. A discussion on the use of a verbal scale to communicate the evidential strength was reported [87]. Although this concerns forensic science as a whole, fibre examination typically lends itself to the Bayesian approach.

During ETHG meetings several fibre practitioners presented on this subject [88–90].

Palmer has identified certain knowledge gaps for the correct interpretation of more difficult cases, involving for instance fibres on wet bodies and secondary transfer of fibres via head hair. His PhD work [91] provided a great example of combining research and practical aspects of fibre examination. Use was made of Bayesian networks to improve the interpretation process and a sensitivity analysis was conducted to estimate the factors that influence the likelihood ratio.

Vooijs et al. [92] discussed a numerical source level evaluation of fibre evidence. Likelihood ratio equations were developed for four generic scenarios involving a different number of reference materials, traces and matches. A review of the existing literature showed a lack of available data making this numerical approach possible for only a few types of fibres. Moreover, most of the relevant literature was based on colour description which is rather a subjective parameter. The authors stated that a verbal statement on the evidential value is currently more appropriate than using a numerical approach for estimating the frequency of fibres.

12 The Future

Several forensic groups are working in different parts of the world on very similar research themes. In order to rationalize funding and avoid duplicating similar research, more use should be made of international and intercontinental cooperation.

In the past years a lot of research has been performed on cotton fibres, as this is a commonly encountered fibre class. The ability to discriminate cotton fibres has been improved with research on methods such as Raman, MSP-PPL and LC-DAD-MS.

The same could be done for polyester for which an extensive increase in production has been noted. In addition, this generic type is almost exclusively composed of PET and therefore research has to be focused on dye analysis.

13 Summary

During the last three years, there has been a considerable amount of research relating to the forensic fibre examination. Although a large part of research was dedicated to instrumental methods, not all of these are directly applicable to individual fibres. Other research covered target fibre, transfer & persistence and discrimination studies which translate the need of data for evidence interpretation. A guideline for evaluative reporting has been issued which provide a common approach to all forensic disciplines and particularly fibre traces.

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Forensic Chemistry

Fire investigation and debris analysis, 2013 to 2016

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1. Introduction

This review covers the studies related to fire investigation published since the 17th International Forensic Science Managers Symposium in 2013. The literature includes main forensic and fire-related journals and books. Year 2013 was also searched from June onward to complete the previous review performed by Viitala and Hyyppä [1].

Fire investigation is a complex field of forensic sciences as it includes examinations of both scene as a major component and laboratory as a minor component. Paradoxically, the number of scientific articles is much greater for the laboratory than for the scene.

The complexity of fire investigation also arises from the fact that scene examination is mostly conducted by fire investigators who do not have a formal scientific education, even though they apply the scientific method. To the contrary, laboratory examinations are conducted by forensic scientists, who do not have a strong experience in fire scene investigation, but who often have a formal background in chemistry.

The literature reflects this dichotomous situation as very little is published by fire scene investigators in specific journals, although (forensic) scientists saturate literature with articles on laboratory aspects of fire investigation in rather non-specific journals. As such, there are only a few publications covering the entire field of fire investigation, from scene to laboratory.

One reference publication that has been guiding fire investigation since 1992 is the NFPA 921 Guide for fire and explosion investigations. This guide was updated in 2014 [2]. The NFPA 1033 Standard for Professional Qualifications for Fire Investigator, a document detailing the training and qualifications necessary for a fire investigator, was also updated the same year [3]. The National Fire Protection Association also published the fourth edition of the study guide for the previously cited documents [4].

Icove, DeHaan and Haynes issued the third edition of Forensic Fire Scene Reconstruction [5]. This work describes and illustrates a new systematic approach for reconstructing fire scenes, including principles of engineering and fire modeling.

Very recently, Harvey edited a book in French on the general methodology and documentation of fire scenes, which details the scientific method, the related documentation, and the report writing [6]. It is written by different contributors, all experienced fire

investigators and laboratory examiners. It constitutes one of the rare publications on fire investigation in the French language.

2. Phenomenon of fire

The study of fire as a phenomenon is crucial to fire investigation. Usually not a research topic led by fire investigators, but rather fire engineers or chemists, the results are often valuable to better understanding the combustion properties of materials, their ignition, and the behavior of fire under different conditions.

As such, this body of literature is a capital asset to the betterment of fire investigation. This chapter has been divided into three sections: combustion studies, ignition studies and fire behavior, with the caveat that a study may extend beyond a single topic.

2.1 Combustion studies

The distribution patterns of polycyclic aromatic hydrocarbons (PAHs) and isocyanates released by the combustion of polyvinylchloride (PVC) carpet and wood products was analyzed under different fire conditions by Blomqvist et al. [7]. They found that the difference among particle sizes was not significant between underventilated and well-ventilated fires, except under oxidative pyrolysis conditions. Also, they observed that volatile PAHs were usually dominant compared to particle-bound PAHs.

Gratkowski characterized the burning of unmounted tires [8]. He used two different spatial orientations: sidewall (as in a pile stock) and on-tread (as if mounted on a car). In non-accelerated ignition, the tire is likely to be ignited if the flame is applied to the bead, not to the tread. In accelerated fire, the delay between incipient flame and fire growth was much longer for ignition of the tread as compared to ignition of the well or bead. The on-tread orientation decreases the burning time of the tire by about 50%. On the other hand, it increases the peak heat release rate by about 75%. In conclusion, the author explained that a tire in the on-tread orientation constitutes a greater hazard than in the sidewall orientation, for which it could not spread fire to another tire located more than 0.61 m away.

As part of a general study of wildland fires spreading into communities, Suzuki et al. reported the production of firebrands (hot wood embers created during a wildfire and travelling in the air, believed to be the main cause of fire spread in a wildfire) [9]. In their experiment on firebrands generated by structures, they found that more than 90% of firebrands were less than 1 g and 56% were less than 0.1 g. Other authors the effect of siding treatment on the generation and spread of firebrands [10, 11]. The authors observed lighter firebrands in certain projected areas due to cedar siding, thus confirming the influence of siding on the production of firebrands.

Votive candles are often cited as the cause of fire. Hoffman et al. studied the characterization of flaring in container-filled votive candles [Hofmann]. Among the 24 candles tested, they found that flaring occurred only with petroleum-based wax, as no flaring was observed with soy-based wax. Among the petroleum-based wax, only those exhibiting another hydroxyl-based compound led to a pool fire after 15 to 30 minutes of burning. When the liquid pool of wax was ignited, the output of the candle reached 230 W, instead of 50 W in normal burning conditions, with a significant flame above the container.

The use of a barbecue generates a large amount of carbon monoxide (CO). While this is not a problem with outside use, the wide availability of disposable barbecues has led to a change of behavior, with people unfortunately using them inside enclosures. Crewe et al. studied the production of CO and carbon dioxide (CO₂) from disposable barbecues [13].

They found that smoke (produced from the burning of accelerant shortly after ignition of the barbecue) did not correlate with CO concentration. In fact, the smoke will likely clear before the CO concentration will increase. Thus, smoke is not a valid indicator of hazard. Finally, the authors concluded that, in the enclosure used in their experiment, the production of CO and CO₂ is sufficient to incapacitate and kill users in the vicinity.

2.2 Fire behavior

Capote et al. conducted small-scale and full-scale tests on new generation of high-speed trains in Spain, in order to determine the fire behavior inside these vehicles [14]. The full-scale tests revealed that the material used in the train was good in terms of fire spread prevention. In their scenario, with a backpack on a seat as the initial source of fire, flashover conditions could not be reached, although conditions dangerous to passenger's safety were created. The position of the initial fire significantly influences the temperature reached in the car, due to the different ventilation conditions. Lee et al. conducted another study on intercity trains in South Korea [15]. A tunnel was used as a test facility into which a full-scale train car was set on fire. Heat release rate (HRR) and gas concentrations were measured. A full list of material contributing to the combustion was presented. Maximum HRR was measured at 32 MW after about 18 minutes.

Okamoto et al. proceeded to four full-scale burns of minivans, two with ignition on the outside of the vans and two in the passengers' compartment [16]. The goal of the studies was to observe fire behavior as well as to monitor temperatures and HRR. The fire spread was carefully documented. With all windows closed, the passenger compartment self-extinguished after about 20 minutes. With the windows about 20 cm open, the fire reached more than 1,000°C before it completely burned the vehicle. HRR curves reflect the burning of the different compartments of the minivan, with a maximum HRR measured at 4 MW. Finally, the authors concluded that the fuel load in the gas tank bore a significant influence on the fire behavior.

Crewe et al. conducted a full-scale fire test of a 1950 residential house, during which they monitored the temperature, amount of smoke, as well as CO, CO₂ and O₂ concentrations [17]. They observed smoke and toxic gas permeation in the entire house due to its poor state of repair and maintenance (lack of sealing between rooms). They concluded that this fire scenario could easily be fatal to sleeping occupants. Guillaume et al. conducted full-scale tests of single-bedroom apartments, during which temperatures, heat flux, opacity, and gas were analyzed [18]. The goal of the study was to determine the fire behavior in regards to the tenability of the room. The first series, a scenario with a person falling asleep in bed that catches fire, showed that smoke detectors react before tenability disappears. At the time of the alarm, the toxic and asphyxiating effects of the gases will influence tenability, while thermal effect is negligible. A paper basket fire scenario was used in the second series. While the observation about the fire alarm remained the same, the tenability was more heavily influenced by thermal effect than toxic and asphyxiating gases.

Zhang and Usmani wrote a full review of the heat transfer principles in thermal calculation of structures fires [19]. This comprehensive paper covers heat radiation through participating medium as well as thermal calculation in a post-flashover fire environment and in a pre-flashover fire environment. The review serves as a guide for students, researchers, and engineers.

St. John conducted full-scale tests of the jet flame created through the mouth of a 2-gallon container of gasoline dumped over flames [20]. The author was able to reproduce a 4-meter long flame, immediately igniting a mannequin with cotton clothing 1.3 m away.

2.3 Ignition studies

Zong et al. studied the influence of gasoline added to camphor wood on its ignition and burning [21]. Using thermogravimetric analysis, they demonstrated that the addition of gasoline lowered the necessary activation energy, thus reducing the ignition time and temperature. In addition, the mass loss rate was accelerated, particularly during the two phases of dehydration (30°C to about 150°C) and decomposition (250°C to about 400°C). Finally, they observed that there was a saturation point, above which the further addition of gasoline did not accelerate the fire further.

Juita et al. explored the roles of peroxides in the spontaneous heating and ignition of linseed oil [22]. They looked into the relationship between the production of peroxides and the variation of the degree of unsaturation. They found out that metal catalysts decompose peroxides formed during the oxidation process of linseed oil, thus resulting in higher product formation.

The propensity of lit cigarettes to ignite gasoline vapors was further investigated by Marcus and Geiman [23]. They conducted 4,500 instances of exposure of lit cigarettes to ignitable concentrations of gasoline, without one successful ignition, thus confirming older studies that a cigarette is not a suitable ignition source for gasoline vapors. Schudel et al. studied the time a lit cigarette disposed on cellulosic material takes to transition to a flaming fire [24]. Among their five tests with commercial cigarettes, four self-extinguished between 6 and 13 minutes and one developed into a flaming fire after 5 minutes. All the hand-rolled cigarettes tests led to a self-extinguishment in less than 4 minutes. In his response, Babrauskas indicated that the interpretation of Schudel et al. is too simplistic, given the conditions under which the experiments were conducted [25]. He indicated a time of no less than 22 minutes under different experimental conditions.

Urban et al. looked into the ignition of cellulose spot with hot metal particles [26]. They studied stainless steel, aluminum, brass, and copper under different configurations, clashing conductors, as well as machine friction and hot work. They concluded that the ignition mechanism was different for the larger particles than the smaller ones.

Shebeko et al. developed a method to test the safety of construction materials with respect to mechanical sparks [27]. Their set-up consisted of a reaction chamber into which a rotating disc creates the spark on the tested specimen. The combustible was transformed into a gaseous mixture by partial vacuuming and ignition was detected through a manometer. They concluded that lean mixtures were most ignitable and they obtained ignition with hydrogen and acetylene, a result in contrast to methane, petrol, and LPG for which they did not obtain ignition. Holländer et al. also studied the probability of ignition of fuel-air mixtures due to mechanical impacts between stainless steel components [28]. They determined that the composition of the steel and the energy of the impact bear influence on the source and/or probability of ignition. Based on the energy, the mode of ignition changed. Arulmoli et al. investigated the production of sparks from titanium alloy golf club [29]. Apparently, cases of vegetation ignition have been documented on golf courses in the past. Although steel club heads did not produce any sparks, titanium alloy heads produced sparks, microparticles up to 500µm in diameter burning for nearly 1 second, which allows for fuel ignition. Finney et al. tested the propensity of rifle bullets to ignite organic matter after impacting a hard surface [30]. They found out that bullets fired at a steel plate could reliably produce enough energy to cause ignition, particularly those containing steel components and those made of solid copper. During their tests, some bullet fragments exceeded 800°C. Finally, Howitt studied the suitability of hot metal fragments created from heavy mechanical equipment to ignite forest litter upon impact with rocks [31]. The author used a bulldozer driving over rocks and performed direct thermographic measurements on the temperature of the fragments created.

He concluded that such fragments do not reach the minimum requirements to ignite forest litter. As such, rock strikes from heavy equipment should not be considered as a possible cause of forest fires.

Novak and Fukuda offered a brief overview of the open neutral phenomenon, an abnormal condition for which the neutral connection is removed [32]. They explained how it affects the electrical service of a residential structure. Iwashita et al. explored leakage current preceding short circuits in PVC-insulated cables under external radiant heat exposure [33]. As such, they were able to identify the mechanism as follows: Under radiant heat exposure, PVC insulation melts and conductors often come into contact with each other, at which point the leakage current is too small to be recognized. When conductors do not come into contact, the PVC insulation is carbonized and the leakage current increases until an arcing-through-char occurs. Goodson and Green published a short review describing the danger of corrugated stainless steel tubing [34].

In his review of arc breakdown in air over very small distances at 1 atm, Babrauskas concluded that the use of the Paschen curve to calculate electrical arc formation becomes unrealistic with distances below $7\mu\text{m}$ [35]. Thus, he proposed a modified Paschen curve to correctly describe breakdown at 1 atm.

Clarke and Andrews studied the ignitability of gasoline vapors with a Taser [36]. Based on full-scale tests on a mannequin, the authors were able to determine that a Taser could easily ignite gasoline vapors in a scenario in which a suspect soaked himself with gasoline.

Goodson et al. provided a brief introduction to integrated circuits, which are found in any electronic apparatus, and can be the cause of a fire [37]. The authors presented the components of the circuit, bond wires, and fuses. They go on to hypothesize about why a bond wire would no longer be continuous, and how to test it. They concluded that microfocus X-ray technique has the advantage of being non-destructive and can be used with heavy fire destruction of integrated circuits.

Jordan provided a brief introduction to lithium secondary batteries, which can be repeatedly charged and discharged, how they work, how they are used, and their potential issues, particularly in regards to fire hazard [38]. He concluded that they are largely safe to use if the design and safety standards are met.

3. Fire scene examination

3.1 Determination of origin

The study of fire patterns is at the base of the determination of the origin of a fire. Unfortunately, very little research is conducted on this topic.

In another paper, Gorbett et al. proposed a prototype method for determining the area of origin of a fire, a seven-step process named the process for origin determination (POD) [39]:

1. Value
2. Identifying varying degrees of fire damage
3. Identifying fire patterns
4. Fire pattern generation
5. Development of hypothetical area(s) of origin
6. Tests of hypothetical area(s) of origin
7. Selection of the final area of origin hypothesis

Based on 32 different origin scenarios, they demonstrated the reliability of the POD, claiming an increase in accuracy of between 50 and 94% when participants use it. Cox proposed another model following the scientific methods a systematic means of applying fire dynamics concepts to the fire scene: the origin matrix analysis [40]. Cox identified three key factors influencing the nature and extent of fire damages as exposure duration, exposure intensity, and material properties. He proposed a four-step process for assessing damages:

1. Document fire effects
2. Quantify fire effects
3. Document fire patterns
4. Label fire patterns

Gorbett et al. proposed a general review of how fire patterns are created, and they tested whether a low heat initial release rate leaves fire patterns after full-room involvement [41]. They observed that both visible and measurable damage of the area of origin persisted even after full-room involvement. One parameter clearly influencing fire patterns is ventilation. Claflin presented some experimental studies on the effects of multiple ventilation openings on a post-flashover compartment fire [42]. He demonstrated that the fire pattern at the origin of the fire can survive flashover, however it is no longer the pattern with the most damage. This latter is, of course, close to the inflow vents, just below the neutral plane, where the largest heat flux occurred. He warned fire investigators to take into account vent openings when interpreting fire patterns. Li et al. conducted some experiments on char pattern and depth on medium-density fireboard (MDF) in post-flashover compartments [43]. They also concurred that ventilation patterns are likely to confuse fire investigators. Gorbett et al. offered a new method for the characterization of the degree of fire damage to gypsum wallboard [44].

Wheeler conducted three experiments applying arc mapping to determine the area of origin [45]. In one experiment, arc-mapping was ineffective, and in the two others it was determined to be useful. However, the author reminded the reader that origin determination should not rely solely on arc-mapping, but should consider it as one tool among many others to use in determining the origin of the fire.

Coldwell investigated oxidation patterns on vehicle occurring after the fire [46]. Based on a couple of full-scale tests, the author concluded that these are of no value to the determination of fire origin, indicating that there are no clear link between the oxide color and the fire origin.

3.2 Cause analysis

Hoffman et al. conducted a full-scale fire test on a kitchen equipped with a refrigerator, a dishwasher, and an electric clothes dryer [47]. The fire was started by a heat source external to the appliances. During the investigation phase, they found evidence of electrical activity inside the appliances. Their results supported a well-know fact: The presence of an arc fault is evidence of the apparatus being energized at the time of fire, not that the apparatus is the cause of the fire. Benfer and Gottuk conducted a comprehensive study on the analysis of electrical receptacle fires [48]. It included a thorough literature review, an experimental approach, laboratory testing of the receptacles, fire exposure testing, and distinguishing of arc from melt damages. They demonstrated that the signature of overheating due to loose connections persisted after exposure to external fire. Finally, they identified the locations of arcing within the receptacle when exposed to external fire. Wright et al. studied the presence of globules and beads on small-diameter copper conductors in relation to their energized state during fire [49]. They conducted a series of tests on energized and non-energized conductors subjected to fire. When energized, the beads resembled a fusion weld, thus

creating a sharp line of demarcation between the melted and non-melted conductor surfaces. This is used to differentiate beads from globules (melted in a non-energized state).

Wang et al. proposed a new hybrid approach of combining fuzzy set theory and fault tree analysis to investigate crude oil tank fire and explosion [50]. Eckhoff offered a brief review of boiling liquid expanding vapor explosions (BLEVEs) [51]. They reminded readers that it is not possible to forecast with reasonable certainty how much time a vessel exposed to fire will resist before undergoing a BLEVE. Cases from a few seconds to several hours were reported. Eckhoff also offered an interesting review on water vapor explosion [52], even though it is not fire-based, such explosion would likely end up on the desk of a fire investigator.

3.3 Case reports

Mehaffey et al. reported a fire on the exterior of a residential structure with no observable ignition source that was caused by an electrical failure of a clothes dryer located about 9m away from the origin [53]. The authors described an interesting but complex failure of the heating element of the clothes dryer that led the electrical current to be redirected through another circuit.

Cho et al. reported a fire in a shooting range that resulted in the deaths of 15 people [54]. By examining CCTV footage, the authors determined that the fire originated in a stack of balloons, likely caused by a stray bullet coming out of the bullet trap and igniting gunpowder residues. Tests were conducted on a polyurethane foam sound absorber to determine their combustibility when impregnated with gunpowder residues. The rapid spread of the fire caused the high number of victims.

Casson et al. reported an explosion followed by a fire that occurred on a resin-manufacturing site [55]. The cause was determined to be an undesired runaway of the polymerization of methylmethacrylate that generated a rapid vaporization of the monomer, which, in turn, entered into contact with an ignition source. The case triggered the authors to conduct a series of tests conducted to understand how the undesired polymerization was accelerated.

Laboureur et al. reported another case involving 30 tons of ammonium nitrate that exploded after a chemical storage and distribution facility caught fire [56]. They investigated the root cause of the incident and the regulations applicable to this kind of facility. As time of the writing of their article, the cause of fire was not known. However, several regulations violations were observed. Dixit et al. reported a case of another factory fire that was caused by the presence of flammable liquids [57].

3.4 Ignitable liquid residues detection

Fire investigators perform detection of ignitable liquid residues (ILR) at a fire scene before sampling. To this effect, there are different techniques that can be used.

Hogsten wrote a book on the use of accelerant detection canine (ADC) [58]. His contribution identified the variables that would assist the ADC team to reach a higher rate of ignitable liquid confirmation by the laboratory. Different recommendations were made to this effect.

Burda et al. developed a new field test kit to sample ILR from fire scenes [59]. It is constituted of a white absorbent non-woven material made of polypropylene fibers. According to the authors, this field test kit, which can be applied to porous and non-porous surfaces, proved to be much better than cotton wool. They also developed a field hand kit based on the same material. Bruno designed a new field sampling system based on porous layer open tubular (PLOT) cryoadsorption of headspace [60, 61]. The device works with

compressed air. It can be used with a hand piece or with a standoff probe. The author tested the extraction of different components, including diesel fuel in soil. For the latter, the PLOT was held at about -10°C for an extraction duration of 10 minutes. The wafer was then heated at about 60°C and eluted with 1.5ml of acetone before GC-MS analysis. Unfortunately, the results were not interpreted using ASTM standards, thus it is not possible to identify the presence of diesel fuel based on the data presented.

Even though the use of portable hydrocarbon detectors is not widely performed at scenes, Baerncopf and Anuszczyk compared their responses to the one from a GC-MS analysis [62]. They concluded that the two detectors tested showed numerous false positives, false negatives, and inconclusive results. In addition, they encountered difficulties due to the adjustment of the sensitivity of the detectors during use.

Greely published a very interesting study on the presence of ILR in pour patterns [63]. He conducted several well-designed tests aimed at determining the best location within the pour patterns to collect a sample in order to maximize ILR recovery. He concludes that sampling closer to the center of the pour patterns would allow for the maximum ILR recovery. Edges do not constitute good samples, because if ILR are present, they would already be highly weathered.

Turner and Goodpaster tested triclosan (2,4,4'-trichloro-2'-hydroxydiphenyl ether) for their efficacy at killing bacteria in soil in order to preserve ILR [64]. Using 2% triclosan in .2M solution of sodium hydroxide, soil samples were sterilized in less than 60 seconds, maintained their sterility for 77 hours and preserved gasoline residues for at least 30 days. In addition, contrary to bleach, triclosan does not produce corrosive effects on the cans. In the original study, funded by the US National Institute for Justice (NIJ), the authors also attempted to develop a container that would immediately start to extract ILR from the substrate upon collection [65]. However, their design (an activated charcoal strip placed in the container) was ineffective. Li et al. studied the volatility of gasoline [66]. Upon extraction of soil with hexane using a mere five samples, they could not detect gasoline after five days. In another brief paper, the authors studied the persistence of gasoline compounds in soil [67]. Li et al. observed that C1-, C2-, C3- and C4-alkylbenzene volatilize first. After 96 hours, they were not able to detect the presence of gasoline.

Muller et al. presented a new technique to sample ILR from hands using a charcoal strip placed on hands that are, in turn, placed in a sealed bag for an hour [68]. They demonstrated that three hours after spiking 50µl of gasoline or 10µl of diesel fuel, it was possible to identify these products using this technique.

Schwenk studied the cross-contamination between containers that were not designed for fire debris samples [69]. She found that a full chromatographic profile of gasoline could be present in all samples in less than an hour of exposure to a sample of improperly-packaged gasoline.

4. Laboratory examination

4.1 General

Krüger et al. claimed the development of a new method to extract and analyze ILR in a series of five full-scale burn tests [70]. They described sampling fire debris in a transparent plastic bag, then taking an aliquot-sized sample that is placed in 20-ml vial sealed with a septum cap and additional parafilm. Before extraction by SPME, 2ml of MilliQ water is added to the aliquot. The authors identified the presence of ILR, even though the quantity before sampling could not be controlled as it was poured before fire. They also analyzed some

swipe samples, for which they claimed some promising results. They concluded that the longer the fire burns, the less chance one has to detect ILR. Dhabbah et al. published another study using SPME-GC-MS to identify gasoline residues on carpet [71].

Visotin and Lennard reported the use of a portable GC-MS to perform direct analysis of ILR at a fire scene [72]. In their preliminary study of the device, they explained that it uses SPME as an extraction technique, can perform a full extraction and analysis in about 5 minutes, and that the field tests performed allowed them to correctly identify ILR. They concluded that the potential showed by the device could provide valuable forensic intelligence directly at the fire scene, compatible with the current push toward intelligence-led policing. Leary et al. reported the use of another portable GC-MS, also using SPME as an extraction technique, in order to perform ILR analysis on site [73]. The authors showed an analysis time of less than two minutes for gasoline.

Finally, Martin-Alberca et al. wrote a rather comprehensive review of the literature regarding ILR complete examination [74]. She highlighted standards, extraction techniques, analytical techniques, statistical tools, and new knowledge on distortion effects. Hendriske et al. published a recent guide on identifying ignitable liquid residues [75]. This guide, based on ASTM E1618, covers the classification scheme, the general production processes of ignitable liquids, analytical techniques, and interpretation of data. The main chapter is a class-by-class guide for interpreting the chromatograms with detailed explanations for each specific element of the composition of the different ignitable liquids.

4.2 Extraction techniques

ASTM standard practices were updated: E1386 (solvent extraction) in 2015 [76], E1412 (passive headspace concentration with activated charcoal) in 2016 [77], E1413 (dynamic headspace concentration) in 2013 [78], E2154 (SPME) in 2015 [79], and E2451 (preservation of ILR extracts) in 2013 [80].

Cacho et al. applied a new technique, called headspace sorptive extraction (HSSE), to fire debris samples [81]. It consists of polydimethylsiloxane stir bars being first inserted in the debris container and then thermally desorbed in the GC-MS. They optimized the extraction time to 1 hour at 50°C. The bar is then desorbed in the thermal desorption unit at 240°C for 10 minutes. The authors concluded that their technique presents the advantage of reducing the manipulation of the sample and avoiding the use of solvent.

St. Pierre et al. promoted another approach using zeolites [82]. Zeolites, which are crystalline aluminosilicate mineral structures, are found in the form of small beads. The research was aimed at the recovery of low molecular weight, polar compounds, so these beads were placed in a tea bag inside the fire debris container. Then, a solvent-extraction is used on the beads. The authors explored the best extraction parameters for the extraction and concluded that zeolites improved the recovery of low molecular weight, polar compounds compared to activated charcoal. In a further study, the authors explored a dual-mode with zeolites and activated charcoal strips in order to extract optimally the full range of ILR [83]. Their research demonstrated that, as the two extraction techniques are complementary to each other, this dual-mode is effective for extracting both low molecular oxygenated compounds and traditional heavier hydrocarbon-based compounds.

Nichols et al. compared PLOT-cryo extraction technique to Tenax and activated charcoal strip [84]. The authors concluded that it works better than the traditional purge and trap technique, and that it outperformed the traditional activated charcoal strip.

Smale et al. compared cat litter, absorbent matting, cotton pads and passive headspace residue extraction device (PHRED) to extract ILR from concrete [85]. PHRED is a device that is placed on concrete, heats it, and adsorbs ILR on a charcoal strip contained inside the device. They concluded that cat litter and PHRED were suitable to extract ILR after one hour, and the two other techniques did not reveal anything. Their chromatographic data clearly demonstrated that PHRED is the most sensitive technique.

Cheenmatchaya and Kungwankunakorn researched using rice husks as a new preparation of activated charcoal strip (which is usually made of coconut shells) [86]. They concluded that rice husks possess a high apparent surface area, and they encouraged the use of it instead of the costly commercial adsorbent.

Fettig et al. evaluated SPME as an extraction technique for fire debris samples [87]. They used a mixture of three different adsorbents, divinylbenzene/carboxen/polydimethylsiloxane. Interestingly, they prepared their fire debris in a smoke density chamber and a controlled-atmosphere cone calorimeter. They presented the best conditions for the use of their SPME fibers; SPME conditions vary according the ILR that has to be recovered, a fact that is usually unknown at time of extraction.

4.3 Analytical techniques

ASTM standard test method E1618 for ILR analysis by GC-MS was updated in 2014 [88]. Also, a new ASTM standard test method E2997 on the analysis of biodiesel byproducts by GC-MS was published in 2016 [89]. The ASTM standard test method E2881 for the extraction and derivatization of vegetable oils and fats from fire debris and liquid samples with analysis by gas chromatography-mass spectrometry was released in 2013 [90].

Martin-Alberca et al. studied the analysis of acidified fire debris samples, i.e. samples originating from Molotov cocktails in which a mixture of gasoline or diesel fuel with sulfuric acid is used [91]. Using SPME-GC-MS, they identified gasoline and diesel fuel residues from acidified fire debris. They concluded that the presence of *tert*-butylated compounds is definitely an indicator of the presence of acidified debris. Furthermore, Martín-Alberca et al. investigated the chemical modifications occurring in the Molotov cocktail [92]. Aromatic compounds are subjected to a heavy alteration and oxygenated compounds are hydrolyzed, thus significantly changing the chromatographic pattern within minutes. Martín-Alberca et al. also used Raman to analyze acidified ignitable liquids through bottle glass [93]. Despite the fluorescence due to the reaction between the acid and the hydrocarbons, the authors were able to identify both, except in the presence of diesel fuel. They concluded that Raman is a useful technique for a rapid, non-invasive analysis of pre-ignited improvised incendiary devices. Martín-Alberca et al. also studied the use of ATR-FTIR to identify neat and acidified ignitable liquids [94].

Kerr et al. investigated the use of Raman and ATR-FTIR spectroscopy to identify polymers among fire debris samples [95]. They demonstrated that both techniques are complementary and, that with this combination, it was possible to identify HDPE, LDPE, PVC, PMMA, and cotton among burned debris.

Schwartz et al. explored the use of GC-IRMS (isotope ratio mass spectrometry) to discriminate between household ignitable liquids [96]. They looked at compound-specific carbon isotope ratios. Although this works with neat liquids, the authors reported that it was problematic with post-combustion residues. They concluded that GC-IRMS is not suitable for fire debris analysis, but appeared best suited for exclusionary purposes when analyzing neat simple mixtures.

Liu et al. conducted a pilot study regarding the use of a cataluminescence-based vapor sensor array for discriminating flammable liquids [97]. Using 10 different catalytic nanoparticles, they were able to differentiate 10 flammable liquids, all single-component liquids, except for gasoline. They concluded that their system is very promising for field use, though further research is still needed.

Ferreiro-González et al. applied the headspace-mass spectrometry technique to discriminate between ignitable liquid residues [98]. Using chemometric methods, they claimed that they were able to successfully discriminate between six different ignitable liquids and six different burned substrates based on the total ion spectrum.

Finally, Sampat et al. published a general paper on the potential of two-dimensional comprehensive gas chromatography in forensic sciences, including a section on fire debris. They reviewed the papers published on the topic since 2002 [99].

4.4 Interpretation

In a quite comprehensive paper, Baerncopf and Hutches reviewed the modern challenges of interpretation in fire debris analysis [100]. They addressed challenges in liquid classification, matrix interferences, microbial degradations, and non-routine samples.

In a study funded by NIJ, Rankin and Petracho investigated the effect of competitive adsorption of substrates typically found in fire debris on the classification of ILR, and proposed developing and validating an expert system to classify ILR in fire debris and to provide a statistical evaluation of error rates [101]. While they did not find any false positives among the reports from the small group of experts tested, they admitted that larger studies would be necessary before an error rate could be determined. Likewise, their expert system needed refinements before it could be tested.

Hetzel conducted a survey of 71 gasolines collected from the United States in 2008, by analyzing them following ASTM E1618 [102]. She observed some wide variations between the samples in regards to the alkanes pattern, however nothing that would cause a fire debris analyst to incorrectly identify gasoline.

Jhaumeer-Laullooa et al. conducted a brief study of background and pyrolysis products [103]. They analyzed 11 different substrates and reported the background products found. Li et al. also performed some controlled burn tests of carpet with and without gasoline [104]. They concluded that the most encountered combustion and pyrolysis products were also found in gasoline. In his doctoral thesis, Sferopoulos conducted test burning of carpet and foam in order to identify interferences with gasoline [105]. His data presented detailed information on the compounds found during these experiments. He concluded that false identification of gasoline is unlikely. Prather et al. investigated the effect of interference from high-density polyethylene (HDPE) on ILR analysis [106]. More concerned by firefighters' health than fire investigation, Organtini et al. studied the halogenated compounds in fire debris samples [107]. Lee et al. conducted a comprehensive study of the influence of temperature on the pyrolysis products of household materials [108]. They compared isothermal and temperature-programmed pyrolysis on many different substrates, such as asphalt roofing materials, carpets, carpet underlay, vinyl flooring, and cellulosic materials. Their paper included all the different identified compounds, as well as their relative intensities according to the pyrolysis temperature. Ding et al. analyzed the pyrolysis products from a tire using GC-MS coupled to a pyrolyzer [109]. The authors showed that at 600°C, pyrolysis products were alkenes, with the main components being isoprene and *D*-limonene. Above 600°C, aromatic content started to rise. This study allowed the author to better understand the reaction pathways of pyrolysis. Zhang and Yang studied the composition of

the combustion smoke of different flammable liquids, notably gasoline, diesel fuel, and paint thinners [110]. Because of the interpretation of the chromatographic data not following ASTM standards, the meaning of the results is difficult to understand. However, the authors claimed that gasoline, diesel fuel, and paint thinner smokes were different and comprised of many components.

Goldman et al. wrote a comprehensive paper on the analysis of biodiesel by GC-MS [111]. After an introduction describing the manufacture of biodiesel, the authors performed analysis of neat samples, heated headspace samples on an activated charcoal strip, and performed studies of microbial degradation, evaporation, and interferences with matrix. They showed the different influences on the data obtained.

Turner et al. comprehensively studied the effect of sampling season and soil type on microbial degradation of gasoline from soil [112, 113]. Bacteria were identified as *Alcaligenes*, *Bacillus*, and *Flavobacterium*. The authors determined that the most vulnerable compounds were n-alkanes, followed by mono-substituted alkylbenzenes. Interestingly, benzaldehyde was identified as a marker of the extent of the degradation. Finally, they concluded that soil collected during hot and dry summer showed the least degradation of the ignitable liquid. Hutches investigated the microbial degradation of ignitable liquids on molded building materials [114]. She concluded that the degradation occurred to an extent that could prevent identification of gasoline. She explained that the microbial degradation observed with soil can also apply to moldy substrates. Winters and Evans studied the effect of mold, and of burning on firelogs [115]. Although firelogs were mostly made of paraffin wax, this has changed, and they are now made of vegetable oils. Both fire and mold affected the chemical composition of firelogs.

McGee et al. conducted a comparison of GC-MS data processing software [116]. They compared ChemStation, Xcalibur, and MS Workstation to ACD/MS Manager Suite. They concluded that ACD/MS Manager Suite constitutes a viable solution for consolidating and processing data from different sources in one standardized package.

The National Center for Forensic Science has been extremely active in conducting research project on interpretation of fire debris analysis. In a first study, Waddell et al. used principal component analysis (PCA), linear discriminant analysis (LDA), and quadratic discriminant analysis (QDA) to develop a multistep classification procedure of ILR according to ASTM E1618 [117]. As a result, the true-positive rate was more than 80% for cross-validation samples and more than 70% for fire debris samples, while the false-positive rate was 9.9% and 8.9% respectively. In sequel paper, Waddell et al. further investigated soft independent modeling of class analogy (SIMCA) classification of total ion spectra [118]. As a result, the true-positive rate was more than 90% for cross-validation samples and almost 80% for fire debris samples, while the false-positive rate was 5.1% and 8.9% respectively. Using the 445 total ion spectra of the ignitable liquids reference collection (ILRC) database, Waddell et al. explored their classification using hierarchical cluster analysis [119]. As a result, the ignitable liquids clustered based on their chemical composition. In addition, ignitable liquids within each cluster were predominantly from one ASTM E1618 class, thus showing very promising results. Lopatka et al. pursued similar research with fire debris samples, some of which included ILR [120]. The authors showed an overall classification performance of 81%. Sigman et al. included all of the research performed by NCFS, which provides a full statistical assessment of the probability of correct identification of ILR in fire debris samples [121]. In a most recent paper, Sigman and Williams compared support vector machine (SVM), LDA, QDA, and k-nearest neighbors (kNN) methods of binary classification of fire debris samples as positive or negative for ILR [122]. SVM, QDA and kNN showed good

performance, which decreased rapidly with fire debris samples. LDA provided poorer discrimination, but its performance did not deteriorate with fire debris samples.

Frisch-Daiello et al. used self-organizing feature maps (SOFMs) to extract ion spectra [123]. This allowed for the interlaboratory comparison of data without concern for retention time shifts. The authors concluded that clusters in the data were observed to be consistent with classification according to ASTM E1618. Sinkov et al. addressed issues of chromatographic alignment and variable selection that are needed prior to the application of chemometric tools [124]. The authors used an alignment strategy based on a ladder consisting of perdeuterated n-alkanes. McIlroy et al. evaluated the effect of pretreatment procedures on multivariate statistical analysis, which included background correction, smoothing, retention-time alignment, and normalization [125]. They concluded that prior to pretreatment, the first principal component accounted for only non-sample source of variance. After pretreatment, the principal component accounted for significant chemical differences among the diesel samples.

4.5 Other liquids, materials and characterizations

Ferreiro-González et al. reported that they successfully discriminated between different gasoline samples according to their RON using headspace-mass spectrometry [126]. The main advantage of this technique is that it neither requires any sample preparation, nor chromatographic separation. Vergeer et al. used likelihood ratio methods (based on distance functions for the two first and multivariate for the third one) to compare evaporated gasoline residues in order to identify source of gasoline [127]. Strong discrimination was obtained with all three methods. Haraczaja et al. used Carburane, a commercial software originally designed to evaluate petroleum fraction quality, to compare premium gasoline neat samples using GC-FID [128]. The advantage of the software is that it adapts to all analytical techniques. Ugena et al. reported the discrimination of five main brands of fuels using GC and neural networks [129]. They claim a discrimination power close to 100%. To fight smuggling across borders, da Silva et al. developed a method of classifying Brazilian and foreign gasolines adulterated with alcohol using infrared spectroscopy [130]. The authors used two different approaches: partial least squares discriminant analysis (PLS-DA) and soft independent modeling class analogy (SIMCA).

Yang et al. reported a case study in which they had to identify biodiesel spill following an environmental spill [131]. They used SPE-GC-MS in addition to an HPLC analysis to identify the compounds that could not be detected by GC-MS. In a later paper, Yang et al. further investigated the discrimination between biodiesel samples [132].

Borusiewicz reported an atypical analysis of ignitable liquids from food and other biological mediums [133]. In the first case, water contaminated with kerosene was discussed. The second case consisted of a poisoned beverage into which a medium aromatic product was found, compatible with the use of insecticides to poison the beverage. The third case involved the analysis of blood from a dead subject who inhaled gasoline fumes. The authors warned that these unusual matrices could have unusual matrix effects.

In quite a comprehensive paper, Ernst and Streeter studied glycol ethers [134]. They explained that glycol ethers, found in scented oils, can be found in fire debris samples. The authors covered the GC-MS analysis as well as a flame test. They concluded that the identification of a glycol ether in a fire debris sample may help explain the appearance of fire patterns at the scene.

Liu et al. studied dust components of gasoline combustion using HPLC [135]. Even though the authors found 20 compounds among the dust without identifying them, they concluded

that pattern recognition would be useful to identify gasoline combustion dust. Zong et al. investigated the discrimination of soot samples from three different substrates [136]. Using PCA and neural networks, the authors were able to differentiate soot from diesel, polystyrene and ABS during small-scale burns. Ayoko et al. studied the VOCs from the exhaust of LPG and gasoline-powered vehicles [137]. Thirty-three compounds were identified in a comprehensive list with concentrations. Hexane was the most prominent in both fuels.

Al-Abdullah et al. investigated the flashpoints and volatility characteristics of gasoline/diesel blends [138]. They tested mixtures with 0, 5, 10, 50, 90, 95 and 100% of gasoline in diesel. They found that flashpoint decreased sharply when gasoline was added. With 1% gasoline, it decreased from 57 to about 35°C. At 16% gasoline, it reached -40°C, which is the typical flashpoint of gasoline.

Groth et al. explored cigarette brand determination through trace-metal analysis of ash [139]. Trace analysis was performed on ashes from 14 American brands and 17 international brands using ICP-MS. Inter-brand variation was shown to be larger than intra-brand variation. Classification between US and international brands was successful, however when classification of brands was possible, in some cases a range of possible source brands was the only option. Finally, the distinction between varieties of the same brand was not possible.

Alqassim et al. studied the degradation of concrete exposed to fire using x-ray diffraction, petrographic approach, and thermogravimetric analysis (TGA) [140]. They showed that concrete underwent two main irreversible degradations, the first one between 70 and 120°C, which corresponded to the loss of water, and again between 650-700°C, which corresponded to the decarbonation of calcium carbonate. Combining different techniques, the authors indicated that the investigator should be able to determine the temperature reached by the concrete. Boniardi and Casaroli investigated the influence of heat on non-ferrous objects from a metallurgical perspective [141]. They looked at the metal structure in order to estimate the temperature range experienced by various items to estimate the temperature reached in different locations for a given fire. In a subsequent paper, the same authors provided an in-depth approach of microstructural analysis of metallic materials to fire investigations [142]. They offered a detailed review on three-multistrand conductors, aluminum window frames, and bedsprings. Gu et al. developed a new method for identifying the condition of electrical immersion heaters before fire [143]. The authors used SEM/EDX to look at the morphology and elemental composition of the electrical wires, which showed significant differences between a normal use and a misuse.

5. Fire modeling

From a safety perspective, Dadashzadeh et al. proposed a new approach to model the entire sequences involved in a potential accident in oil and gas facility, using use computational fluid dynamics (CFD) codes [144]. As such, they modeled the vapor cloud explosion and the consecutive fire. They concluded that the integrated approach provided a clear advantage over the modeling of single phenomenon.

Lai et al. conducted FDS simulations and full-scale experiments to measure the impact of natural ventilation design in Green buildings [145]. They concluded that in non-fire rooms, the FDS simulated temperatures were consistent with the full-scale tests. This is not the case in fire rooms, where the thermocouples did not provide data representative of smoke descent, due to the proximity of fire. Finally, the authors demonstrated that a room with a natural ventilation shaft provided better control of the smoke layer than a room without a natural ventilation shaft. In addition, they demonstrated that FDS simulation could

compensate for the lack of usable data from full-scale tests. Fire modeling was also used to simulate railway rolling stock fire scenarios, based on Guillaume et al.'s pyrolysis model [146]. With the model, the authors were able to reproduce fire growth, heat release rate, and temperatures observed in the real-scale scenario. Weinschenk et al. applied FDS to provide some insights into a single-family structure fire [147, 148]. They calculated temperatures in some key locations, thus explaining the downward development of the fire that ended up killing a fire captain. Overholt et al. also applied FDS to analyze the fire that occurred in another single-family residential structure that resulted in the death of two firefighters [149]. With their simulation, they were able to demonstrate that the rear basement window failure led to a steep HRR increase from 2 MW to 32 MW. The two firefighters were in the flow path when the rapid change of conditions occurred.

A prison fire that occurred in 2010 in Chili killed 81 inmates. Although the cause of the fire was determined to be intentional, the investigation could not determine how fast the fire grew and whether the prison guards acted accordingly. Jahn et al. used CFD to analyze the development of the smoke and to simulate temperatures [150]. The authors demonstrated that the fire grew so quickly that it became uncontrollable before the guards could intervene. Another question of timeline was confirmed using FDS at a murder scene in which a smoldering fire was observed [151]. Hofmann et al. concluded that the fire started a few hours before discovery, at a time where the suspect had access to the crime scene.

Price et al. explored a prototype inverse fire model (IFM) in order to predict heat release rate of a compartment fire using smoke layer information obtained from building environmental sensors [152]. However, the authors concluded that IFM-based estimations of heat release rate have an accuracy of about 40%. Finally, Overholt and Ezekoye introduced a Bayesian approach to the quantitative testing of fire scenario hypothesis through fire models [153].

6. Aspects of forensic pathology and toxicology in fire investigation

Owen et al. explored the possibility of using bladder swabs, instead of conventional samples such as blood, muscle or bone) to identify cremated victims through DNA analysis [154]. Out of the 28 cases analyzed by their laboratory in 2012, all of them provided positive DNA results through bladder swabs. As such, the authors recommended the use of bladder swabs in incinerated bodies, as it is an efficient and cost effective means of obtaining DNA.

Pahor et al. conducted a study to determine whether gasoline residues could be detected in fire victims' lung tissues and heart blood [155]. They conducted tests on pigs, which they subjected to gasoline breathing before euthanasia. They also used pigs that did not inhale gasoline, but that had gasoline poured on them. Using headspace thermal desorption GC-MS, the authors found traces of gasoline in lung tissues and heart blood from the pigs that only inhaled gasoline. In a case for which two dead persons were found in a burned car, Karinen et al. found methyl *tert*-butyl ether (MTBE) in postmortem blood and urine samples [156]. Because the autopsy and routine toxicology results did not explain the cause of death, the authors analyzed the samples for MTBE. They concluded that gasoline poisoning caused the deaths and that MTBE can be a suitable marker of gasoline exposure when other volatiles have evaporated. Using infrared imaging combined with ordinary color imaging, Yamauchi et al. demonstrated the improvement in the detection of soot particles in the respiratory and gastrointestinal system [157]. Soot particles appear black, while blood is transparent, and tissues are white.

McAllister et al. reported a case study in which they compared the results of COHb analysis to autopsy data and the two hypotheses regarding origin and cause of the fire [158]. This allowed them to determine that only one scenario produced the level of CO found in the

victims. Michiue et al. used a similar approach to reconstruct mass fire casualties, combining fire investigation data with COHb analysis of the victims [159]. Hill proposed the same approach of combining COHb data with the fire origin and cause investigation to test hypotheses [160]. Oshima et al. reported three fire cases in which they performed COHb analyses [161]. As such, they demonstrated that CO poisoning occurred prior to the fire. Finally, Ferrari and Giannuzzi proposed a novel approach to the assessment of carboxyhemoglobin, hydrogen cyanide, and methemoglobin in fire victims [162]. They performed blood analysis on 32 fire victims of a prison fire caused by a polyurethane mattress fire in Argentina in 2006. The authors found no correlation between HCN and MeHb.

Bernitz et al. studied tongue protrusion as an indicator of vital burning [163]. The authors found a statistically significant dependence between tongue protrusion and the presence of soot in the respiratory tract and stomach. Since the latter is a good indicator of vital burning, the authors concluded that tongue protrusion could be used as an additional indicator of vital burning. In a critical reply, Bohnert did not share the same opinion [164], claiming that the correlation detected by Bernitz had no relevancy. In addition to the response from Bernitz [165], the Bernitz study triggered a series of comments from Hejna and Janik [166], Madea and Doberentz [167] as well as Nikolić and Živković [168], who claimed that tongue protrusion is not a vital sign. Recently, Bohnert and Hejna published a retrospective of 61 fire fatalities for which they did not find a significantly increased incidence of tongue protrusion in vital burning [170].

Goncalves et al. investigated the estimation of the condition of human remains prior to exposure to fire [170]. They looked into the effects of age, sex, time span from death to cremation, duration, and temperature of cremation. Among their conclusions, the authors indicated that warping is most useful as an indicator of the pre-burning condition of human remains. Keough et al. conducted an assessment of skeletal changes after post-mortem exposure to fire [171]. The authors concluded that, based on the data, the pattern of heat-induced changes may assist in estimating decomposition from burned remains. Three cases of cremation on wooden pyre were reported by Alunni et al. [172]. The authors highlighted the specific thermal alterations resulting from these circumstances and reminded readers of the importance of a good collaboration between the fire investigator and the forensic pathologist.

Finally, Rossi et al. reported a suicidal fire death case where the cause of death was a highly unusual trigemino-cardiac reflex [173].

7. Human behavior

Guldåker and Hallin examined the correlation between living conditions and the occurrence of intentional fires [174]. As such, they looked at spatio-temporal distribution of fires in Malmö, Sweden. They observed that high exposure to social stress and high proportion of young males increased the risk of more frequent intentional fires. Among many other observations, they proposed different solutions in order to prevent fires. Grubb and Nobles evaluated arson occurrences in Los Angeles from 2005 to 2012 [175]. They analyzed the space-time interaction of these incidents using the Monte-Carlo simulation-based Knox method. They concluded that the results obtained may aid in efforts to investigate and prevent arson. Corcoran et al. examined malicious hoax calls and suspicious fires in Queensland, Australia [176]. They explained that these incidents are a significant burden to fire departments. As such, the authors used a local Moran temporal plot to target hotspot with finite resources. Bruenisholz et al. published a critical review of the situation regarding deliberate fires [177]. In this first of a series of three papers, the authors detailed current

knowledge on the subject and the challenges of repetitive fires. Based on successes of this application with other repetitive crimes, they developed a new approach that uses an intelligence process cycle as a framework to perform a systematic analysis of fire events. In another paper on the same subject, the authors mention intelligence-led policing as the framework for tackling the problematic of repetitive fires [178].

Harpur et al. investigated the circumstances around fatal residential fires involving children 5 years old and under [179]. They determined that the most common cause was fire-play with inadequate supervision and relaxed attitude to fire safety at home. In regards to juvenile fire-setters, Johnson et al. proposed that the forensic psychological evaluation also include an assessment of the parents [180]. This would help understand the broader context surrounding the subject.

Purser looked at two cases of fire fatalities in care homes in order to determine fire scenario development and occupant behavior [181]. He pinpointed different problems, such as doors left open and training inadequacies. He concluded with some recommendations for improving safety. On the same topic, Thompson and Wales conducted interviews of victims to determine their experiences, actions, and motivations during an accidental dwelling fire [182]. One of the key findings was the victims' desire to tackle the fire in its early stage to put it out. Xiong et al. studied the risk factors (inherent to the victim) contributing to death in accidental residential fires [183]. The top seven factors discovered were: psychotropic and sedative drug intake, discarded cigarettes, living alone, being over 70 years of age, being asleep, location in the room of fire origin at ignition, and alcohol intake.

Anderson and Janssens examined low-energy (lighter, candle, match, or some form of space heater) versus smoking materials (pipe, cigar, or cigarettes) ignition fires in the US, UK, Japan and Finland [184]. They reported that smoking materials ignition fires tend to be more fatal than low-energy ignition fires, even though the latter exhibit a larger proportion, leading to more losses and injuries. Finally, they proposed a statistical model that predicted whether a fire was ignited by a low-energy means or by smoking materials based on age and race of the victim, as well as the season of the year at which the fire took place.

Finally, Rohde et al. conducted a comprehensive review of the correlation between smoke alarms, injuries, and deaths in fire [185]. The authors reviewed the literature and pinpointed areas of future investigation. They reminded readers that the death rate in households with smoke alarms is about half of that in households without smoke alarms.

8. Diverse publications

In a two-part paper on arson investigation and science, Srutin reviewed errors that have and can occur in fire investigation [186, 187]. Ost-Prisco addressed legal issues surrounding a fire cause determination based upon negative corpus [188]. According to the author, negative corpus opinions are no longer acceptable as the sole basis for an opinion that a fire was incendiary.

Plucinski studied the timing and circumstances of vegetation fires for the Perth region of Australia [189]. The lessons learned from the trends observed by the author will help apply prevention and mitigation programs.

Carvel provided a comprehensive review of fire safety in tunnels [190].

Sullivan and Schumacher provided a description of wet chemical extinguishing systems for commercial kitchen [191]. Their comprehensive paper also comprised defects found in these

systems. Horton investigated fires in which creosote build-ups in kitchen exhausts systems create an important risk [192].

Morgan et al. studied the persistence of pollen in vehicle fires [193]. The authors found that the pollen persisted long enough to resist a vehicle fire.

Gardner et al. investigated latent fingerprint enhancement techniques on items recovered after a fire [194]. The authors determined that temperature was the most important factor in determining whether a print could be recovered or not. Where soot is present, recovery is possible until about 300 °C. No statistical differences were observed between the different techniques studied, which should be chosen on a case-by-case basis.

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Forensic Chemistry

Explosives, 2013-2016

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1. Introduction and Coverage of the Literature

This current review starts with a recommendation to read the previous two papers covering explosives analysis from 2007-2010 presented in 2010 by Richard Strobel and my previous review from 2013. [16, 7] This review is perhaps more truncated than the last two papers in that the breadth and impetus into explosives research has slowed. There have been austerity measures imposed on governmental organizations, academia, and even on private companies, but there are still “hundreds of citations listed...because of result of the ability to survey specialty journals which were previously unknown to the forensic practitioner”. [16] Additionally, war efforts have slowed significantly in the past three years and the impetus to devote resources in detection, post blast analysis, and prevention in that mode have likewise seemingly decreased. That said, the overall threat from explosives, especially in domestic settings, has increased. Several events in Europe, Asia, Middle East and in the Americas have shown that terrorists are still keen on employing explosives. If anything, the work of the forensic chemist in the area of explosive analysis will increase due both to an increase in cases and the use of novel or hard to detect explosives. The forensic explosive analyst should regularly review literature in the wider scientific community with an emphasis on suitability for employing new techniques in the scheme of analysis. These include both applied and theoretical published research. It helps to get an early start in researching these techniques because of the increasingly stringent accrediting requirements for any new technique. Even after a given technique is discovered and tested, vigorous validation and documentation is required before the technique is actually used in case work.

There are many applications in the overall field of explosives which may be of interest to the forensic analyst tasked with examining explosives for law enforcement purposes. The explosives detection field, which is primarily for security purposes, is both the fastest growing and most proliferated area from which forensics can draw. There are a host of references in this area, ranging from theoretical research to applied systems that are already in field use. Some of these papers may appear limited on the surface but are worth perusing, especially if the technique can practically be more broadly applied.[7]

There are 646 references in this review. A direct pathway to the abstracts of the articles are included via a hyperlink to the abstract, or full text article where available. Additionally, the categories in the reference list can be easily accessed using the Navigation pane in Microsoft Word. This will aid the navigation of the bibliography section, starting on page 23 of this document. Many of these references could fall into two or even three categories. They will not be presented in multiple places, so it would be advantageous for the reader to peruse all of the sections.[7]

The organization of this paper follows roughly the same pattern as the previous reviews. In many cases the reference papers could easily fit into more than one category. The placement in any given category was based on the authors' original candid assessment but could be construed to be better fitted in another.

2. Review Articles

There were again several papers in this three year cycle deemed by the authors to be most relevant if placed in the review category. Some are broad schemes of analysis, while many are reviews of a specific class of instrumentation. Still others are self-described as reviews.

Abdul-Karim et al review the aspects of post-blast explosives deposition based on known theoretical constructs.[1] It is unknown how useful this will be to crime scene professionals because many factors are simply uncontrolled in a real explosion.

Brown, Greenfield, McGrane, and Moore have written two major review papers on the advances in explosives analysis.[2,3] One focuses on photon and neutron methods and the other animal, chemical, ion, and mechanical methods. This second paper does a good job of reviewing the molecular imprinted polymers, which has seen a large increase in publications from the last three year cycle. It also delves into another emerging area, immunochemistry. Finally it tackles mass spectrometry and other ion methods.

Calcerrada and company review advances in capillary electrophoresis, including the portable and miniature systems and even microchip CE, and compare them to conventional CE systems.[4]

In Cleveland and Morris' book, *Handbook of Energy 2*, they more broadly look at improvements in engineering energetic materials and have an interesting retrospective on historically important improvements in explosives.[5]

Martin-Alberca and Garcia-Ruiz have completed a comprehensive review of the schemes of analysis for consumer fireworks and what formulations are typically found in them.[10] It provides a superb primer on contents and methods of analysis although it appears to be light on some techniques such as X-ray powder diffraction.

Fountain et al discuss recent advances in all spectroscopic detection of explosives and seek to integrate signatures into algorithms for evaluating sensor performance.[6]

Ma, Wang and Wang review nanomaterials for luminescence detection of nitroaromatics.[9] These already have shown great selectivity and the challenges now are ease of fabrication, and cost. They examined various approaches and methodologies. These types of sensors are likely to be found in commercial products and scene data relayed to the bench forensic explosives expert. The basis of this technology should be known to bench chemists.

A similar review by Shanmugaraju and Mukherjee describe fluorescent chemosensors.[15]

Zyryanov et al have an excellent review of the low mass chemosensors for nitroaromatic detection. This is perhaps the most comprehensive review of these types of sensors and explores mostly visual detection through colorimetric and photometric methods.[19]

Lefferts and Castell review vapor sensing of explosives.[8] They discuss the challenges presented by vapor detection and examine techniques from animal olfaction to the

electronic nose. As we rely on vapor detection to discover concealed devices this review summarizes the tools available for early detection.

J. Oxley reviews the history of explosives detection for security purposes and presents “an overview of the history, existing practices, and potential future technologies of explosive detection.”[12] For widespread security applications X-ray remains the primary bulk detection technology while airports and the like rely upon ion mobility spectrometry (IMS) for trace detection. The difficulties with IMS in a security environment where samples can be almost anything humans touch are well documented, but no new overall trace detection technology is enjoying widespread usage.

A review of detecting organic gunshot residues by mass spectrometry is presented by Taudte et al. They discuss currently used mass spectrometry and ionization techniques for the detection of various organic constituents of smokeless powders.[17]

Whetstone and Kearfort examine explosives detection with active neutron interrogation and review these types of neutron techniques, including fast neutron analysis, thermal neutron analysis, pulsed neutron analysis, neutron elastic scatter and fast neutron radiography.[18]

3. Explosive Standards and References, Laboratory Quality Control, Contamination Prevention

There are two sources found in the bibliography.

4. Sampling and Concentration of Explosive Traces

Improving the sampling and concentration of explosives is perhaps the most crucial aspect of explosives analysis and detection. Ridding the sample of interferences is important for many reasons including possible false positives and saturating the system with non-target compounds. Therefore much work is still being conducted on this front.

DeGreeff et al look at using a new sampling chamber for the headspace analysis by GC/MS with TNT, HMTD and TATP. The design seems applicable to any organic high explosive.[27]

Fan and Almirall developed a unique sampling method by packing a glass capillary with glass microfibers. Their system is reported to have a surface area of 5000 times that of a single solid-phase (SPME) fiber. This allows for a reported 30 times improvement for NG, 2,4-DNT and DPA.[29] It should also prove to be a vast improvement over detecting less volatile organic explosive and related compounds than we have now.

Hashimoto et al describe a novel cyclone explosive particle concentrating chamber that vaporizes the particle in the chamber and then uses atmospheric pressure chemical ionization (APCI) before introducing it into a linear ion trap mass spectrometer. Reported detection limits mimic typical IMS screening systems and also are close in sampling times. [35] It is unclear if this system has the same issues as IMS but theoretically better data can be obtained.

For micro-gas chromatography, James et al discuss a gas preconcentrator and its potential to improve efficiency and the limit of detection.[37]

Tomaszewski et al describe a series of tests on the efficiencies of carbon/silica adsorbents with various levels of silylation and testing the system with nitramines. They found the

“recovery rate of explosives studied in the SPE procedure depends mainly upon the amounts of carbon deposits (accessible surface area of the carbon phase)”. [43]

Zheng et al report on the preparation of a poly (glycidyl methacrylate-co-ethylene dimethacrylate capillary column with cucurbit[6]uril monotaxane for preconcentration and separation of nitroaromatics. [44]

5. Identification of Explosives, Explosive Residues and Explosive Properties

There are some reports on the properties of explosives and theoretical modeling of explosive behavior.

5.1 General and Properties

Abdul-Karim et al take a look at post blast particle morphologies for aluminized RDX and show that the inorganic portion have spherical shapes and the organic portion had irregular shapes that varied reportedly depending upon location from the detonation. [45]

Narin, Özyörük and Ulas describe a two-dimensional code for looking at the deflagration to detonation transition in granular solid explosives. [61]

Janesheki, Groven, and Son have a very interesting and timely report on characterizing the detonation failure factors of Homemade Explosives (HME). Dealing with primarily ammonium nitrate (AN) and two fuels, they investigate failures due primarily to chemical composition and configuration. [51]

Trying to get more accurate total thermodynamic energy of a given explosive, Lorenz, Lee and Chambers propose using a device called the Disc Acceleration Experiment (DAX) measured with photonic Doppler velocimetry. [58] Several other instruments were used, all representing a better overall picture than the older tests cited in tomes like the Picatinny Arsenal publications.

5.2 TATP

Ezoe, Imasaka and Imasaka analyze TATP with an ultraviolet femtosecond multiphoton ionization pulse with time of flight mass spectrometry and found a small improvement in detection limits, better than those with EI and CI. Furthermore they introduced TATP into human blood to see if the interferences found therein would change the detectability of the analyte. It reportedly did not. [72]

Jiang et al employ a dopant-assisted positive photoionization method with IMS and coupled that with a time-resolved thermal desorption introduction to analyze TATP. They also placed TATP in various complex matrices such as soft drinks and cosmetics. Finally, they did the same for HMTD with good results. [73]

Ray et al present a study using a nanostructured titanium dioxide nanotube for the detection of TATP. [76]

5.3 PETN

Bhattacharia et al delve into the aspect of doping PETN crystals to show that doping will slow the mass-loss rate from PETN crystals. These “impurities” often retard the loss of mass from the surface of PETN crystals and the dopants used here reduced it by 35%. [77]

A potential very useful study of the sources of isotope ratio variation among PETN sources was conducted by Howa, Lott, and Ehleringer. They surveyed 175 PETN samples from 22

manufacturing facilities. They report to be able to discriminate PETN from the same manufacturer. They also report that the precursor pentaerythritol was the source of the variation and not nitric acid.[78]

5.4 ANFO

In a similar fashion, Brust et al explored using isotopic profiling using IRMS and elemental profiling using ICP-MS to discriminate batches of fertilizer grade ammonium nitrate. Using N and O ratios they studied 103 samples from 19 manufacturers. They found 32 elements to be useful for differentiation using ICP-MS (and looked for 66). Linear discriminate analysis was used to show the effectiveness of combining these two techniques for differentiating between sources.[79]

Chakraborty, Bagchi and Chandra Lahiri report on a post-blast case of ammonium nitrate and wax from a bombing in Midnapore, West Bengal.[80]

Hernandes et al characterize ANFO using ESI-FTMS in both negative and positive ionization modes. They use both direct desorption and an ambient sonic spray ionization with a simple single quadrupole mass spectrometer.[82]

Howa, Lott and Ehleringer precipitated ammonium ions using a sodium tetrphenylborate solution in order to do isotope ratio analysis of the ammonium ions solely. They found that the “isolation of ammonium precipitate from solutions containing dissolved nitrates did not influence the nitrogen isotope ratios of test ammonium salts.” This technique allows for separating the ammonium from the nitrate and could be used in future isotope ratio investigations.[83]

5.5 Peroxide Explosives (General)

Aernecke et al measure the vapor pressure of HMTD using secondary electrospray ionization mass spectrometry. They present a vapor curve over the temperature range from 28 to 80 degrees Celsius. It is a worthwhile endeavor to peruse their data to understand possible head space analysis for suspected HMTD and if one can predict the likelihood of detecting HMTD over a period of time on scene after a suspected HMTD explosion and collection in an appropriate container.[85]

Türker and Variş take a close look at the properties of TEX explosive. TEX is an explosive with an isowurtzian cage structure, a nitramine similar to CL-20.[90]

5.6 Other Explosives including Novel or New Explosives

Two types of advances in the production of novel explosives are reported here. As in the last review there are many nanoparticle investigations. Additionally, the need for stability in harsh environments and a push toward environmentally friendlier explosives drive development of new military explosives. Also included are some recently declassified materials. Several studies report on the behavior of these explosives. In the bibliography of this section one will find many citations not discussed.

Anderson et al replace aluminum in an RDX based explosive composition with silicon for further progress in the area of insensitive munitions (IM) and report on their findings. They prove significant reaction of the silicon and note that the energy release is less.[91]

Cheng, Ma, Liu and Shen describe that magnesium hydride sensitized emulsion explosives have far better resistance to pressure desensitization than traditional sensitizers such as glass microspheres.[97] We are unaware whether manufacturers are currently testing or

marketing emulsions with this sensitizer but is another compound to look for while analyzing intact or post-blast emulsion explosives.

A theoretical paper by Lan, Zhai and Yang explores the influence of dimethyl hydantoin (DMH) on GAP/RDX propellants and improves binding energies.[115]

Similarly, Lin et al look at the properties of TATB (1,2,5-triamino-2,4,6-trinitrobenzene) bonded with a styrene copolymer.[118]

Lin, Ma, Shen, and Wan look at the effect of aluminum “fiber” content in RDX based explosives and concentrate on underwater explosion performance.[119]

Further exploring the mechanics of design for explosives, Miao, Shen and Yu show that the critical thickness of explosives can be reduced (in this case with emulsion explosives) by using a honeycomb structure with double sided cladding.[123]

Reese, Groven, and Son propose, in a very interesting paper, to substitute NG with 1,4-dinitrato-2,3-dinitro-2,3-bis(nitratomethylene) butane (SMX). The focus is to prevent degradation of the liquid NG with this SMX room-temperature solid replacement.[129] The same authors propose another composite propellant with another compound.[130] It is unknown how cost effective these would be as that would be a limiting factor in its usage.

Richard and Weidhaas explored the biodegradation of another novel explosive, IMX-101. [131]

Tappan and Chavez investigate the combustion properties of an amino substituted N4BIM explosive formulation.[136]

Tichapondwa et al report on performance testing the substitution of calcium sulfate for barium sulfate used in barium sulfate-silicon long time delay compositions for a more environmentally friendly composition. The composition is supported for Si content between 30-70% of the total mixture.[137]

Türker write about nitrogen analogs of the explosive TEX, a caged structured explosive.[140]

Vargeese, Muralidharan and Krishnamurthy study the catalytic effects of nano-sized titanium particles incorporated in ammonium nitrate based solid propellants.[141]

Walsh et al study the deposition rates and effects of IMX-104, an insensitive military explosive, for decreasing the likelihood of environmental contamination.[142]

Another “green” oxidizer is studied by Wilharm, Chin and Pliskin who propose using potassium ferrate (VI) as an alternative to perchlorates.[144] It appears to be a promising green alternative.

Wu, Luo and Ge studied the plasticizing advantages of glycidyl azide polymers (GAP) in modifying nitrocellulose based powders and show that although some parameters (drop weight impact sensitivity) are reduced with GAP powders, other factors show it is a viable dopant to NC powders for stability.[146]

Xing et al explore the theoretical aspects on the role of aluminum in thermobaric explosives (TBX) and look at different reaction phenomena in the detonations of TBX's.[149]

Yang, Li and Ying propose a replacement propellant in the form of RDX and an inert polymer binder and compare many performance factors to traditional nitrocellulose propellants. The propellant is constructed with a novel process.[150]

Yang et al prepare an energetic cocrystal containing benzotrifuroxan and 1,3-dinitrobenzene. [152]

6. Instrumental Analysis of Explosives

6.1 LC/HPLC/UPLC

Much of the work of the forensic scientist in the laboratory is devoted to utilization of instrumental techniques to identify explosive traces. LC/HPLC/UPLC are excellent separation techniques and can be a part of a positive identification if coupled with specific detection methods, or by using orthogonal methods.

Brust et al quantitate PETN and its degradation products using LC with APCI-MS. The method was employed in a case to discriminate between post-blast PETN (and degradation products) and the natural ratios found in unexploded PETN.[160]

Russel et al use HPLC and HPLC-MS to analyze insensitive munitions (IMX) which are increasingly being used in military applications for their insensitivity and environmental friendliness. The two most common are IMX 101 and IMX 104 which contain four constituents in various ratios.[162] These are likely to be seen in future criminal or terrorist attacks and/or seizures. The explosive analyst would be well served to review both the specifications for these products and methods of analysis as presented here.

Schram, Vailhen and Bridoux quantify trace amounts of organic high explosives in water samples using UHPLC-MS/MS. They employed stir bar sorptive extraction followed by liquid desorption. 10 factors for the experimental design and 8 analytes were considered.[163]

Walsh uses a trifunctionally bonded amide phase HPLC/MS system to rapidly separate NTO, nitroguanidine and DNAN. Analytical runtime was reported at 3 minutes.[164]

Zhu et al describe a hydrophilic interaction chromatography technique (HILIC) to analyze the polar precursors of HMX. These compounds, TAT and DAPT are polar and difficult to analyze using reverse phase LC.[166]

6.2 Ion Chromatography

The technique of Ion Chromatography is used in forensic post-blast analysis for the analysis of both inorganic and some organic explosives. The mass spectrometer is rapidly becoming the detector of choice even for simple ions but other detectors are still used as well.

Gilchrist presents, in a 2015 doctoral dissertation for King's College in London, using high resolution ion chromatography for low explosives. One aspect was to use HRMS to achieve confirmatory identifications of analytes at pictogram levels.[167] As IC/MS progresses in the area of low mass ions, it will progress to more usage of HRMS.

Similarly, Gilchrist, Nesterenko, Smith and Barron report how organic solvent and temperature enhanced IC with HRMS enhance separation and identification of both organic and inorganic low weight ions.[168]

6.3 Gas Chromatography

Brust et al report on impurities in TNT with a quick 4 minute GC-MS method and detect organic impurities less than 1% by weight in intact samples.[170] It would be interesting to see if these findings could translate to post-blast recovery where there is very little residue and whether some of these impurities (DNT's for example) will not be distorted by the actual detonation. That stated, it is useful for intact comparisons at this point.

Chajistamatiou and Bakeas identify nitrocellulose (NC) by GC-MS using EI. They do this by looking at the NC trimethylsilyl derivatives.[171]

Chang, Yew and Abdullah report on the optimization of headspace solid-phase microextraction for the volatiles in smokeless powders. Multivariate analysis showed that sample temperature and extraction time were the two biggest parameters for optimization, and subsequently determined that 66 degrees Centigrade and 21 minutes were optimal. This allowed enough time for extraction to be able to differentiate unique powders.[172]

Field et al use direct liquid deposition of standards onto sorbent-filled thermal desorption tubes for later vapor detection using GC with an electron capture detector for quantitative analysis and thus eliminated the requirement for vapor standards (direct) by combining the sensitivity of their instrument with direct liquid deposition.[173]

Leary, Dobson and Reffner describe the performance characteristics that are important for field use GC/MS instruments including a review of those currently available and their performance.[174] It is an excellent overview of the current state of these types of instruments.

Seneviratne, Ghorai and Murray have developed a method for incorporating small molecule separation with laser desorption with capture on a SPME fiber for injection into a GC/MS instrument. This technique shows promise in dealing with explosives in complex matrices [177].

6.4 Capillary Electrophoresis

A few papers are noted in this section. CE is a powerful analytical technique for separating analytes. Coupled with mass spectrometry it can identify many species of interest to the forensic chemist.

Ali, Alharbi and Sanagi explore the use of nano-capillary electrophoresis in environmental analysis. They couple this with several types of detectors including UV-Vis, conductivity, AA, RI, ICP and MS among others.[179] While not focused exclusively on explosives they include explosives in their suite of analytes.

Bresinger et al use perfluorooctanoic acid (PFOA) as both a chromatography reagent and as a complexation reagent in micellar electrokinetic chromatography (MEKC) for the separation of neutral explosives such as RDX, HMX, tetryl, and PETN with mass spectrometric detection in the negative ion mode. Additionally some nitro aromatic species were directly detected, without forming the complexes. Their detection limits were reported in the high parts per billion.[180]

Saiz et al build a dual channel capillary electrophoresis system, one for cations and one for anions for the specific purpose of analyzing consumer fireworks.[182] As they can be run simultaneously, this system could prove to be a time saver and eliminate the possibility of sample degradation over time spent switching systems. Additionally, you would be running the exact same sample.

6.5 General Spectroscopy: Fluorescence, Luminescence, Spectrophotometric, UV, Chemiluminescence

There are dozens of papers reporting work in this area. They are too numerous and sometimes esoteric to comment on most of them. Some are used in commercial, military, security and law enforcement applications, while some could be in the future. Still others will prove to be too costly to mass produce. Most of these involved making sensors that are specific to one type of explosive or a class of explosives.

A paper presented by Asha, Bhattacharyya, and Mandal is typical of the types of papers in the citations in that it shows the discrimination of nitro aromatic explosives using a luminescent metal-organic framework.[185]

Another area often researched is the use of conjugated or straight polymer applications. Barata and Prata report on detecting DMNB, the only ICAO detection agent in widespread use, in the vapor phase via a Calix (4) arene-based carbazole-containing conjugated polymer.[188]

Still yet another area of research has been conducted in the making of nanosensors of various geometric configurations. Chen et al show trace aromatic explosive detection using fluorescent gC_3N_4 nanosheets.[192] Many of these types of applications involve a good deal of engineering.

As an example of the high degree of engineering, Darr et al employ an ultra-thin oxide capping layers and plasmonic silver gratings to improve the utility of the fluorescent conjugated polymer films previously mentioned, in this case for portable chem/bio sensing applications.[198]

Fernández de la Ossa, Amigo and Garcia-Ruiz show how near infrared hyperspectral imaging (NIR-HIS) can provide a fast non-contact non-destructive method for analyzing explosives on handprints. They look at ammonium nitrate, black powder, smokeless powders, and dynamite. By using a partial-least squares discriminant model they show detection and discrimination.[208]

Gonzalez et al build luminescent silicon nanocrystals with dodecyl groups on paper to detect nitrobenzene, nitrotoluene, and dinitrotoluene.[216]

Hu et al use near IR mediated ratiometric luminescent sensors for the detection of explosives through multimode visualized assays of explosives.[221]

Rembelski, Barthet, Frenois and Gregis employ a heating variable to their fluorescent chemical sensors and report on its utility.[252]

Sun et al report on detecting nitro aromatic explosives using a fluorescent polymer film with self-assembled three-dimensionally ordered nanopores. The films are simply prepared by a dip coating process of a mixture of polystyrene and fluorophore pyrene on a glass slide.[262]

Xu reports on employing a Ti (oxo) salt blended into a cellulose microfibril network to produce a tunable interface for detecting hydrogen peroxide in the vapor phase which produces a yellow color. Further work to improve sensitivity used a naphthalimide based fluorescent sensor.[278]

Xu et al also report this and expand on it by sensing also TATP, DADP, and HMTD in a later paper published in ACS Applied Materials & Interfaces.[279]

6.6 Mass Spectrometry

Mass spectrometry continues to be the gold standard in explosives analysis. There are several applications and various ways mass spectrometry can be achieved. In many cases an identification of a species can be accomplished. Some common nitrate esters still prove problematic for a straight forward unambiguous identification, however.

Agarwal et al report on a soft chemical ionization switchable source for the detection of picric acid. They use a time-of-flight mass spectrometer with a switchable reagent ion source.[289]

Chen, Hou, Hua and Li develop a method and system for introducing wet air into the low temperature plasma stream (in the ionization source) to improve the sensitivity for the detection of explosives.[292]

Cheng et al use ambient mass spectrometry with a dual electrospray and APCI source to simultaneously detect polar and nonpolar compounds. They could modulate between ESI only, APCI only, or ESI+APCI. A pulsed laser was used on sample surfaces.[293]

Clemons, Nnaji, and Verbeck used direct analyte-probed nanoextraction coupled to nanospray ionization and reportedly solve or mitigate the selectivity issues and matrix effects of direct inject electrospray.[295] Hopefully this will prove valuable to those working with post-blast samples.

Forbes and Sisco use direct analysis in real time (DART) for exploring trace samples of erythritol tetranitrate (ETN), an explosive gaining popularity. They look at the competitive ionization between ETN, erythritol itself, and nitric acid.[299]

Jjunju et al constructed a lightweight hand-held APCI ion source for the in situ analysis of nitroaromatic explosive compounds.[305]

Kaplan-Sandquist, LeBeau and Miller look at applying matrix-assisted laser desorption ionization with time of flight mass spectrometry to fingerprints with explosive and pharmaceutical contaminants. The explosive depositions were TNT and RDX.[306]

Kaupila et al compare desorption atmospheric pressure photoionization (DAPPI) to desorption electrospray ionization (DESI) and compared the two with a suite of explosive analytes. They report DAPPI is more sensitive for TNT than DESI but the opposite was true for HMX. They conclude "DAPPI could become an important method for the direct analysis of nitroaromatics from a variety of surfaces. For compounds that are thermally liable, or that have very low vapor pressure, however, DESI is better suited." [307]

Krawczyk analyzes HMTD by ESI-MS on a UPLC-TOF instrument. Ions were formed by use of alkali metal salts. The most surprising aspect was the discovery that HMTD actually undergoes oxidation to tetramethylene diperoxide diamine dialdehyde (TMDDD) and the author explains that in other papers the results should be attributed to TMDDD. The method described in this paper matches the "most sensitive methods" available.[311]

Lubrano et al use a modified SPME fiber using a butyl chloroformate coating to detect ammonia from ammonium nitrate.[312]

Schwarzenberg et al differentiate isomeric dinitrotoluene and aminodinitrotoluene using electrospray high resolution mass spectrometry.[315] High resolution mass spectrometry will likely be the gold standard going forward.

Using the negative ion mode on porous supporting tips, Wong, Man, Che, Lau and Ng explore the electrostatic charging effect of these tips and its application to explosive detection.[320] It's interesting to note the effect of negative ionization on various substrates.

6.7 Isotope Ratio Mass Spectroscopy, IRMS

Howa, Lott and Ehleringer report on carbon and nitrogen isotopes in factory samples of RDX and HMX and show the discriminating power of this technique. Samples of RDX and HMX made in the same factory with two different processes could still be discriminated from other factories.[321]

Lott, Howa, Chesson and Ehleringer also refine their technique using thermal decomposition and look at inorganic nitrates and urea. They also use elemental analysis.[322]

Zalewska, Sikora and Buczkowska use differential ion mobility spectroscopy and constructed a device.[323]

6.8 FTIR

Fourier Transform Infrared Spectroscopy is a workhorse instrument in forensic explosives analysis. Some useful papers are commented upon, below. Many commercial platforms and sampling devices are available.

Banas et al report on a practical study of using FTIR on fingerprint deposition of explosives. They employed tape lifts and analyzed these with an FTIR.[324] This is a useful forensic tool.

Cuisset uses infrared cross-sectional analysis of nitro-compound vapors for traditionally low vapor pressure explosives by analyzing the more volatile compounds present either as a remnant of the manufacturing process or natural degradation. RDX and TNT are investigated.[325]

In a very useful paper, Martin-Alberca et al have studied a variety of commercial fireworks and their post-combustion products via ATR-FTIR. They have identified 22 standard compounds in the original fireworks.[327]

Wang et al have studied HMX, RDX, TATB and TNT in single, binary and tertiary component bonded plastic explosives. By using principle component analysis they could discriminate 100% for the sets of data generated in house but only 40% for actual real world PBX samples.[331]

6.9 Raman Spectroscopy

Raman spectroscopy was, at the last review paper's publication, more reserved for stand off detection than for forensic practices. This has rapidly changed and it is now used in several laboratory applications as well in stand off detection.

Almaviva et al report that they can detect traces of explosives in a single fingerprint from 6-10 meters distance by using an eye-safe UV Raman instrument. They also deposited samples on fabric, leather, and synthetics.[334]

Almeida et al report using Raman hyperspectral imaging and independent component analysis on explosives residues on banknotes from an ATM theft.[335]

Hamad et al use nano-structured copper substrates, fabricated using ultrafast laser pulses, and surface enhanced Raman scattering (SERS).[344]

Malka, Rosenwaks and Bar report on photo-guided sampling with a compact Raman spectrometer.[347] This combination could prove useful in field applications.

6.10 DSC, Thermal Analysis, TG

Some citations are found in the bibliography below.

7. Nanotechnology

As stated in our previous review, “one of the most exciting aspects in explosives in the last decade has been the development of nanotechnology. The microsensing field will be applicable both in field and laboratory testing of explosives.” [7] There are dozens of citations in this area, but two papers caught the interest of the authors.

Du Plessis has an excellent review of the advances in research done on porous silicon with oxidizers. This is one of the best overall reviews on this class of nano-explosive materials (as opposed to nano-sensors). Du Plessis reviews a decade of research and advances and it is likely that this class of explosive will be recovered in a device or crime scene.[366]

Peveler reviews, then builds, nanomaterial based sensors for detection of explosives, mostly through the construction of gold nanosensors, including gold nanostars. Then Peveler builds quantum dot arrays. The detection of five explosives, 2,4-DNT, 2,4,6-TNT, tetryl, RDX and PETN were explored.[377]

8. Detection

Some references are probably not directly applicable to forensic analysis but may be useful to the forensic analyst. A few references will be mentioned up front and the rest arranged in more succinct categories.

Tourne has an excellent review of the developments in explosive characterization and detection.[395] It is a highly recommended paper for the forensic explosive analyst to peruse.

Li, Bassett, Askim and Suslick use a colorimetric sensor array to detect twelve peroxide samples.[389]

Mbah, Steward and Elgebor fabricated a disposable solid phase electrolyte/electrode interface for the detection of trace amounts of TATP and HMTD and their precursors. It appears their apparatus is saturated after the first test.[390]

Peters reports using microfluidic paper-based analytical devices using inexpensive paper utilizing colorimetric reactions for five or more simultaneous analysis. Confirmation of results were completed using EC-ESI-TOF-MS with 18-crown-6 ethers.[392]

8.1 Canine Explosives Detection

Bali, Armitt, Wallace and Day propose tripentanone peroxide (TPTP) as an analog species for TATP in dog training. There are some similar degradation compounds that the authors believe show promise as a safer alternative to having laboratories synthesis TATP for their K-9 teams.[398]

Davis et al explore the possibility that explosive K-9's suffer from gastric disease similar to endurance racing dogs. They report that five days of sustained (but less than racing dog exercise) work on seven dogs shows that gastric disease increased.[399]

Kranz et al investigate what compounds dogs might be alerting to when working with C-4. They note previous studies have suggested an array of chemicals including 2-ethyl-1-hexanol and the suggestion that these be used in place of C-4 as training aides. But they find that 2-ethyl-1-hexanol comes from benign sources such as common plasticizers and products like PVC tile and pipe, electrical tape, and even credit cards. They recommend that because of this 2-ethyl-1-hexanone is not a good substitute.[401]

Lazarowski and Dorman did some investigation into trained Labradors on pure potassium chlorate (PC) and subsequent PC mixtures and found 87% of the 20 dogs did not alert to one or more of the four PC mixtures. They also worked with separate components of the PC mixtures and PC itself and found marked improvement.[403]

Lazarowski et al determined that dogs trained on AN did indeed have decent alerting to AN in various forms and did not alert to either sodium nitrate or ammonium sulfate at higher than chance rates.[404]

Miller et al describe how elephants in Angola were adept at avoiding minefields so they tested three elephants on TNT. The elephants would indicate on TNT better than chance. [405]

Mitchell writes in *Vet Times* about the growing use of giant African pouched rats in some countries for chemical, including explosive, detection.[406]

Sherman et al report on an emotional reactivity test for screening explosive dogs for the United States Marine Corps. The test included measuring of cortisol levels in saliva and plasma to assess the stress level experienced by the canine and was determined to provide useful information regarding the suitability of the canine for work as an improvised explosive detection dog.[409]

Zubedat et al describe how when increasing non-task related stress on a human-dog pair that the human reacts more poorly but the dog's performance increases. The intentional stressors were put on the human.[410]

8.2 LIBS Detection

Bauer, Farrington, Sorauf and Miziolek demonstrate the utility of using laser-induced breakdown spectroscopy for the detection of metal powders found in explosive mixtures as well as other fuels.[413]

Moros, Fortes, Vadillo and Laserma have a chapter on LIBS detection of trace explosives, concentrating on why LIBS will work in some cases and what its weaknesses are. They then discuss different types of available instrumentation including hand-held and stand off units and finally discuss data fusion strategies to differentiate between explosives and harmless materials.[417]

8.3 Neutron

Bagdasaryan et al discuss using a tagged neutron method to detect TNT, tetryl, RDX and ammonium nitrate through different thicknesses of paper and discriminate them from benign materials such as sugar, water, silk and polypropylene.[421]

Ding, Guo, and Shen collected the oxygen coefficients of 396 explosives, 117 other “dangerous” materials such as oxidants and combustibles, and 9 common packing materials. They found that explosives can be distinguished.[424]

Israelashvili et al look at a new detector concept combining imaging with fast-neutron and gamma spectroscopy.[425]

8.4 Terahertz

Beigang et al compare different terahertz instruments and approaches in a proceedings paper.[433] This is decent review of current technology in this area of detection.

Hamdouni et al design a novel detector with metamaterials embedded in a one-dimensional photonic crystal. The transmission of their design reportedly ranges from 3 to 8 THz.[439]

In a focused practical test, Lepodise, Horvat and Lewis experiment with temperature variation from 7 to 245 K while analyzing 2,4-dinitrotoluene and show changes in response with temperature changes.[440]

Walczakowski, Palka and Szustakowski publish a study on the influence of different types of common clothing on the remote identification of explosives with THz. They investigated results at 1,3 and 5 meters on clothing fabric such as viscose, polyester, cotton, spandex, wool, nylon, leather, flax and textiles with multiple types of fibers.[447] This is a useful paper for those who use THz technology in practical detection settings.

8.5 Nuclear Techniques

Apih et al describe a review of nuclear magnetic resonance technology and its current state in detecting explosives and describe some prototypes to overcome weak signals. It works in the differentiation in benign liquids in closed containers versus liquid explosives.[449]

Prado describes a “Bottled Liquid Scanner” using NMR to detect peroxide explosives in sealed containers. The device apparently can scan several bottles at once and reportedly has a low false alarm rate.[455]

8.6 X-Ray

There are a couple of references cited below.

8.7 Ion Mobility Spectroscopy

Liang et al describe the use of adding chlorinated hydrocarbons into the drift gas and first show its improvement in detecting black powder. Other explosives showed marked improvement too, including ANFO, TNT and PETN.[464]

Similarly, Peng, Hua, Wang, Zhou and Li use acid-enhanced evaporation coupled with thermal desorption IMS to show a reported 3000 time increase in response for inorganic explosive oxidizers such as potassium nitrate, potassium chlorate and potassium perchlorate. Organic explosives were unaffected by the acidification process which drastically enhanced the inorganic detection.[468]

Samotaev et al describe a non-harmful process to heat human fingers for detection of explosives and other substances using IMS.[470]

8.8 Novel Detection

The references cited in this section are varied. Several are not necessarily completely novel but are self-described as such or offer some significant variation from the standard technology on which they rely. Several could easily fit into other categories.

Bharawaj and Mukherji detect explosive vapors of RDX and TNT using gold nanoparticles coated on a unique U-bent fiber optic probe for surface plasmon resonance.[474]

Civiš et al offer a very unique way of looking at explosives and propellants. Testing 38 types of commercially produced explosives and propellants, the authors describe using laser-induced breakdown (LIBS) coupled with selected ion flow tube mass spectrometry (SIFT-MS) and then quantitatively comparing the results. Using PCA they show that there are “similarities in the quantitative compositions of the decomposition products for similar explosives and propellants.”[481]

Using electromagnetic exploration geophysics techniques such as ground penetrating radar, Grant, Barrowes, Shubitidze and Arcone show that detection of AN in the sub-surface can be achieved because AN is hygroscopic and will produce a response.[491] While easily applicable to dry environments, the question is whether this will prove useful in a very humid environment and with water saturated soils.

Mallin describes a TATP sensor for continuous monitoring using a thermodynamic gas sensor that measured the heat of decomposition of TATP and a metal oxide catalyst film. [500]

Miller, Woods and Rhoads take a specific look at the near-resonant response of particulate plates formed by hydroxyl terminated polybutadiene and observe the thermodynamic effects of plastic-bonded composites. The authors surmise that this will increase the understanding of vapor based sensor materials.[503]

Much research has been devoted to multi-channel colorimetric testing of explosives using inexpensive paper platforms. Peters et al describe such a device for simultaneous detection of several explosives.[506]

Phelan et al describe a mine and IED detection system using an ultra wideband stepped-frequency radar system.[507]

Roberts, Petraco, and Gittings use a presumptive color test for nitroglycerin in smokeless powders using dimethylaminocinnamaldehyde (p-dmac).[510]

Spitzer et al describe vertically aligned titanium dioxide nanotubes on a cantilever for the detection of the vapors of TNT and PETN.[513]

Vovusha and Sanya researched the binding of RDX, TATP, HMTD, TNT, HMX and PETN to two different substrates, one with boron nitride, and one with graphene and used first principles density function theory to test the binding to each.[516]

Wang, Yang and Zhang describe a microdroplet sensor capillary with a UV fiber sensor for the detection of explosives in soil.[519]

Wojtas et al describe an explosive vapor concentrator to improve detection of explosives. They used TNT.[521]

Finally, Zhou et al report on an electro-spun aggregation-induced emission-active polyhedral oligomeric silsesquioxane copolymer film to improve response by reported 9 fold for explosive vapors.[528]

8.9 Stand Off

Ahmed, Jassar and Jaaz propose and test the use of the internet and the internet of things to connect a network of wireless sensors in order to provide information about potential threats to a central intelligence point rapidly. and test a wireless network of explosive sensors. They explain a metropolitan-wide management system can transmit data in 0.28 seconds or less. The development of explosives sensors was not within the scope of the project.[534] It is unclear which type of stand-off technology they used, but it is interesting to see the integration of said systems in a wide area.

In an interesting test and report that could have ramifications for explosives scene work, Ceco et al tested stand-off Raman spectroscopy (at 532nm) for trace detection of ammonium nitrate and TNT on aluminum post-blast witness plates and plastic containers. [538] If deployed this could give investigators an idea of the explosive used without much disturbance of a crime scene.

Christesen et al investigate a number of laser spectroscopy techniques in a review paper. They recommend orthogonal techniques for positive identification of various illicit materials including explosives.[539]

Using improved specific optical design and electro-optics components, Garibbo, Palucci and Chirico introduce an improved design for Raman scattering stand-off detection of explosives. This system was tested in a Paris Metro station after development.[542]

Kabessa et al describe the use of genetically engineered bacterial biosensors to detect landmines and concealed explosives and report detection of 2,4-DNT at a distance of 50 meters.[545] In a separate citation, Kabessa et al describe their scheme of biosensors on a larger area.[546]

Maksimenko evaluates the efficiency of a tunable CO₂ laser in the IR range for stand-off detection.[548]

In another wireless engineering proposal, Priyanga and Sukanya propose using cellular networks for fast data transfer from explosive sensor networks.[551]

Schwarze et al propose the use of long-wave IR Risley prism laser-beam steering systems as an alternative to conventional scanning and report that it should be better because of its compact size, low power, fast scanning capability and large field of view.[555]

Skvortsov writes a review of terahertz time-domain spectroscopy and active spectral imaging. Overall technique problems and issues are addressed and those interested in this technology should review this paper.[558]

9. Environmental

Developments in analytical techniques used in environmental analysis of explosives provide another area where forensic analysis can find new methods and techniques. Environmental requirements mandate the monitoring of explosive compounds and by-products during the manufacturing process and later in the environment at large.

Giordano et al use micellar electrokinetic chromatography in water samples taken from a Hawaii estuary. This technique is popular and has been used in forensics for organic high explosive analysis. This study found that microbial degradation of certain explosives (TNT, 1,3,5-trinitrobenzene, and 1,3-dinitrobenzene) was probable. They note that their samples were slightly saline versus fresh water samples in other studies.[569]

Junk, Liu, Perkins, and Liu investigate the crystalline structure of 2-hydroxylamino-4,6-dinitrotoluen (2-HADNT), a breakdown product of TNT and a serious environmental contaminant.[573]

Lu et al report on detecting nitrobenzene compounds in surface water using IMS but with molecularly imprinted polymers (MIP-IMS). They report more than 87% of nitrobenzene compounds could be adsorbed onto their MIP with 90-105% (sic) recovery.[576]

Mark et al study how eleven different soils interact with a relatively new insensitive munition explosive 3-nitro-1,2,4-triazol-5-one (NTO), doing a series of kinetic and equilibrium batch experiments.[577]

Sanchez et al developed a silicon micro-analytical platform with a 3D micro-preconcentrator on a hydrophobic zeolite and a chemical gas sensor for a miniaturized vapor detector.[580]

Shemer, Palevsky, Yagur-Kroll and Belkin, as in some previous referenced citations, explore genetically engineered bacteria to detect explosives.[582]

Ueland et al use a microfluidic (capillary channels) paper-based system for explosives detection in soils by fluorescent quenching.[585]

10. Other (Safety, Definitions, Etc.)

One of the biggest issues in explosives analysis is fragmentation distribution patterns. Bors, Cummings and Goodpaster trace and track pipe bomb fragmentation patterns using different types of pipe and different low explosive fillers and map velocities, fragmentation and distribution in different temperature environments. They published their work in two different journals.[593, 594]

Duque, Perry and Anderson-Cook look at how microwaves permeate explosives using several types of explosives.[598]

Keshavaez, Seif and Soury proposed a theoretical framework for predicting the brisance of a given explosive other than the "sand test".[610]

Okada et al describe the experimental testing done in response to an accidental explosion. Conducting 40 laboratory runs they concluded that when ammonium sulfate and sodium hypochlorite are mixed in the presence of platinum black an explosion will occur due to the formation of nitrogen trichloride. They set parameters for safely mixing these two based on their experiments.[617]

Pakkirisamy et al describe how adding water to what they term a "flowerpot" mixture that is barium nitrate, potassium nitrate, aluminum and dextrin lowers the self-heat commencement from 170.62 degree Celsius to 95.71 degrees Celsius. Even at 40 degrees Celsius decomposition begins. Although water slurries reduce friction and spark hazards other hazards remain.[619]

Pittman et al review 100 years of ammonium nitrate disasters.[620] The authors highly recommend this article for forensic analysts and investigators alike.

Soler-Rodríguez and Míguez-Santiyán describe the dangers of tetranitromethane.[627]

10. Final Notes

Papers that were not referenced above can be found in the extensive bibliography. Many of these seem promising as technology advances.

11. Acknowledgements

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Forensic Chemistry

Drugs, 2013-2016²

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Prefacing Remarks

1. With the exception of synthetic cannabinoids and cannabimimetics, all references are subdivided by individual drug, drug group/class, or general topic, then chronologically (year only) within each subsection, then alphabetically by first author within each year. Synthetic cannabinoids and cannabimimetics are in a separate category (1.D), and are subdivided as individual compounds, groups of compounds, and finally as groups with other drugs.
2. Many citations included in this report are dated prior to July 1, 2013, because they had not yet been abstracted prior to the 2013 report. In addition, many of the references in this report are cited as “Ahead of Print;” because their actual publication citation was never subsequently published in *Chemical Abstracts*. For this reason, the year listed with “Ahead of Print” may not reflect the actual year of publication; however, the rest of the citation will remain the same, allowing the full citation to be easily found by Internet searching.
3. All citations are formatted in accordance with Uniform Requirements for Manuscripts Submitted to Biomedical Journals, except that journal names are not abbreviated.
4. In contrast to recent reports, no restricted articles are cited in this report.
5. A small number of citations are bolded, reflecting topics judged to be of notable importance.

² Contains many citations published prior to July 1, 2013 – see Prefacing Remarks.

1. Routine and Improved Analyses of Abused Substances

Improved methods of analysis, i.e., faster, more discriminatory, more sensitive, less costly, etc., are needed for all abused substances. Additionally, standard analytical data are required for previously unknown or rarely encountered substances and/or new "designer drugs."

Drug seizures and clandestine laboratory operations are continuously monitored to provide a comprehensive overview of new developments. Ongoing research in the forensic community, as well as in the general fields of analytical chemistry and toxicology, provide new and/or improved methods of analysis for abused substances. Reports providing standard analytical data for new drugs of abuse and/or improved analytical protocols for known drugs of abuse are generated for the forensic and enforcement communities.

1.A – Individual Compounds or Substances

1.B – Individual Natural Products Containing Abused Substances

1.C – Common Groups or Classes of Compounds or Substances

1.D - Synthetic Cannabinoids and Cannabimimetics

1.E – Mixed or Unrelated Individual (Named) Compounds or Substances

1.A – Individual Compounds or Substances (except individual synthetic cannabinoids and cannabimimetics, which are compiled under 1.D)

Alprazolam: 2014 by UV/Vis after derivatization with DDQ (1); **2015** as a contaminant in "natural waters" by adsorptive cathodic stripping voltammetry (2);

2-Amino-1-(4-bromo-2, 5-dimethoxyphenyl)ethan-1-one (bk-2C-B): 2015 characterization by GC/MS (with and without derivatization with 2,2,2-trichloroethyl chloroformate), LC/HRMS, and NMR (3); synthesis and identification of bk-2C-B by NMR, GC, LC, and HR-MS (4);

5-(2-Aminopropyl)indole (5-IT): 2015 an overview (5);

Amphetamine: 2013 impurity profile of amphetamine produced from APAAN (6); **2014** identification of 4,6-dimethyl-3,5-diphenylpyridin-2-one as a route specific by-product for amphetamine synthesized by the APAAN to P2P, Leuckart route (7); **2015** determination of relative enantiomer migration order using racemic amphetamine (8); **2016** impurity profiling of P2P-derived amphetamine; identification and characterization of the by-products from the APAAN and alpha-methylstyrene routes to P2P and their respective impurities following Leuckart reduction (9);

Barbital: 2013 determination by RP-HPLC (10);

Benzphetamine: 2014 production and impurity profiling of benzphetamine HCl (11);

1-Benzylpiperazine (BZP): 2013 a review (social focus, but includes “analytical methodologies for the identification of BZP in forensic settings”) (12); 2015 determination of the isotopic makeup of BZP synthesized from 3 different sources by IRMS (13);

4-Bromo-2,5-dimethoxyamphetamine: 2015 by LC-MS/MS (14);

2-(4-Bromo-2,5-dimethoxyphenyl)-N-[(2-methoxyphenyl)methyl]ethanamine (25B-NBOMe): 2015 by HP-TLC (15); a review (16);

Buphedrone (2-(Methylamino)-1-phenylbutan-1-one): 2013 characterization with GC/MS, HPLC-DAD, and LC-MS/MS (17);

Buprenorphine: 2016 abuse and diversion of the buprenorphine transdermal delivery system (18);

Camfetamine (N-Methyl-3-phenyl-norbornan-2-amine): 2014 an overview (19);

Chloral Hydrate: 2015 detection of chloral hydrate adulteration in alcoholic beverages (20);

2-(4-Chloro-2,5-dimethoxyphenyl)-N-[(2-methoxyphenyl)methyl]ethanamine (25C-NBOMe): 2013 characterization by GC-EI-MS (with and without derivatization with TFAA), LC-ESI-QTOF-MS, FTIR, and NMR (21); 2014 an overview (22);

4-Chloromethcathinone (Clephedrone): 2014 characterization by GC/MS, NMR, GC, and CE (23);

Cocaine: 2012 rapid separation and characterization of cocaine and various cutting agents by differential mobility spectrometry-MS (24); optical detection using a highly specific triple-fragment aptamer (25); 2013 by electrochemical determination (26); by GC/FID (27); determination on circulated banknotes by CE with UV detection (28); separation of cocaine and phenyltetrahydroimidazothiazole mixtures (29); profiling of cocaine seized in Naples, Italy, by 1H-NMR (30); analysis by GC/MS, ATR/FTIR, and chemometric methods (31); detection of contamination of Brazilian currency by HPLC/UV (32); detection of hygrine and cuscohygrine as possible markers (to distinguish coca chewing from cocaine abuse) by GC/MS (33); fluorescent sensing of cocaine based on a structure switching aptamer, gold nanoparticles, and graphene oxide (34); comparative analysis of solvent impurity profiles obtained by HS-GC/MS (35); detection by a fluorescent biosensing system (36); 2014 IMS evaluation of cocaine occupational exposure in forensic laboratories (37); electrochemical detection using disposable sensors (38); determination of levamisole and tetramisole in cocaine by enantioselective HPLC with circular dichroism detection (39); the stability of cocaine and its metabolites in municipal wastewater (presents the case for using metabolite consolidation to monitor cocaine utilization) (40); impurity profiling of cocaine seized by the Brazilian Federal Police in 2009-2012 (41); determination of cocaine, benzoic acid, benzoylecgonine, caffeine, lidocaine, phenacetin, benzocaine, and diltiazem by HPLC/DAD (42); analysis of “crack” by

Scotts color testing, TLC, GC/FID, and GC/MS (43); quantification by IR and PLSR (44); detection by microfluidic paper sensors (45); determination of the isomeric truxillines in illicit cocaine via CGC/FID and their use and implication in the determination of cocaine origin and trafficking routes (46); a bio-inspired solid phase extraction sorbent material for cocaine (47); radiographic (CT) features of intracorporeally smuggled (body-carried) liquid cocaine versus solid cocaine (48); determination of cocaine, its metabolites, and its pyrolytic products by LC-MS using a chemometric approach (49); determination by diffuse reflectance measurements in the near IR (50); colorimetric detection with aptamer-gold nanoparticle conjugates coupled to an android-based color analysis (51); qualitative analysis by DESI-MS (52); the evaluation of trace cocaine on banknotes (53); novel optical fibre-based cocaine sensors (54); a study of the inclusion complex between p-sulfonated calix[4]arene with cocaine HCl by fluorescence and ¹H NMR (55); **2015** determination of cocaine on Brazilian banknotes (analytical methodology not identified in the abstract) (56); multicriteria FTIR/ATR wavenumber selection to differentiate cocaine base versus HCl (57); an electroanalytical method for the quantification of aminopyrine in cocaine (58); chemical profiling of cocaine seizures in Finland by GC/MS (59); comparison of canine detection of methyl benzoate released from 4 different species of snapdragon versus actual cocaine (60); differentiation of South American crack and domestic (US-produced) crack cocaine via HS-GC/MS (61); the influence of medium and elicitors on the production of cocaine, amino acids, and phytohormones by *Erythroxylum coca calli* (62); a study of the inclusion behavior of p-sulfonated calix [4,6,8] arene with cocaine HCl by fluorescence and ¹H NMR (63); a discussion of levamisole in cocaine preparations (64); quantification of cocaine and adulterants by IR and PLSR (65); determination of cocaine in creek water via SPE with subsequent analyses by either HPLC or GC (66); quantification of cocaine, caffeine, 4-dimethylaminoantipyrine, levamisole, lidocaine, and phenacetin by GC/NPD (67); copper thiocyanato complexes and cocaine (a case of “black cocaine”) (68); chemical profiling of cocaine in Brazil from 2010 to 2013, a discussion of the increase in aminopyrine in cocaine (analytical methodology not identified in the abstract) (69); HS-GC-MS analysis of South American commercial solvents to monitor their use in the illicit conversion of cocaine base to HCl (70); profiling cocaine and some common adulterants by FTIR/ATR (71); a review of nanomaterial-based cocaine aptasensors (72); profiling of cocaine by FTIR/ATR, GC/MS, and HS-GC/MS determination of minor alkaloids and residual solvents (73); ultra-high frequency piezoelectric aptasensor for the label-free detection of cocaine (74); identification of different forms of cocaine and substances used in adulteration using NIR Raman spectroscopy and infrared absorption spectroscopy (75); determination of cocaine, its main metabolites, and its pyrolytic products by HPLC-UV-CAD (76); voltammetric determination of cocaine using carbon screen printed electrodes chemically modified with uranyl Schiff base films (77); optical fibre fluorescent chemical probes for the detection of cocaine (78); **2016** detection and unambiguous identification of traces of cocaine on Euro banknotes using FAPA-MS (79); analysis of cocaine and its adulterants by TLC coupled to paper spray ionization MS (80); fast on-site screening of cocaine with a wearable fingertip sensor based on voltammetry (81); **geographically sourcing cocaine's origin by delineation of 19 major coca growing regions in South America** (82); determination of cocaine, diltiazem, benzocaine, levamisole, caffeine, phenacetin,

lidocaine, and dipyrone by LC/DAD (83); use of a small-molecule-dependent split aptamer assembly for detn. of cocaine (84); detection by a fluorescence immunoassay (85); use of a key aptamer structure-switching mechanism for the ultrahigh frequency detection of cocaine (86); the stability of cocaine impurity profiles during 12 months of storage, by GC/MS and HS-GC-MS (87); removal of benzoylecgonine in water matrices by UV254/H₂O₂ processing using a flow microcapillary film array photoreactor (88); determination of procaine in cocaine by a paper-based device coupling electrochemical sample pretreatment and colorimetric detection (89); polarographic determination of the stability constant of the complex formed between cocaine and cobalt thiocyanate (90); detection by a electrochemical aptasensor (91); a fluorescent aptasensor for cocaine based on a G-quadruplex and ruthenium polypyridyl complex molecular light switch (92);

Clobazam (7-chloro-1-methyl-5-phenyl-1,5-dihydro-benzo[1,4]diazepine-2,4-dione): 2015 the dynamic behavior of clobazam on HPLC chiral stationary phases (93); 2016 spectroscopic and quantum chemical studies of the molecular geometry, frontier molecular orbital, NLO, and NBO analysis of clobazam (94);

Codeine: 2013 detection using a label-free electrochemical biosensor based on a DNA aptamer (95); 2014 a rapid colorimetric method for the detection of codeine sulphate using unmodified gold nanoprobe (96); analysis of codeine phosphate sustained release capsules by HPLC (97); 2015 development of an abuse- and alcohol-resistant formulation of codeine phosphate (98); 2016 photocatalytic degrdn. of codeine by UV-irradiated TiO₂ (99);

Deschloroketamine (2-Methylamino-2-phenylcyclohexanone): 2016 characterization of deschloroketamine by GC/MS, LC/HRMS, MS/MS, and NMR (100);

Desomorphine ("Krokodil"): 2014 a review (101); 2015 an overview and review (102); analysis by TLC, UV/Vis, ¹H NMR, and FTIR (103);

Diazepam: 2015 differentiation of licit and illicit diazepam tablets by DSC (104); 2016 determination of the compatibility between diazepam and tablet excipients by DSC, thermogravimetry, and IR (105);

3,4-Dichloro-N-[2-(dimethylamino)cyclohexyl]-N-methylbenzamide (U-47700): 2016 the first reported fatality associated with U-47700 (and implications for forensic analysis) (106);

1-(2,3-Dihydro-1H-inden-5-yl)-2-phenyl-2-(pyrrolidin-1-yl)ethanone ("Indapropyphenidone"): 2015 characterization by GC/MS, LC-HRMS, NMR, and X-ray crystallography (107);

Diltiazem: 2015 analytical characterization of two new related impurities of diltiazem (2-(4-methoxyphenyl)-5-methyl-4-oxo-2,3,4,5-tetrahydrobenzo[b][1,4]thiazepin-3-yl acetate and 2-(4-methoxyphenyl)-4-oxo-5-vinyl-2,3,4,5-tetrahydrobenzo[b][1,4]thiazepin-3-yl acetate) by HRMS and NMR (108);

1-(3,4-Dimethoxyphenyl)-2-(ethylamino)pentan-1-one (DL-4662): 2015 characterization by NMR, GC/MS, and HPLC (109);

4,4'-Dimethylaminorex (4,4'-DMAR): 2015 chemistry, pharmacology, and toxicology (110); an overview (111);

1,3-Dimethylamylamine (DMAA): 2014 determination by ¹H NMR (112); **2015** identification by DART-QTOF-MS (113);

1,3-Dimethylbutylamine (DMBA): 2015 identification in dietary supplements by UHPLC/MS (114);

N,N-Dimethyltryptamine (DMT): 2014 conformational, spectroscopic and nonlinear optical properties (a theoretical study) (115); a review (also presenting the results of a global survey) (116);

Eszopiclone: 2013 determination by UHPLC and HPLC (117);

N-Ethyl-alpha-ethylphenethylamine: 2013 characterization by GC/MS, LC-TOF-MS, and 1D- and 2D-NMR (118);

2-(Ethylamino)-1-(4-methylphenyl)-1-pentanone (4-MEAP): 2015 analysis by GC/MS, NMR, and LC/EIS (119);

Ethylone (3,4-Methylenedioxy-N-ethylcathinone): 2015 synthesis and characterization of two conformational polymorphs of ethylone HCl by FTIR, FT-Raman, powder XRD, GC-MS, ESI-MS/MS and NMR (¹³C CPMAS, ¹H, ¹³C) (120);

Etizolam: 2014 synthesis (121);

Fenethylamine: 2016 a review (122);

Fentanyl: 2012 impurity profiling of illicit fentanyl using UHPLC-MS/MS (123); **2015** discussion of a case of abuse via extn. of fentanyl from transdermal patches (124); organic and inorganic impurity profiling of fentanyl produced by 6 different methods, using GC-MS, LC-MS, and ICP-MS (125); **2016** impurity profiling using multivariate statistical analysis of orthogonal mass spectral data (includes GC/MS, LC-MS/MS-TOF, and ICPMS) (126);

Flephedrone: 2015 characterization by ¹H, ¹³C, ¹⁵N HMBC, and ¹⁹F NMR (127);

Flubromazepam: 2013 characterization by NMR, GC/MS, LC-MS/MS, and LC-Q-TOF-MS (128);

Flunitrazepam: 2013 electroanalytical sensing using screen-printed graphite electrodes (129); **2014** electroanalytical sensing using electrogenerated chemiluminescence (130); **2015** novel reductive-reductive mode electrochemical

detection by HPLC with dual electrode detection (131); **2016** detection in beverages using portable Raman (132);

6-Fluoro-3,4-methylenedioxyamphetamine: **2015** crystal structure (133);

4'-Fluoro- α -pyrrolidinobutyrophenone (4F-PBP): **2015** structural characterization by ^1H , ^{13}C , ^{19}F NMR, and MS (134);

Heroin: **2012** a review of crystal water in heroin HCl standard (135); **2013** high resolution impurity profiling by UHPLC (136); determination of heroin, morphine, 6-MAM, codeine, and 6-acetylcodeine drug samples using HPLC with "parallel segmented flow," which enables the simultaneous use of UV-absorbance, tris(2,2'-bipyridine)ruthenium(III) chemiluminescence, and permanganate chemiluminescence (137); **2014** determination of heroin, 6-acetylmorphine, acetylcodeine, morphine, noscapine, papaverine, caffeine, acetaminophen, lactose, lidocaine, mannitol, and piracetam by ^1H NMR and 2D DOSY ^1H NMR (138); comparison of quantitation of illicit heroin HCl samples obtained by quantitative NMR versus results obtained by CE (139); an overview of the detection of heroin (140); inorganic impurity profiling and classification of illicit heroin by ICP-MS (141); **2015** acetaminophen, caffeine, diazepam, phenobarbital, and alprazolam in heroin by GC/MS (142); characterization and origin of the 'B' and 'C' compounds in the acid/neutral forensic signatures of heroin (143); classification of illicit heroin by UPLC-Q-TOF analysis of acidic and neutral manufacturing impurities (144); **2016** site- and species-specific hydrolysis rates of heroin to the mono-acetylmorphines (145);

Human Growth Hormone (HGH) (and related substances): **2014** identification of the growth hormone-releasing hormone analogue [Pro1, Val14]-hGHRH in a confiscated product (146); identification and quantification of GHRP-2 by NMR and MS (147); **2015** quantification of HGH by isotope dilution-HPLC/MS (148);

Hydrocodone: **2014** synthesis from thebaine in six steps (149); **2015** wastewater effluent hydrocodone concentrations as an indicator of a drug disposal program success (analytical methodology not identified in the abstract) (150);

Hydromorphone: **2016** two orthorhombic polymorphs of hydromorphone (151);

γ -Hydroxybutyric Acid (GHB) (also γ -Butyrolactone (GBL), 1,4-Butanediol (BD), and Tetrahydrofuran (THF)): **2013** a comprehensive study of the worldwide distribution of GBL using internet monitoring, comparison of packaging, and carbon isotopic measurements (152); detection of GHB, GBL, and BD in dietary supplements and foods, by GC/MS (using isotopologues for quantitation) (153); development of a fluorescent sensor for GBL (154); **2014** a review of the relative risks of GHB and GBL (155); development of a fluorescent sensor for GHB (156); **2015** analysis of GBL and 1,4-BD by chemical ionization-ion trap-GC/MS (157); **2016** comparative study of GHB and other derivative compounds (GBL, butyric acid, and succinic acid) by spectroelectrochemistry Raman on platinum surface (158); detection of BD in spiked drinks (analytical methodology not provided in the abstract) (159);

Ibogaine: 2013 determination by GC-MS/MS (160);

2-(4-Iodo-2,5-dimethoxyphenyl)-N-[(2,3-methylenedioxyphenyl)methyl]ethanamine (25I-NBMD): 2013 characterization by LC, ESI-QTOFMS, GC/MS, and MS/MS (161);

Ketamine: 2012 a simple color testing reagent for screening (162); 2013 screening in orange juice by TLC (163); a review of O-chlorophenyl cyclopentyl ketone (the precursor for ketamine) (164); 2014 wearable devices based on ionic liquid-based SPME for the environmental monitoring of ketamine (165); estimation by UV/Vis (166); electroanalytical sensing using electrogenerated chemiluminescence (167); a review (168);

Lisdexamfetamine Dimesylate: 2012 synthesis and characterization by FT-IR, NMR, ESI-TOF/MS, GC-MS, and HPLC (169);

Lysergic Acid Diethylamide (LSD): 2014 determination by adsorptive stripping voltammetry (170);

Mephedrone (4-Methylmethcathinone): 2013 by SERS with a portable Raman (171); 2014 a study of phase transformations (to minimize transitions between polymorphic forms during storage) (172); use of mephedrone as an exemplar in an interpretative spectroscopy exercise in a second-year bioscience program (173); analysis of purity and cutting agents in street-level samples from South Wales collected between Nov. 2011 and March 2013, by FTIR (4-fluoromethcathinone and 4-methylethcathinone were also found) (174); structures of mephedrone hydrogen sulfate and its polymorphs under ambient and high pressure conditions (175); 2015 computational studies on molecular structure and interpretation of vibrational spectra, thermodynamical and HOMO-LUMO analyses of mephedrone using density functional theory and ab initio methods (176); spectrophotometric determination (177); identification of 1,2,3,5-tetramethyl-4-(4-methylphenyl)-1H-imidazol-3-ium salt (TMMPi), formed during the synthesis of mephedrone (analysis by GC/MS, LC/MS, NMR, and crystal structure determination (178); 2016 detection via an anthracene molecular probe (by NMR) (179);

Methamphetamine: 2012 analysis of the enantiomeric makeup of methamphetamine in OTC inhalers (also includes a toxicology study) (180); fates of precursors and byproducts in soil from the Leuckardt, Nagai, and dissolving metal reductive syntheses of methamphetamine (181); evaluation of the effects of synthesis conditions on the $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, and $\delta^2\text{H}$ stable isotope ratio values of methamphetamine (182); 2013 detection of pharmaceutical impurities in methamphetamine by GC/FID and GC/MS (183); rapid quantitation of methamphetamine by FTIR/ATR and Chemometrics (184); impurity profiling by CE using a highly sulfated gamma-cyclodextrin as a chiral selector (includes methamphetamine, amphetamine, ephedrine, pseudoephedrine, norephedrine, and norpseudoephedrine) (185); screening of methamphetamine, pseudoephedrine, and ephedrine by a portable lab-on-a-chip instrument (186); quantitation of airborne

methamphetamine by SPME and GC/MS (187); detection in indoor air using dynamic SPME followed by GC/MS (188); elemental profiling of methamphetamine using ICPMS (189); influence of precursor solvent extraction on stable isotope signatures of methamphetamine prepared from OTC pharmaceuticals using the Moscow and hypophosphorous syntheses (190); stable isotope analysis of methamphetamine, to help determine precursors (191); molecular fluorescence spectroscopy of methamphetamine in methanol (192); rapid, nondestructive screening test for methamphetamine in clandestine laboratory liquids by Raman (193); impurity profiling of methamphetamine synthesized from P2P prepared from phenylacetic acid or its esters (194); terahertz spectra of methamphetamine HCl (195); **2014** differentiation of ephedrine and pseudoephedrine based methamphetamine samples by 2D-HPLC (196); determination of methamphetamine in sewers using a Polar Organic Chemical Integrative Sampler followed by HPLC-MS/MS (197); real time quantitative (Simon) colourimetric test for methamphetamine detection using digital and mobile phone technology (198); a review of methamphetamine profiling (199); **use of IRMS for methamphetamine profiling (comparison of ephedrine and pseudoephedrine-based samples to P2P-based samples)** (200); use of 10-ethylacridine-2-sulfonyl chloride for detection of methamphetamine (201); **2015** “amine-rich carbon nanodots” as a fluorescence probe for methamphetamine precursors (202); photocatalytic degradation of methamphetamine in wastewater by UV/TiO₂ (203); use of methamphetamine impurity profiling for intelligence gathering (204); detection by a fluorescence nanosensor (with comparison with HPLC) (205); identification of trans-N-methyl-4-methyl-5-phenyl-4-penten-2-amine HCl as an impurity in methamphetamine synthesized via reductive amination of P2P made from phenylacetic acid/lead (II) acetate (206); enantiomeric profiling of methamphetamine by LC-MS-MS (207); **2016** determination of the synthetic routes of methamphetamine using GC-MS and multivariate analysis (208); demethylation of methamphetamine by UV treatment at wastewater treatment plants (209); detection of trace methamphetamine by dual-mode plasmonic naked-eye colorimetry and a SERS sensor with a handheld Raman spectrometer (210);

Methaqualone: **2013** simultaneous determination of methaqualone, saccharin, paracetamol, and phenacetin in illicit drug samples by HPLC (211);

Methcathinone: **2012** detection by HPLC (212); **2013** qualitative and quantitative analysis by LC/MS/MS (213); quantitative analysis by GC/MS (214);

Methiopropamine: **2015** indirect electrochemical detection of methiopropamine (MPA) and 2-aminoindane (2-AI) by Raman spectroscopy, presumptive (color) testing, HPLC, and electrochemical analysis (this mixture was referred to as “synthacaine”) (215); by selective reagent ionisation-TOF-MS for analysis of a mixture of methiopropamine and benzocaine (also referred to as “synthacaine”) (216);

Methoxetamine: **2013** by GC-MS and ¹H- and ¹³C-NMR (217); **2014** a review (218);

2-Methoxydiphenidine (2-MXP): 2015 synthesis and characterization (includes the positional isomers; toxicological focus) (219);

para-Methyl-4-methylaminorex: 2014 an overview of deaths from use (220);

3,4-Methylenedioxy-N-benzyl cathinone (BMDP): 2013 characterization by LC/high res QTOF-MS, EI-MS, IR, and 1D- and 2D- 1H- and 13C-NMR (221);

3,4-Methylenedioxymethamphetamine (MDMA): 2013 enantiomeric purification by batch chromatography with a cyclodextrin chiral selector (222); use of organic and inorganic impurities in MDMA for comparative analyses (223); impurity profiles of MDMA synthesized by different routes or by variations in the same routes, by GC/MS and GCxGC-TOF-MS (224); 2014 the effects of extrn. procedure and GC temp. programming on MDMA impurity profiles (225); by voltammetry (226); 2015 analysis by direct laser ablation with TOFMS (227); compression studies (228); impurity profiling of MDMA synthesised from catechol (229); chemiluminescence detection of MDMA in street drug samples (230);

3,4-Methylenedioxy-4-methylaminorex (MDMAR): 2015 synthesis of the cis- and trans- isomers, with characterization by “chromatographic, spectroscopic, mass spectrometry, and crystal structure analysis” (231);

Methylenedioxypropylone (MDPV): 2013 injection of MDPV among needle exchange program participants in Hungary (232); 2014 a review, including sepn. and analysis by TLC, GC/MS, HPLC, and LC/MS (233); analysis by GC/MS and LC/MS (234); a review (235); see also phencyclidine (below) for a related citation;

4-Methylethcathinone (4-MEC): 2013 by GC/MS, HPLC-DAD, and LC-MS/MS (236);

Methylhexaneamine: 2013 by GC/HR-TOFMS with soft ionization (237);

β-Methylphenylethylamine (BMPEA): 2015 by LC-QTOF-MS (238);

4-Methylthioamphetamine (4-MTA): 2012 identification of common impurities found in 4-MTA produced by the reductive amination and nitropropene routes (239); identification and synthesis of by-products found in 4-MTA produced by the Leuckart method (240);

Mianserin (a psychoactive tetracyclic antidepressant): 2012 by TLC, color testing, and UV (241);

Midazolam: 2015 a review of published, validated methods for determination of midazolam in pharmaceuticals (242);

Morphine: 2013 evaluation of stationary phases based on silica hydride, using morphine as the model compound (243); determination in compound liquorice tablets by HPLC with online SPE (244); 2014 detection using electroactive polymers (245);

highly sensitive detection based on molecular imprinting polymers using surface plasmon resonance (246); determination in pharmaceutical samples by kinetic spectrophotometry (247); conformational complexity of morphine and morphinum in the gas phase and in water (a DFT and MP2 study) (248); degradation of morphine in opium poppy processing waste composting (249); **2015** “fingerprinting” using chromatographic purity profiling and multivariate data analysis (250); a study of the stability of morphine sulfate orally disintegrating tablets (analytical methodology not identified in the abstract) (251); a review of sugar derivatives of morphine (252); **2016** a structural and computational study (to determine morphine’s mechanism of action as an antioxidant) (253); photostability of 6-MAM and morphine exposed to controlled UV irradiation in water and methanol (254); characterization and origin differentiation of morphine base, HCl, and sulfate (and other unspecified “morphine derivatives”) by DSC/TG and FTIR (255); detection using cathodically electropolymerized, molecularly imprinted poly(p-aminostyrene) films (256); determination in pharmaceutical products by on-line SPE and HPLC (257);

Oripavine: **2014** a review of the chemistry of oripavine and its derivatives (258);

Oxycodone: **2013** analysis of oxycodone/acetaminophen tablets by HPLC (259); a study on the effectiveness of reformulated (abuse deterrent) oxycodone tablets (260); **2014** a review (261); the impact of a reformulation of extended-release oxycodone designed to deter abuse in a group of prescription opioid abusers (262); reductions in reported deaths following the introduction of extended-release oxycodone with an abuse-deterrent formulation (263); **2015** impact of the introduction of an abuse-deterrent sustained-release formulation in Australia (264); an overview of the level and methods of tampering with a tamper-resistant formulation (265); **2016** evaluation of the tamper-resistant properties of biphasic immediate-release / extended-release oxycodone/acetaminophen tablets (266);

Phenazepam: **2012** analysis of phenazepam by GC/MS and LC-MS/MS (267);

Phencyclidine (PCP): **2013** false-positive PCP immunoassay caused by MDPV (268);

Phenobarbital: **2014** detection by an electrochemical sensor based on molecular imprinted polymer (269); detection by an electrochemical sensor based on molecular imprinted technique and electropolymerization membrane (270); characterization of the monosolvates between phenobarbital and acetonitrile, nitromethane, dichloromethane, and 1,4-dioxane by single-crystal and powder X-ray diffraction, thermoanal. methods, FTIR, Raman, and solid-state NMR (271); **2015** simultaneous determination of phenobarbital and aspirin by HPLC (272); **2016** a study of polymorphism of phenobarbital by structural, thermal, and VT-Raman spectroscopy (273);

Phenyl Acetyl Carbinol (L-PAC and R-PAC): **2014** isolation/selection of the best yeast culture and its metabolic control for the biotransformation of benzaldehyde to 1-hydroxy-1-phenyl-2-propanone (274); use of substituted benzaldehydes for the manuf. of substituted L-PAC analogs (which were subjected to reductive amination to

give the corresponding substituted pseudoephedrine/ephedrine analog, which were then either reduced or oxidized to produce the corresponding methamphetamine or methcathinone analogs) (275); **2015** biosynthesis of R-PAC in [BMIM][PF₆]/aqueous biphasic system using *Saccharomyces cerevisiae* (276);

Phenyl-2-propanone (P2P, Phenylacetone): **2016** a detailed analysis of the impurities formed when P2P is synthesized via an aldol condensation of benzaldehyde and Me Et ketone (MEK), followed by a Baeyer-Villiger reaction, followed by ester hydrolysis (route specific markers for this synthesis include 3-methyl-4-phenyl-3-buten-2-one, 2-methyl-1,5-diphenylpenta-1,4-diene-3-one, 2-(methylamino)-3-methyl-4-phenyl-3-butene, 2-(methylamino)-3-methyl-4-phenylbutane, and 1-(methylamino)-2-methyl-1,5-diphenylpenta-4-ene-3-one) (277);

Pregabalin: **2016** a literature review (278);

Pyrazolam (8-Bromo-1-methyl-6-pyridin-2-yl-4H-[1,2,4]triazolo[4,3-a][1,4]benzodiazepine): **2013** characterization by GC/MS, LC-MS/MS, LC-QTOFMS, and NMR (also includes a toxicology study) (279);

alpha-Pyrrolidinopentiophenone (alpha-PVP): **2013** thermal degradation during GC/MS analysis (280); **2016** structure by crystallography (281);

Scopolamine: **2013** detection in spiked samples by portable CE with contactless conductivity detection (282);

Sibutramine: **2012** quantitative determination in adulterated herbal slimming formulations by TLC-image analysis and TLC-densitometry (Dragendorff reagent was used for spot detection) (283); **2013** detection of illicit adulteration of botanical food supplements, by color tests, TLC, HPLC-DAD, MS, and NMR (284); **2015** detection and quantitation in herbal medicines by NIR (285);

Testosterone: **2014** stable carbon isotope ratio profiling of illicit preparations (by GC-IRMS) (286); **2016** screening for in aquatic environments by DART-MS (287);

Tianeptine: **2016** identification by “a multi-pronged analysis approach” (not detailed in the abstract) (288);

Tramadol: **2014** a survey of abuse of tramadol in the U.K. (289);

1-(3-(Trifluoromethyl)phenyl)piperazine (TFMPP): **2014** an FTIR, FT-Raman, UV/Vis, and DFT quantum chemical study (290);

Zolpidem: **2014** development of modified-release tablets of zolpidem tartrate (291);

Zopiclone (see also Eszopiclone): **2015** quantitative determination of zopiclone and its impurity by four different spectrophotometric methods (292); quantitative determination of zopiclone and its impurity by HPTLC (293).

1.B – Individual Natural Products Containing Abused Substances (except natural products laced with synthetic cannabinoids and/or cannabimimetics)

Overviews and/or Reviews: **2013** an overview of the hallucinogenic plant and fungal species naturally growing in Mediterranean countries (including *Phalaris aquatica*, *Peganum harmala*, *Mandragora officinarum*, *Hyoscyamus niger*, *Atropa belladonna*, *Datura stramonium*, *Cannabis sativa*, *Psilocybe semilanceata*, and *Amanita muscaria*) (294); **2014** natural products as lead structures for the synthesis of “smart” and “recreational” drugs (295); comprehensive comparison of different MS techniques for the detection, identification, and characterization of bioactive substances in herbal materials, including saponins, alkaloid, tropane alkaloids, lycopodium alkaloids, phenethylisoquinoline alkaloids, benzyltetrahydroisoquinolines, morphine, berberine, dauricine, quinolines, flavonoids, flavones, flavanols, anthocyanidins, etc. (296); a review, covering kava, kratom, *Salvia divinorum*, bufotenine, glaucine, betel, pituri, lettuce opium, and kanna (297);

Ayahuasca: **2015** quantitative determination of the alkaloids in *Tetrapteryx mucronata* (a plant occasionally used in Ayahuasca preparation) by HPLC-ESI-MS/MS (bufotenine, 5-methoxy-N-methyltryptamine, 5-methoxy-bufotenine, and 2-methyl-6-methoxy-1,2,3,4-tetrahydro- β -carboline were identified) (298); **2016** analysis by DART-HRMS (299);

Betel (*Piper betle* Linn): **2013** an overview of its phytochemistry, pharmacological profile, and therapeutic uses (300);

Coca (*Erythroxylum*): **2012** identification using DNA analysis (301); **2014** chemosystematic identification of 15 new cocaine-bearing *Erythroxylum* cultigens grown in Colombia for illicit cocaine production (302); selection and validation of reference genes for quantitative gene expression studies (303);

Damiana (*Turnera diffusa*): **2013** identification and discrimination of damiana in herbal blends by GCxGC (304);

***Datura stramonium* (Jimson weed, Angel Trumpet):** **2013** isolation of (3R,5R,7Z)-3-hydroxy-5-dec-7-enolide, (R)-tuberolactone, daturadiol, monolinoleoyl glycerol, linoleic acid, and lutein from *Datura stramonium* (analytical methodology not identified in the abstract) (305); a review, including testing methods for Flos Daturae (306); **2014** a review of the use of *Datura* for poisoning (307); **2015** analysis of phytochemical alkaloids in *Datura stramonium* by GC/MS (308); DNA molecular identification of *Datura* medicinal plants using ITS2 barcode sequence (309); determination of hyoscyamine and scopolamine in *Datura stramonium* by HPLC (310); fingerprint analysis of Daturae flos using rapid resolution LC-ESI-MS (311);

Ephedra: **2013** determination of ephedrine and pseudoephedrine in *Herba ephedrae* from different habitats and species by HPLC (312); a review and overview, covering the past 10 years (313); optimum conditions for extracting ephedrine from

Ephedra sinica by response surface methodology (based on HPLC analyses) (314); **2014** correlation between the main alkaloid contents and the powder fractions of pulverized Ephedra sinica (analysis by HPLC) (315); **2015** determination of the total alkaloids content, total phenolics content, and total flavonoids content, and determine their relationship in dry herb of Ephedra major, Ephedra distachya subsp. helvetica, Ephedra monosperma, Ephedra fragilis, Ephedra foeminea, Ephedra alata, Ephedra altissima, and Ephedra foliata, by UHPLC/UV (316); the influence of genetic factors on the ephedrine alkaloid composition ratio in ephedra (317); identification and determination of biogenic amines in Ephedrae herba by RP-HPLC with precolumn derivatization (318);

Hawaiian Baby Woodrose (*Argyreia nervosa*): **2015** determination of its alkaloid composition (319);

Khat (*Catha edulis*): **2012** determination of of cathinone, cathine, and phenylpropanolamine in khat by GC/MS and GC/FID (320); **2013** evaluation of the effect of various drying techniques on the levels of cathinone in khat (321); optimized GC analysis for cathine, phenylpropanolamine, and cathinone in khat following derivatization with MSTFA (322); analysis by CE (323); **2015** isolation of kaempferol, quercetin, and myricetin skeletons from khat, with structural analysis by ¹H and ¹³C NMR, and UV (sugars determined by TLC after acid hydrolysis) (324); use of cation-exchange solid-phase and liquid-liquid extraction for the determination of khat alkaloids by reversed phase HPLC-DAD (325); rapid differentiation of khat using single point and imaging vibrational spectroscopy (326); use of a (-)-norephedrine-based molecularly imprinted polymer for the solid-phase extraction of psychoactive phenylpropylamino alkaloids from khat (327); a review (328); a review (329);

Kratom (*Mitragynine speciosa*): **2013** by microscopy, TLC, and HPLC (330); by HPLC/DAD (331); **2014** by DART-MS (332); quantification of mitragynine in Kratom by an indirect competitive enzyme-linked immunosorbent assay (333); identification of mitragynine and O-desmethyltramadol in kratom (analytical method not identified in the abstract) (334); comparison of GC/MS, SFC with DAD, and HPLC with MS and DAD for detection of mitragynine and other indole and oxindole alkaloids in kratom (335); **2015** identification and characterization of indole and oxindole alkaloids in kratom using LC-accurate-QTOF-MS (336); a review (337); a review of its phytochemistry (338); detection of mitragynine and its analogs (analytical method not identified in the abstract) (339); a review of the chemistry of mitragyna alkaloids (340); the chemistry of the mitragynines (341); an overview and review (342); an overview of the physicochemical properties of mitragynine (includes UV and HPLC analyses) (343); **2016** monitoring the mis-use of kratom in sports (344); a review (345); extraction of mitragynine from kratom (346);

Marijuana and Hemp (*Cannabis sativa*) and associated Phytocannabinoids: **2012** comparison of bulk and compound-specific $\delta^{13}\text{C}$ isotope ratio analyses for the discrimination of marijuana samples (347); effects of electrical lighting power and irradiance on indoor-grown marijuana potency and yield (348); of THC in marijuana, by HPLC (349); **2013** effects of cultural conditions on the hemp fibres (350); of marijuana extracts by HPLC/UV following cloud point extraction (351); chemical

profiling of different hashish seizures by GC/MS and statistical methodology (7 cannabinoids were profiled; analytical methodology not identified in the abstract) (352); production, characterization, and application of hemp essential oil (353); optimisation and characterisation of marijuana extracts obtained by supercritical fluid extraction, focused ultrasound extraction, and retention time locking GC/MS (354); by laser-ablation-ICPMS – a review, covering many other applications (355); a study of marijuana potency from the 1970s to the 2000s (356); supercritical CO₂ extraction of cannabis seed oil (and its fatty acid composition analysis) (357); use of ultrasound to extract flavanoids from cannabis (with analysis by UV) (358); determination of cannabinol in “hemp food” by UHPLC-MS/MS (359); potency survey in the Venice, Italy area from 2010-2012 (360); **2014** identification and quantification of cannabinoids in cannabis by HPLC/MS (361); cold pressing and supercritical CO₂ extraction of hemp seed oil (362); simultaneous quantification of THC, THC-Acid-A, CBN, and CBD in seized drugs by HPLC/DAD (363); a surface plasmon resonance-based method for detection and determination of cannabinoids (THC, CBD, and CBN) in hashish, using silver nanoparticles (364); variation in mineral composition in the leaves, bark and core of 5 fibre hemp cultivars (365); comparison of 2 different conventional working electrodes for detection of THC using square-wave voltammetry (366); Bayesian classification criterion for discriminating between drug type (illegal) and fiber type (legal) cannabis at an early stage of the growth (367); analysis of marijuana samples of varying age by the Duquenois-Levine color test (368); variation in preliminary phytochemical screening of cannabis leaf, stem and root (369); separation of aroma compounds from industrial hemp by supercritical CO₂ extraction and on-line fractionation (370); fast fingerprinting of cannabinoid markers by laser desorption ionization using silica plate extraction (371); elucidation of the Duquenois-Levine chromophore (372); the kinetics and thermodynamics of hempseed oil extraction by n-hexane (373); evaluation of fatty acid profile, antioxidant capacity and metabolic content of cannabinoid-free cannabis grown in the Po valley, Italy (374); identification of 5,5-dimethyl-1-vinylbicyclo[2.1.1]hexane as a volatile marker of hashish (375); analytical and phytochemical characterization of the unsaponifiable fraction of cannabis seed oil (376); resolution of co-eluting compounds of cannabis comprehensive 2D-GC/MS with Multivariate Curve Resolution-Alternating Least Squares (377); metals and organic compounds in the biosynthesis of cannabinoids - a chemometric approach to correlating the metal content in the different parts of cannabis with the soils where plants were cultivated (and with their cannabinoids content) (378); synthesis of all 4 stereoisomers of THC (379); understanding cultivar-specificity and soil determinants of the cannabis microbiome (includes descriptions of the endorhiza-, rhizosphere-, and bulk soil-assocd. microbiome of 5 distinct cannabis cultivars) (380); cannabis potency in the Venice area (Italy) (2013 update) (381); extraction of flavonoids from cannabis by ultrasound (and its scavenging activity towards the DPPH radical) (382); **2015** minor oxygenated cannabinoids (9 α -hydroxyhexahydrocannabinol, 7-oxo-9 α -hydroxyhexahydrocannabinol, 10 α -hydroxyhexahydrocannabinol, 10 α R-hydroxyhexahydrocannabinol, Δ 9-THC aldehyde A, 8-oxo- Δ 9-THC, 10 $\alpha\alpha$ -hydroxy-10-oxo- Δ 8-THC, 9 α -hydroxy-10-oxo- Δ 6a, 10 α -THC, and 1'S-hydroxycannabinol) from high potency cannabis (structural elucidation was accomplished by 1D and 2D NMR, HRMS, and GC/MS) (383); supercritical CO₂ extraction of hemp seed oil (384); *ab initio* quantum mechanical calculations on THC (385); fatty acid

composition, and oxidation stability of the hempseed oil from 4 cannabis cultivars (386); determination of the conformation of THC by linear and nonlinear CD (387); using compact mass spectrometry for detection and quantification of cannabinoids in cannabis (388); potential oil yield, evaluation of elemental profiling methods, including laser-induced breakdown spectroscopy, ICPMS, LA-ICP-MS, and μ XRF for the differentiation of cannabis grown in different nutrient solutions (389); quality analysis of cannabis seed oils extracted by the hot-pressing method, the cold-pressing method, or by an aq. enzymic method (390); analysis of cannabinoids and terpenes in cannabis by HPLC/DAD and GC/FID (391); synthesis of THC and related derivatives via a Diels-Alder route (392); isobaric drug analyses of THC and CBD by DART and hydrogen/deuterium exchange (393); molecular imaging of cannabis leaf tissue with MeV-SIMS (394); analysis of marijuana by LC techniques (a literature survey 1990 – 2015) (395); review of marijuana testing rules in Colorado, methods used for testing, and test results (396); increasing sample throughput of cannabis analyses by using a highly selective stationary phase combined with superficially porous particle technol. for HPLC and LC-MS/MS (includes comparison versus UHPLC) (397); screening of cannabinoids in industrial-grade hemp using 2D-LC with chemiluminescence detection (398); use of ^1H NMR and HPLC/DAD to determine cannabis chemotype, extract profiling, and specification (399); the relationship between cannabinoid content and composition of fatty acids in hempseed oils (400); characterization of the smell of marijuana by SPME with multidimensional GC/MS (401); analysis of residual solvents in cannabis extracts by GC (402); an overview of recent improvements in chromatography for analysis of marijuana (403); determination of the relative percentage distribution of THCA and Δ^9 -THC in herbal cannabis seized in Austria - impact of different storage temperatures on stability (404); feasibility of facile quantification of cannabinoid content in cannabis to discriminate drug- from fiber-type cannabis in the field (405); cannabinoid dose and label accuracy in marijuana edibles (406); determination of THC, CBD, and CBN by GC/MS (focus on athletic doping) (407); differences in the extraction of THC, THCA, and CBN from cannabis by long-lasting liq. extrn. in a Soxhlet app. versus pressurized liq. extrn. (408); improving quality control methods for extracting cannabis by flash chromatography (409); determination of selected metals in leaves of cannabis by flame AA (410); simultaneous extraction of total flavonoids and total phenolic compounds from hemp (411); **2016** evolution of 8 cannabinoids and 23 terpenes during the growth of cannabis plants from different chemotypes (412); comparison of new and traditional fiber hemp cultivars (stem, bark, and core yield, and chemical composition) (413); heated headspace SPME of marijuana for chemical testing (414); rapid quantitative chemical analysis of cannabinoids in seized cannabis using heated HS-SPME and GC/MS (415); qual. and quant. detn. of CBD-A, CBD, CBN, THC and THC-A in “cannabis-based medicinal exts.” by HPLC/UV and HPLC-ESI-QTOF-MS (416); report from a Colorado private laboratory on regional cannabis potency (THC, CBD, CBN, THCA, CBDA, THCV, CBDV, CBG, and CBC) by UHPLC analysis (417); potency trends in confiscated cannabis (includes analytical methods; time frame not indicated in the abstract) (418); changes in cannabis potency (focusing on THC and CBD) over the last 2 decades (1995-2014) (419); a discussion of the chem. diversity, biosynthesis, and biol. activity of the various compds. in cannabis, and how these compds. can be used to chem. classify cannabis cultivars (420); analytical testing for the cannabis industry

(consumer safety vs. regulatory requirements) - an overview of current protocols for testing for the active phytochem. constituents (i.e., cannabinoids and terpenes), but also for potential contaminants including heavy metals, residual solvents, pesticides, mycotoxins, and microbiol. contaminants (421); use of flash chromatography for rapid extraction of cannabinoids from marijuana edibles (422); analysis of cannabis grown in eastern Oregon for THC, THC-A, CBD, and CBN (edibles, concentrates, and waxes were also tested) (423); comparison of fiber and seed productivity of 14 com. hemp cultivars were tested in 4 contrasting environments (Latvia, the Czech Republic, France, and Italy) (424); the influences of cultivation setting on the lipid distributions, concentrations, and carbon isotope ratios in cannabis (these lipids can currently be used to trace cultivation methods of cannabis and may become a more powerful marker in the future, once the mechanism(s) behind the patterns is uncovered) (425); detection of Δ^9 -THC and Δ^8 -THC (and also CBD and CBN) by HPLC/UV (426); cleanup of marijuana edibles using automated flash column chromatography (427);

Marijuana (Genetic and/or Proteomic Analyses): **2012** investigations into transgenic marijuana (428); **2013** extraction of high quality DNA from seized Moroccan hashish (429); analysis of THCA Synthase gene expression by real-time quantitative PCR (430); chemotype and genotype of cannabinoids in hemp (431); by DNA analysis (432); polymorphism of DNA and accumulation of cannabinoids by cultivated and wild hemp (433); characterization of seeds by DNA analysis (434); **2014** a simple and efficient method for high quality genomic DNA isolation from cannabis containing high amount of polyphenols (435); diversity analysis in cannabis based on large-scale development of expressed sequence tag-derived simple sequence repeat markers (436); application of DNA barcoding in cannabis identification (437); a PCR marker linked to a THCA synthase polymorphism is a reliable tool to discriminate potentially THC-rich plants of cannabis (438); nomenclature proposal and SNPSTR haplotypes for 7 new cannabis STR loci (439); characterization of 15 STR cannabis loci - nomenclature proposal and SNPSTR haplotypes (440); **2015** the phytoremediation potential of hemp - identification and characterization of heavy metals responsive genes (441); genetic structure of 5 dioecious industrial hemp varieties (442); genetic identification of cannabis using chloroplast trnL-F gene (443); genetic resources of cannabis in the gene bank at INF&MP in Poznan (which holds about 150 accessions from various regions of the world) (444); cold acclimation induces distinctive changes in the chromatin state and transcript levels of COR genes in 9 cannabis varieties with contrasting cold acclimation capacities (445); sequence heterogeneity of cannabidiolic- and tetrahydrocannabinolic acid-synthase in cannabis and its relationship with chemical phenotype (446); the genetic structure of marijuana and hemp (447); gene duplication and divergence affecting drug content in cannabis (448); characterisation of cannabinoid composition in a diverse cannabis germplasm collection (449); **2016** proteomic characterization of hempseed (450); the inheritance of chemical phenotype in cannabis (regulation of the propyl-/pentyl cannabinoid ratio, and completion of a genetic model) (451); monitoring metabolite profiles of cannabis trichomes during flowering period using ¹H NMR-based metabolomics and real-time PCR (452); use of embryos extracted from individual cannabis seeds for genetic studies and forensic applications (a unique profile for each individual was obtained,

and a clear differentiation between hemp and marijuana varieties was observed) (453); identification and characterization of the hemp WRKY transcription factors in response to abiotic stresses (454);

Marijuana – Miscellaneous Topics: **2014** the effects of photoperiod on phenological development and yields of industrial hemp (455); detection of pesticides in seized illegal cannabis plants by UPLC/MS-MS in pos. ESI mode using MRM and GC/MS using scan mode (456); **2015** germination characteristics of hemp seeds under single NaCl treatments of varying concentrations (457); method development towards quantifying marijuana consumption using sewage based drug epidemiology (458); medical marijuana's public health lessons - implications for retail marijuana in Colorado (459); determination of herbicides paraquat, glyphosate, and aminomethylphosphonic acid in marijuana samples by CE (460); an overview of the occupational hazards for employees working in the state-permitted marijuana industries (461); issues with retail promotion of marijuana edibles (462); method development towards quantifying marijuana consumption using sewage based drug epidemiology (463); a series of editorials (published in *Nature*) concerning various aspects of state-permitted marijuana (464); an overview of health and safety issues for state-permitted marijuana businesses (465); a review on the ingredients in and safety of “hemp seed food” (466); **2016** the appropriateness of applying ISO/IEC 17025 standards to cannabis testing laboratories (467); quantification of THC-COOH in wastewater from a residential treatment plant as a tracer of cannabis use, using LC-MS/MS (468); oral cannabidiol does not alter the subjective, reinforcing, or cardiovascular effects of smoked cannabis (469); an overview of the changing regulations and rules of the state-permitted cannabis industry (470); the effects of ethephon (a plant growth regulator) on changes in the amt. of many terpenoid compds. in cannabis, including THC, CBD, chlorophyll, carotenoids, α -tocopherol, and pyruvate (471); an overview of the American Herbal Product Assocn.'s (AHPA) industry guidelines on manufg., producing, dispensing, and lab. operation stds. as they apply to state-permitted cannabis (including the American Herbal Pharmacopeia's (AHP) cannabis monograph) (472); an overview on preserving personal cultivation rights while regulating commercial cultivation as agriculture (focusing on the excessive energy, water, and other resources needed for cannabis cultivation) (473); evaluation of three multiresidue methods for the determination of 61 pesticides on marijuana by LC-MS/MS (474); an overview of the establishment of the cannabis subdivision of the American Chemical Society (475); use of “cannavaping” as a means for administering “medical marijuana” (476); antifungal activity of the volatiles of high potency cannabis against *Cryptococcus neoformans* (477); quantification of THC-COOH in wastewater to assess cannabis consumption in Washington state (478);

Marijuana (“Synthetic Marijuana”) - See “Synthetic Cannabinoids and Cannabimimetics” (Subsection 1.D)

Mimosa: **2013** characterization and purity of DMT isolated from *Mimosa tenuiflora* inner barks (479);

Mushrooms (including *Psilocybe* mushrooms): **2013:** simultaneous determination of mushroom toxins by LC-TOF-MS (480); **2014** analysis of mushrooms by Fluorescent Random Amplified Microsatellites (F-RAMS) (15 samples of *Amanita rubescens* and 22 samples of other hallucinogenic and non-hallucinogenic mushrooms of the genera *Amanita* and *Psilocybe* were profiled) (481); **2015** identification of psilocybin, psilocin, baeocystin, norbaeocystin, and aeruginascin in *Pholiotina cyanopus* by LC/MS (482); genetic identification of hallucinogenic and other poisonous mushrooms (483); **2016** DNA-based taxonomic identification of basidiospores in hallucinogenic mushrooms in "grow-kits" (including LC-UV quali-/quantitative determination of psilocybin and psilocin) (484);

Opium / Opium Poppy / Poppy Seeds (see also Papaver below, and Opiates in Subsection 1.C): **2013** the effects of potassium, boron, and strontium on poppy cultivation (such enhancements may impact impurity profiling studies based on elemental analysis) (485); **2014** simultaneous detn. of morphine, codeine, thebaine, oripavine, papaverine, and noscapine in poppy straw by 2 HILIC methods (486); a review of cold pressed poppy seed oils (487); unambiguous characterization of analytical markers in 4 opium samples using an ion mobility trace detector-mass spectrometer (488); physicochemical properties of opium marc (a waste product from commercial opium processing) (489); management of opium marc as a hazardous waste (490); results from an effort to detect opium fields from a Hyperion image covering a study area in Southwest Afghanistan (491); **2015** comparative analysis of volatile flavor compounds of poppy seed oil extracted by two different methods via GC/MS (492); analysis of alkaloids in poppy straw by HPLC (493); **2016** analysis of opium poppy by 2D-HPLC (494); analysis of poppy seeds (intended for use as food) that had been adulterated with poppy straw (i.e., containing morphine and codeine) by IRMS (495);

Papaver (other species): **2016** measurement of some benzyloquinoline alkaloids in *Papaver bracteatum* (496); developmental accumulation of thebaine and some gene transcripts in different organs of *Papaver bracteatum* (497);

Papaver (Genetic and/or Proteomic Analyses): **2011** characterization of SSR markers in opium poppies (498); **2014** a review of benzyloquinoline alkaloid biosynthesis in opium poppy (499); development of genomic simple sequence repeat markers in opium poppy by next-generation sequencing (500); comparative analysis of *Papaver somniferum* genotypes having contrasting latex and alkaloid profiles (501); transcriptome profiling of alkaloid biosynthesis in elicitor induced opium poppy (502); recessive loci Pps-1 and OM differentially regulate PISTILLATA-1 and APETALA3-1 expression for sepal and petal development in *Papaver somniferum* (503); variation in fatty acid composition of three Turkish opium poppy lines (504); **2015** regulation of the alkaloid biosynthesis by miRNA in opium poppy (505); comparative study for stability and adaptability through different models in developed high thebaine lines of opium poppy (506); **2016** molecular genetic diversity and association mapping of morphine content and agronomic traits in Turkish opium poppy germplasm (507);

Peyote (and other mescaline-containing cacti): 2013 analysis of “peyote tea” by GC/MS and GC/MS/MS in PCI mode (508); 2014 phytochemical study of *Echinopsis peruviana* (509);

Plant Materials (Multiple Plants in Single Studies): 2013 identification of plant materials used as supporting matrices for pharmaceuticals, nutritional supplements, and illicit drugs, by DAD, evaporative light scattering detection, and MS (510); a review of chromatographic herbal fingerprints (the “herbs” and the chromatographic method(s) were not identified in the abstract) (511); isotopic analyses to discriminate between organic and “conventional” plants (512); the effects of 11 elements (Co, Mo, Zn, W, Cr, Cu, B, Fe, V, Mn, Ni plus Ca for second species) on the formation and accumulation of indoles and isoquinolines in seedlings of *Catharanthus roseus* L. and *Papaver somniferum* L. (513); analysis of the plant materials used as support matrices, by DNA analysis, GC/MS, and LC/MS (514); an overview and review of the application of 2D-IR for determining the composition, origin, and authenticity of herbal medications (515); 2014 evaluation of mycotoxins, mycobiota, and toxigenic fungi in opium poppy, licorice root, Indian rennet, and others (516); the study of elemental profile of some important medicinal plants by Flame AA (the study included *Papaver somniferum*) (517); comparison of plant DNA extraction kits for plants identification in forensic botany (the plant species were not identified in the abstract) (518); determination of metabolites in finely powdered plant material by Direct Laser Desorption Ionization MS (519); chemotaxonomical classification of the Solanaceae *Atropa belladonna*, *Datura stramonium*, *Hyoscyamus niger*, *Solanum dulcamara*, and *Duboisia* by FTIR/ATR in combination with cluster anal. (520); use of hyperspectral data for detection of cannabis and poppy sites, including those mixed with masking vegetation (521); 2015 transcriptome profiling of *Catha edulis* and *Ephedra sinica* identifies genes potentially involved in amphetamine-type alkaloid biosynthesis (522); phytoaccumulation of heavy metals in natural vegetation, including cannabis (523); application of chemometrics for identification of psychoactive plants (*Salvia divinorum*, *Mitragyna speciosa*, *Psychotria viridis*, and *Calea zacatechichi*) using GC/MS, AAS, and ICP/MS (524); the chemical properties of cold-pressed vegetable oils from seeds of hemp (*Cannabis sativa* L.), blue poppy (*Papaver somniferum* L.), and several other plants (525); biosynthesis of amphetamine-like alkaloids in *Catha edulis* and *Ephedra* spp. (526); profile of toxic metals in 12 different plant materials, including marijuana, by AA (527); use of EI-LC/MS with supersonic molecular beams for analysis, including cannabis (528); 2016 determination of Mn, Ni, Rb, and Sr in powdered stimulant plants (ginseng, guarana, and others) using high-resolution continuum source AA followed by chemometric classification (529); phytochemical profiling of plants using GC/MS (including cannabis) (530); use of high-throughput DART-HR-TOFMS to screen plant-based drugs of abuse for psychotropic alkaloids and adulterants (plants not identified in the abstract) (531); analysis of *Datura* spp. seeds, kratom powder, kava powder, *Salvia divinorum* leaves, *Kanna* crushed leaf material, *Mimosa hostilis*, *Banasteriopsis caapi*, and Morning Glory seeds by DART-HRMS (532);

***Psychotria viridis* (and related species):** 2015 examination of *Psychotria viridis* (DMT was identified by TLC and HPLC) (533); 2016 structural characterization of dimeric indole alkaloids (brachybotryne, its N-oxide deriv., along with bufotenine)

from *Psychotria brachybotrya* by NMR spectroscopy and theoretical calculations (534);

***Salvia divinorum*:** **2013** differentiation of *Salvia divinorum* from marijuana and tobacco by DNA analysis (535); **2014** quantitative determination of salvinorin A in *Salvia divinorum* (analytical methodology not identified in the abstract) (536); analysis of “legal high” products containing *Salvia divinorum* for Salvinorins A, B, C, and D (analytical methodology not identified in the abstract) (537); **2015** determination of salvinorin A in commercial products available in Mexico, using HPLC (538); **2016** an overview of the chem. and pharmacol. of *Salvia divinorum* and salvinorin A (539).

1.C – Common Groups or Classes of Compounds or Substances (except Synthetic Cannabinoids and Cannabimimetics)

(2-Aminopropyl)indoles: **2013** 2-, 3-, 4-, 5-, 6- and 7-(2-aminopropyl)indole – analyses by GC/MS and LC/MS (540);

Amphetamine-Type Stimulants (ATs) and Related Phenethylamines (PEAs):

2011 Impurity profiling of various ATs by physical characterization, qualitative and quantitative analyses, and identification of adulterants, byproducts, and precursors, using GC, GC/MS, and cluster analyses (541); **2012** analysis of 2-, 3-, and 4-methylmethamphetamine and 2-, 3-, and 4-methylamphetamine, by GC/MS and GC/IRD (542); analysis of methamphetamine, amphetamine, and ecstasy by inside-needle adsorption trap based on molecularly imprinted polymer followed by GC/FID (543); **2013** analysis of 4-bromo-2,5-beta-trimethoxyphenethylamine (BOB), 4-methyl-2,5-beta-trimethoxyphenethylamine (BOD), 3,4-methylenedioxy-beta-methoxyphenethylamine (BOH), and 4-methyl-2,5-dimethoxy-beta-hydroxyphenethylamine (BOHD), by LC-MS/MS (toxicological focus) (544); differentiation of stimulant amphetamines, hallucinogenic amphetamines, and non-amphetamines (none specified in the abstract) by GC/FTIR and cluster analysis (545); determination of ephedrine, methamphetamine, and amphetamine by SERS (546); analysis of amphetamine and methamphetamine by GC-MS after propylchloroformate derivatization (547); determination of diethylpropion, fenproporex, and sibutramine in counterfeit tablets, by FTIR/ATR (548); determination of amphetamines and precursors by a portable instrument combining miniaturized GC and IR Absorption Spectroscopy (549); determination of (unspecified) amphetamines by GC/FTIR (550); synthesis and characterization of 2-, 3-, and 4-methylamphetamine by GC/MS, HR-ESI-MS, NMR, and IR (551); a chemometric system for the automated detection of 159 ATs, using GC/FTIR (552); a review of the 2C series of PEAs (553); analysis of methamphetamine, MDMA, and other ATs by GC/MS after derivatization with iso-Bu chloroformate and SPME (toxicological focus) (554); detection of volatile compounds that could indicate an AT by SPME-GC/MS (P2P was detected in every stimulant sample, and 1-phenyl-1,2-propanedione was detected in some stimulant samples) (555); determination of (unspecified) amphetamines by GC/FTIR (556); a review of impurity profiling and

syntheses of methamphetamine, MDMA, amphetamine, DMA, and PMA (557); identification of phenethylamine, ephedrine, and MDMA by Raman, SERS, and DFT (558); analysis of six (unspecified) isomers of mono-methoxyethylamphetamines and mono-methoxydimethylamphetamines (MeO-DMA) by GC-EI-MS/MS (559); **2014** detection of amphetamines by cluster analysis (560); determination of N-ethyl- α -ethyl-phenethylamine (ETH), N,N-diethylphenethylamine, and phenethylamine in dietary supplements by LC-MS/MS (561); synthesis and SARs of N-benzyl phenethylamines as 5-HT_{2A/2C} agonists (562); potential interferences in the GC/MS analyses of methiopropamine, 4-fluoroamphetamine, 4-fluoromethamphetamine, and 4-methylamphetamine (563); synthesis of [¹³C₆]-labeled amphetamine, methamphetamine, MDA, MDMA, MDEA, PMA, PMMA, 3,5-dimethoxyphenethylamine, 4-bromo-2,5-dimethoxyphenethylamine, and 2,5-dimethoxy-4-iodophenethylamine (564); enantioselective hydrogenation of α,β -disubstituted nitroalkenes to synthesize chiral amphetamines (565); synthesis of phenethylamine via anti-Markovnikov hydroamination of alkenes catalyzed by a two-component organic photoredox system (566); simultaneous enantiomeric separation of methamphetamine, ephedrine, pseudoephedrine, and the chlorointermediates formed during the Emde method, after derivatization with trifluoroacetic anhydride (567); detection of amine-based stimulants by a novel fluorescent sensor (568); chiral separation of cathinone and amphetamine derivatives by HPLC/UV using sulfated β -cyclodextrin as a chiral mobile phase additive (569); **2015** comparisons of chiral analyses of 10 cathinone and amphetamine-derivatives by CEC, SFC, and 3 different LC methods (570); simultaneous voltammetric detection of MDMA and PMA (571); analysis of ATs by DSC (572); enantioselective synthesis of ephedrine, amphetamine, and their analogues via two stereocentered Co(III)-catalyzed hydrolytic kinetic resolution of racemic syn-benzyloxy epoxide (573); analysis of amphetamine, methamphetamine, norephedrine, norpseudoephedrine, ephedrine, pseudoephedrine, dimethylamphetamine, and methylephedrine by chiral CE/MS (574); determination of MDMA, methamphetamine, MDA, and MDEA by portable CE with contactless conductivity detection (575); fast separation of 11 cathinones and 4 phenylethylamines by SFC-positive-ESI-triple-quad-MS (576); “novel” sympathomimetics in supplements actually recapitulate the work of synthetic chemists at pharmaceutical firms during the 1930s and 1940s (577); characterization of N-(ortho-methoxybenzyl)-3,4-dimethoxyamphetamine, N-(ortho-methoxybenzyl)-4-ethylamphetamine, N-(ortho-methoxybenzyl)-4-methylmethamphetamine, and N-(ortho-methoxybenzyl)-5-(2-aminopropyl)benzofuran by MS, IR, and NMR (578); **2016** electrochemiluminescent detection of methamphetamine and amphetamine (579);

Barbiturates: **2013** analysis of barbital, phenobarbital, pentobarbital, amobarbital, secobarbital, butalbital, pentothal, and butabarbital by IR and Raman (580); **2014** by colorimetric sensing (581); computing the acidities of barbituric and thiobarbituric acid (582); a theoretical study on the isomerization and tautomerism of 16 isomers of barbituric acid, using MP2 and B3LYP (583); **2016** an overview of the polymorphism and tautomerism of barbituric acid (584); a review of the chem. of barbituric acids employed in the design and synthesis of different types of compds (585);

Benzodiazepines: 2013 cross reactivity of 3-hydroxy-flunitrazepam, 7-amino-nitrazepam, brotizolam, delorazepam, pinazepam, α -hydroxy-midazolam with a commercial immunoassay test (includes LC-MS/MS analyses) (586); analysis of 11 different benzodiazepines and metabolites by SERS (benzodiazepines not identified in the abstract) (587); an FTIR/ATR spectral library of benzodiazepines (588); analysis of nitrazepam, clonazepam, lorazepam, chlordiazepoxide, alprazolam, clozapine, and diazepam by HPTLC with densitometric measurement and UV scanning (toxicological focus) (589); 2014 quantum chemical study of some benzodiazepines by density functional theory (590); determination of clonazepam and its related substances in pharmaceutical formulations by HPLC (591); determination of bromazepam, clonazepam, and diazepam in the Guanda River, Brazil (analytical methodology not identified in the abstract) (592); detection of diazepam, flunitrazepam, and temazepam in spiked drinks by GC/MS (593); a review of the analysis of benzodiazepines by LC with electrochem. detn. (since 2006, with earlier reports given in summary) (594); analysis of diazepam, alprazolam, clorazepate, temazepam, and bromazepam by confocal Raman microscopy (595); differentiation of benzodiazepines by Raman (596); low temperature separation of the interconverting enantiomers of diazepam, flunitrazepam, prazepam, and tetrazepam by dynamic HPLC on chiral stationary phases (597); detection of benzodiazepines in drinks by electrophoretic fingerprinting (598); 2015 use of supported liquid extraction for the analysis of benzodiazepines by SERS (599); characterization of clonazolam, deschloroetizolam, flubromazolam, and meclonazepam by NMR, GC-EI-MS, LC-MS/MS, LC-QTOF-MS, and IR (600); determination of diazepam, clonazepam, and alprazolam in dietary supplements by UHPLC-HR-Quad-MS (601); predictive modelling of the toxicity of benzodiazepines using descriptor-based QSTR, group-based QSTR, and 3D-toxicophore mapping (602); a study of the mechanism of mass spectral fragmentation of benzodiazepines (603); analysis of chlordiazepoxide, midazolam, nitrazepam, estazolam, oxazepam, lorazepam and alprazolam by HPLC with UV or DAD detection (604); 2016 analysis of benzodiazepines by chip-based electrochromatography coupled to ESI-MS detection (605); determination of chlordiazepoxide; lorazepam; diazepam; oxazepam; medazepam in an alc. "grappa" drink by packed sorbent (MEPS)-UHPLC-UV (606);

Benzofurans: 2015 pharmacological profile of 5-APB, 5-APDB, 6-APB, 6-APDB, 4-APB, 7-APB, 5-EAPB, 5-MAPDB, and the benzodifuran 2C-B-FLY (607);

Bromo-, Chloro-, and Fluoro- Amphetamines and Methamphetamines: 2013 analysis of 2-, 3-, and 4-chloro- and 2-, 3-, and 4-fluoro- amphetamines by CE-LIF, following derivatization with fluorescein isothiocyanate (includes comparisons against CZE-UV, sweeping-MEKC-UV, and LC-Q-TOF-MS) (608); synthesis and characterization of fluoroamphetamines and fluoromethamphetamines by GC/MS and LC-MS/MS, before and after derivatization with various reagents (compounds not specified in the abstract) (609); 2015 discrimination of 2-, 3-, and 4-fluoroamphetamine by Raman (610); differentiation of ring-substituted bromoamphetamine analogs by GC/MS (611); identification of the regioisomers of the chloroamphetamines and chloromethamphetamines by GC-MS/MS (612);

Cathinones: **2012** mass spectral fragmentation of 25 cathinones (not identified in the abstract) by GC-HR-TOF-MS using a soft ionization source (613); analysis of 4-MMC, 4-, 3-, or 2-fluoromethcathinone, 4-methoxymethcathinone, N-ethylcathinone, and N,N-dimethylcathinone by GC/MS (includes a stability study) (614); **2013** characterization of 31 synthetic cathinones (not identified in the abstract) by GC/MS, IR, and NMR (615); analysis of mephedrone, methylone, and MDPV by ambient ionization MS using arrays of low-temperature plasma probes, and also following injection of trifluoroacetic anhydride directly into the plasma stream for online derivatization (616); analysis of BMDP, butylone, MDPBP, MDPV, methylone, and pentylone by HPLC-HR-QTOF-MS (617); analysis of 38 cathinones (not specified in the abstract) by hybrid Q-TOF-MS and LC/MS/MS (618); an overview and review (619); analysis of (unspecified) "bath salt" cathinones by DART-MS (620); an overview and review of synthetic cathinones (621); analysis of 16 cathinones using "presumptive testing" (not specified in the abstract), TLC, and GC/MS (622); an overview of "bath salts" (including mephedrone, MDPV, and possibly others) (623); characterization of metaphedrone and pentedrone by single-crystal X-ray diffraction (624); analysis of 4-methylmethcathinone, three positional isomers of fluoromethcathinones, 4-methoxymethcathinone, N-ethylcathinone, N,N-dimethylcathinone, buphedrone, and pentedrone by GC/MS (625); a review of mephedrone, MDPV (and possibly others) (626); **2014** enantiomeric analysis of 10 new cathinones by CEC on a chiral stationary phase (627); identification of trace-levels of synthetic cathinones using Raman (cathinones not identified in the abstract) (628); analysis of 13 synthetic cathinones and associated psychoactive substances by ESI-high performance-IMS (629); identification of MDPV, 3,4-methylenedioxy- α -pyrrolidinobutiophenone (MDPBP), 4-fluoromethcathinone (4-FMC), butylone, mephedrone, naphyrone, 4-methylethcathinone (4-MEC), ethcathinone, α -pyrrolidinopentiophenone (α -PVP), and 3-methyl- α -pyrrolidinopropiophenone (3-MPPP) by GC/FID and GC/MS (630); screening and comparative analysis of synthetic cathinones by portable microchip electrophoresis (631); chiral separation of 12 cathinones by cyclodextrin-assisted CE with UV and MS detection (632); use of DART-MS in-source collision induced dissociation and high mass accuracy for determination of new psychoactive cathinones (633); screening for 16 cathinones by "presumptive testing", TLC, and GC/MS (634); electrochemical detection of (\pm)-methcathinone, (\pm)-mephedrone, and (\pm)-4'-methyl-N-ethylcathinone (635); electroanalytical sensing of mephedrone and methylethcathinone (636); synthesis and characterization of 9 new derivs. of cathinone (obtained by modifying the carbonyl group to create cyclic ketals and thioketals, oximes, and hydrazones of cathinone and of cathinone phthalimide) (analytical methodologies not identified in the abstract) (637); QSAR modelling of 4-methylbuphedrone and 4-methoxy-N,N-dimethylcathinone, with comparison to methylone (638); characterization of 4-fluoromethcathinone, ethcathinone, buphedrone, methedrone, pentedrone, 3,4-dimethylmethcathinone, 4-methylethcathinone, and others by FTIR, GC/MS, $^1\text{H-NMR}$, and wavelength dispersive XRF (639); **2015** analytical and synthetic studies on substituted cathinones (no details provided in the abstract) (640); analysis of methcathinone, 3,4-methylenedioxymethcathinone, 3,4-methylenedioxypropylvalerone, and 4'-methyl- α -pyrrolidinopropiophenone by LC/MS (641); isotopic profiling of cathinones for comparative analyses (642); identification and characterization of α -PVT, α -PBT, and their bromothieryl analogs (643); a review of the R- and S- isomers

of cathinones, focusing on MDPV (644); an overview and review of the neurotoxicity of the cathinones (645); identification and characterization of 4-fluoro-PV9 and α -PHP by HPLC, HPLC/DAD, ESI-Ion-Trap-MS in MS2 and MS3 modes, GC/MS, thermogravimetric anal., DSC, FTIR, UV/Vis, and NMR (646); compatibility of highly sulfated cyclodextrin with ESI at low nanoliter/minute flow rates and its application to CE-ESI/MS analysis of cathinone derivatives (647); the electrochemical detection of mephedrone (4-MMC) and 4'-methyl-N-ethylcathinone (4-MEC) (648); a study of the decomposition of the HCl salts of 8 cathinone derivatives in air (649); crystal structures of two forms of MDPV HCl and one form of ethylone HCl (650); preparation and characterization of the tertiary cathinones N,N-dimethylcathinone, N,N-diethylcathinone, and 2-(1-pyrrolidiny)-propiofenone by NMR and MS (the enantiomers were also prepared and identified by HPLC and CD (651); analysis of (\pm)-4'-methylmethcathinone and (\pm)-4'-methyl-N-ethylmethcathinone by HPLC/UV and amperometric detection ("NRG-2" is a focus) (652); **2016** differentiation of cyclic tertiary amine cathinone derivatives (the cyclic amines azetidine, pyrrolidine, piperidine, and azepane were incorporated into a series of cathinones related to MDPV) by product ion-EI-MS and MS/MS (653); thermal degradation of 4-ethylmethcathinone, 4-methylethcathinone, buphedrone, butylone, ethcathinone, ethylone, flephedrone, 3,4-methylenedioxy- α -pyrrolidinobutiophenone, 3,4-methylenedioxypropylone, mephedrone, methcathinone, methedrone, methylone, 4-methyl- α -pyrrolidinobutiophenone, naphyrone, pentedrone, pentylone and pyrovalerone under GC/MS conditions (654); identification of methylone and pentedrone by NMR, IR, UV/Vis, MS/MS, and HR-TOF-MS (655); identification and characterization of iso-4-BMC, β -TH-naphyrone, mexedrone, and 4-MDMC by LC-QTOF-MS, GC/MS, and NMR (656); chiral separation of new cathinones on chiral ion-exchange type stationary phases (657);

"Ecstasy Tablets" (that is, Tablets or Powders specified in their Titles or Abstracts as Ecstasy – these may in fact contain MDMA, a mixture of MDMA with one or more other Drugs, or only one or more non-MDMA Drugs): **2013** elemental analysis of Ecstasy tablets by graphite furnace atomic absorption, for comparative analysis (abstract indicates copper, magnesium, barium, nickel, chromium, and lead) (658); **2014** determination of metals (Zn, Al, Ca, Mg, K, Na, Ba, Fe, B, Cu, and Pt) in Ecstasy tablets using ICP-OES and XRF (659); **2015** a discussion of "luminescent" Ecstasy tablets (a marketing ploy) (660); detection of MDMA, methamphetamine, and 20 other substances in Ecstasy tablets, including caffeine, 2C-B, piperazines, amphetamines, and phencyclidine, by GC/MS (661); **2016** comparison of the purity and adulteration of the crystalline (powder) samples versus tablets in the Spanish Ecstasy market 2000-2014, by TLC, GC/MS, and UV (662);

Ephedrines: **2012** interconversion of ephedrine and pseudoephedrine during heptafluorobutyric anhydride derivatization (663); **2013** comparison of RP-UHPLC and HILIC for quantitation, with medium-resolution accurate MS (664); **2014** identification of ephedrine by use of charge-transfer complexes (with analysis of the complexes by elemental anal., IR, Raman, ^1H NMR, and UV-Vis (665);

Ergot Alkaloids: **2014** a review of the biosynthetic pathways of ergot alkaloids (666); detection of ergometrine, ergosine, ergotamine, ergocornine, ergocryptine, ergocristine) in rye and triticale grains (analytical methodologies not identified in the abstract) (667); determination of ergotamine tartrate in tablets using LC with fluorimetric and UV detection (668); an overview of the biosynthesis of the ergot alkaloids (669); identification of ergot alkaloid in two *Argyrea nervosa* “legal high” products by HPLC-HRMS/MS (670); a review of the detection of ergot alkaloid derivatives by TLC (671); aptamer-based extraction of ergot alkaloids from ergot contaminated rye feed (672); **2015** determination of ergot alkaloids in grain products by LC-ion trap-MS (673); an evaluation of fast dissolving tablets of ergotamine tartrate (674); determination of ergovaline in tall fescue seed and straw using a QuEChERS extraction method by HPLC with fluorescence detection (675); **2016** an overview and review (676); quantitative and qualitative transcriptome analysis of four industrial strains of *Claviceps purpurea* with respect to ergot alkaloid production (677); determination of ergot alkaloids in Morning Glory cultivars by LC-Q-TOF-MS (678); screening for total ergot alkaloids in rye flour by planar SPE-fluorescence detection and MS (679);

Fentanyl Derivatives: **2014** analysis of the inclusion complexes between cyclodextrins and fentanyls by NMR and computational studies (680); an efficient, optimized synthesis of fentanyl and related analogs (681); **2015** improved and optimized syntheses of fentanyl and related analogs (682);

2-, 3-, and 4-Fluorophenmetrazines: **2016** synthesis, characterization, and differentiation of the fluorophenmetrazine isomers (683);

“FLY” Compounds: **2014** synthesis of labeled 2C-B-FLY and Bromo-DragonFLY for use as internal standards (684);

Methiopropamine (and its 3-thienyl isomer): **2013** synthesis and analysis/differentiation by GC (685);

NBOMe Compounds: **2013** characterization of 25D-NBOMe [2-(2,5-dimethoxy-4-methylphenyl)-N-(2-methoxybenzyl)ethanamine], 25E-NBOMe [2-(4-ethyl-2,5-dimethoxyphenyl)-N-(2-methoxybenzyl)ethanamine], and 25G-NBOMe [2-(2,5-dimethoxy-3,4-dimethylphenyl)-N-(2-methoxybenzyl)ethanamine] (686); **2014** an overview and review (687); **2015** a review (688); detection of NBOMe's (and other NPSs) on blotter papers by direct ATR-FTIR (689); analysis of 25I-NBOMe, 25B-NBOMe, 25C-NBOMe and other dimethoxyphenyl-n-[(2-methoxyphenyl)methyl]ethanamine derivatives on blotter paper by DART-AccuTOF-MS and HPLC-triple quadrupole-MS (690); an overview (691);

Opiates: **2012** determination of morphine and codeine by HPLC-quadrupole mass-selective detection (may be a toxicological study) (692); **2013** analysis of morphine and codeine by TLC and densitometry (693); **2014** some insights into hydrate formation and stability of morphinanes by powder X-ray diffraction, IR, DSC, and isothermal calorimetry (694); isomerization of codeine and morphine into hydrocodone and hydromorphone using a water-sol. rhodium complex formed from

com. available [Rh(COD)(CH₃CN)₂]BF₄ and 1,3,5-triaza-7-phosphaadamantane (695); a review of the TLC of morphine analogs (compounds not identified in the abstract) (696); **2015** potential use of oriental poppy hairy roots for producing thebaine, morphine, and codeine (697); a review covering the synthesis of buprenorphine, naltrexone, naloxone, and nalbuphine from naturally occurring opiates such as thebaine and oripavine (698); the stereochemistry and spectral assignment of thebaine derivatives based on a 1D NOESY NMR study (699); degradation of morphine and codeine by gamma radiation in methanol (700); radiation induced destruction of thebaine, papaverine, and noscapine in methanol (701); a review of AH-7921 (702); separation of morphine, hydromorphone, and norcodeine using ESI and paper spray coupled to high-field asymmetric waveform IMS (703);

Opiates (Bio-Engineered): **2014** use of a microbial biomanufacturing platform for natural and semisynthetic opioids, using *Saccharomyces cerevisiae* (704); **2015** heroin from bio-engineered yeast (705); heroin from bio-engineered yeast (706); failure of an attempted large-scale effort to produce thebaine using home-brew type conditions (707); synthesis of morphinan alkaloids from norlaudanosoline using *Saccharomyces cerevisiae* (708); a feasibility study for production of thebaine and hydrocodone from sugar by bio-engineered yeast (709); a review, detailing the current status of microbial benzyloquinoline alkaloid synthesis and derivatization (710); a call to regulate the synthesis of morphine by bio-engineered yeasts (711); **2016** metabolic engineering for the production of plant isoquinoline alkaloids (712); complete biosynthesis of opioids (thebaine) by yeast (713); a review of the production of thebaine and hydrocodone from D-glucose by fermentation (714); total biosynthesis of opiates (thebaine) by stepwise fermentation using engineered *E. coli* (715);

1-(1-Phenylcyclohexyl)piperidine (PCP) and 1-(1-phenylcyclohexyl)pyrrolidine (PCPy) analogues: **2014** characterization by GC-ion trap-EI-, CI-, and HR-MS, LC-ESI-triple-quadrupole linear ion trap-MS/MS, IR, DAD, and ¹H and ¹³C NMR (716);

Phenothiazines: **2013** separation and identification of prochlorperazine, promethazine, chlorpromazine, and trifluoroperazine (717);

Phosphodiesterase-5 Inhibitors – Cialis (tadalafil), Levitra (vardenafil), Viagra (sildenafil), and similar drugs: **2013** a multivariate-based wavenumber selection method for classifying Cialis and Viagra into authentic or counterfeit classes by ATR/FTIR (718); analysis for residual solvents in counterfeit tablets and capsules of Cialis and Viagra (analytical method not indicated in the abstract) (719); simultaneous qualitative and quantitative analysis of counterfeit Cialis by Raman (720); analysis of 38 compounds (sildenafil, tadalafil, vardenafil and their analogs) in illicit erectile dysfunction products by LC-ESI-MS/MS (721); differentiation between counterfeit and authentic Cialis and Viagra by ATR/FTIR with PCA (722); analysis and profiling by UPLC/MS (723); characterization of sildenafil citrate tablets from different sources by NIR chemical imaging and chemometric tools (724); **2014** profiling authentic and counterfeit Viagra and Cialis using XRF, direct infusion ESI-MS, UPLC-MS, and ATR-FTIR (725); simultaneous determination of of sildenafil,

tadalafil, vardenafil and acetildenafil in health-care foodstuffs by UHPLC/MS (726); qualitative and quantitative analysis of sildenafil in traditional medicines and dietary supplements by HPLC/UV and IR (727); **2015** isolation and structural characterization of chloropropanoylpretadalafil in a dietary supplement by HPLC-UV, GC/FT-IR/MS, and HRMS (728); detection of sildenafil citrate in herbal formulations by UV/Vis (729); differentiating genuine and counterfeit Viagra tablets by dynamic thermal analysis (730); **2016** use of transmission-mode desorption electrospray MS-MS to screen for synthetic phosphodiesterase-5 inhibitors in samples of adulterated herbal dietary supplements (731); analysis of dietary supplements containing phosphodiesterase type-5 (PDE-5) inhibitors by LC/MS and HPLC/UV (732);

Piperazines: **2012** differentiation of methylenedioxybenzylpiperazines and ethoxybenzylpiperazines by GC/IRD and GC/MS (733); **2013** characterization of six ring regioisomeric dimethoxybenzoylpiperazines (DMBzPs) by GC/MS and GC/IRD (734); analysis of the six-ring regioisomeric dimethoxybenzyl-N-methylpiperazines (DMBMPs) by GC/MS (735); a presumptive color spot test method for the detection of benzylpiperazine and piperazine analogues (736); determination of chlorophenylpiperazine isomers by CE (737); analysis of phenyl and benzyl piperazines by HPLC with chemiluminescence detection (738); **2014** six ring regioisomeric dimethoxybenzoyl-N-methylpiperazines (DMBzMPs) by GC/MS and IR (739); analysis of regioisomeric bromodimethoxy benzyl piperazines related to 4-bromo-2,5-dimethoxybenzylpiperazine by GC/MS and FTIR (740); differentiation of the 1-(methylenedioxyphenyl)-2-piperazinopropanes and 1-(methoxyphenyl)-2-piperazinopropanones by GC/IRD and GC/MS (741); **2015** analysis of six ring regioisomeric dimethoxyphenylpiperazines (DOMePPs) by GC/MS and IR (742); analysis of 23 benzylpiperazine (BZP) and trifluoromethylphenylpiperazine (TFMPP) containing tablets by HPLC and IRMS (743); an overview of 1-cyclohexyl-4-(1,2-diphenylethyl)piperazine (MT-45) (744);

Steroids: **2013** determination of tetrahydrogestrinone and related anabolic androgenic steroids by MEKC (745); a study of authentic and counterfeit products (primarily stanozolol, testosterone, and nandrolone) seized in Brazil from 2006 to 2011 (746); analysis of methandienone and methyltestosterone in tablets by color testing and GC/MS (747); a review of the bioanalytical challenges in detecting unknown anabolic androgenic steroids (in doping control analysis) (748); screening for steroids in traditional medicine and nutraceutical products using electrospun cellulose acetate nanofibers as thin layer chromatographic media (749); **2015** analysis of anabolic steroids by GC-EI/MS, GC-EI/MS/MS, LC-ESI/MS/MS, LC-Ag+CIS/MS/MS, and GC-ESI/MS/MS (for doping control) (750); determination of anabolic-androgenic steroid adulterants in counterfeit drugs by UHPLC-MS/MS (751); identification and quantification of anabolic steroid esters by DART-HRMS (752); an overview and review of the anabolic androgenic steroids in supplements (753); determination of anabolic agents in dietary supplements by LC-HRMS (754); a summary of the designer steroids that are most commonly sold in dietary supplements (as of Apr. 2014) (755); **2016** improved detection of steroids and evidence for their regiospecific decompositions using anion attachment MS (756); analysis of steroids in dietary supplements by non-targeted mass spectrometry (757); analysis of anabolic steroids by GC-CI-TQuad-MS (758);

Tryptamines (see also Mushrooms): 2013 characterization of AMT (3-(2-aminopropyl)indole) and 5-IT (5-(2-aminopropyl)indole) by ¹H- and ¹³C-NMR, GC-EI/CI-ion trap-MS, U/HPLC-DAD, and HPLC/MS (759); simultaneous determination of tryptamine analogues in designer drugs using GC/MS and LC-MS/MS (only 5-methoxy-N,N-diethyltryptamine and 5-methoxy-N-methyl-N-isopropyltryptamine were identified in the abstract, among many more) (760); 2015 a review of the use, analysis, and toxicity of tryptamines (only DMT is specifically noted in the abstract) (761); 2016 synthesis of psilocin, bufotenin, serotonin, and various homologues and branched tryptamine derivatives (762); characterization of N,N-diallyltryptamine (DALT), and 2-phenyl-, 4-acetoxy-, 4-hydroxy-, 4,5-ethylenedioxy-, 5-methyl-, 5-methoxy-, 5-methoxy-2-methyl-, 5-ethoxy-, 5-fluoro-, 5-fluoro-2-methyl-, 5-chloro-, 5-bromo-, 5,6-methylenedioxy-, 6-fluoro-, 7-Me, and 7-ethyl DALTs, by NMR, GC/MS, EI/MS, low and high mass accuracy MS/MS, PDA, and GC solid-state IR (763).

1.D - Synthetic Cannabinoids and Cannabimimetics [Notes: Compounds are listed either by their acronym or full name as was specified in their respective abstract – no effort was made to transcribe acronyms to full chemical names or vice versa. Articles that include both synthetic cannabinoids and/or cannabimimetics with other drugs are detailed separately.]

Individual Synthetic Cannabinoids and Cannabimimetics: 2013 identification of (1-(cyclohexylmethyl)-1H-indol-3-yl)(4-methoxynaphthalen-1-yl)methanone by LC/MS and NMR (764); purification and characterization of 3-methyl-6-[3-(trifluoromethyl)-phenyl]-1,2,4-triazolo[4,3-b]pyridazinol (CL 218872) by MS, IR, and NMR (765); characterization of JWH-213 by LC-PDA-MS, GC/MS, high-res MS, and NMR (766); analysis of N-[3-(2-methoxyethyl)-4,5-dimethyl-2(3H)-thiazolylidene]-2,2,3,3-tetramethylcyclopropanecarboxamide (A-836339) by LC/MS, GC/MS, high-res MS, NMR, and X-ray crystallography (767); identification of [1-(tetrahydropyran-4-ylmethyl)-1H-indol-3-yl]-(2,2,3,3-tetramethylcyclopropyl)methanone (A-834,735) by LC-ESI-QTOFMS, GC/MS, 1D- and 2D-NMR, and FTIR (768); 2014 an outbreak of exposure to a novel synthetic cannabinoid (abstract not available) (769); analysis of methyl 2-[[1-(5-fluoropentyl)-3-methyl-1H-indol-3-ylcarbonyl]amino]butyrate (770); structural elucidation of a new open chain isomer of the cannabimimetic cyclopropoylindole A-796,260 by NMR and MS (771); determination of HU-210 by HPLC (772); identification of JWH-018 by LC-MS/MS (773); 2015 isolation and identification of AB-FUBINACA (774); structural elucidation of N-(1-amino-3,3-dimethyl-1-oxobutan-2-yl)-1-(5-fluoropentyl)-3-(4-fluorophenyl)-pyrazole-5-carboxamide (a homolog of AZ-037) by NMR and MS (775); characterization of naphth-1-yl 1-(5-fluoropentyl)-1H-indole-3-carboxylate (CBL-2201) by ¹H, ¹³C, and ¹⁵N NMR, FTIR, and GC/MS (776); new monoclonal antibodies specific for 1-(5-fluoropentyl)-3-(2-iodobenzoyl)indole (AM694) (777); identification of N,N-bis(1-pentylindol-3-yl-carboxy)naphthylamine (BiPICANA) by LC/MS, HRMS, NMR, and X-ray crystallography (778); analysis of AB-CHFUPYCA [N-(1-amino-3-methyl-1-oxobutan-2-yl)-1-(cyclohexylmethyl)-3-(4-fluorophenyl)-1H-pyrazole-5-carboxamide] by GC/MS, LC/MS, LC/HRMS, and NMR (779); 2016 determination of the absolute

configuration of MDMB-CHMICA by vibrational and electronic CD spectroscopy, X-ray crystallog., and HPLC (780); separation and structural characterization of JWH-018-cyclohexyl methyl derivative (NE-CHMIMO) by flash chromatography, GC/MS, IR, and NMR (781); analysis of 3-benzyl-5-[1-(2-pyrrolidin-1-ylethyl)-1H-indol-3-yl]-1,2,4-oxadiazole by GC/MS, GC/HRMS, UHPLC/HRMS2, FTIR, and 1H and 13C NMR (782);

Multiple Synthetic Cannabinoids and Cannabimimetics:

[Note: Each year in this subsection is separated by a line space.]

2012 separation and structural characterization of JWH-412 and 1-[(5-fluoropentyl)-1H-indol-3yl]-(4-methylnaphthalen-1-yl)methanone using GC/MS, NMR, and flash chromatography (783); analysis of cannabinoids by IR, GC/MS, LC/MS, and 1H NMR (784); analysis of CP-47,497-C8 JWH-250, and RCS-4 by TLC, GC/MS, light-optical microscopy, and “phytochemical reactions” (785);

2013 analysis of JWH-018, JWH-019, JWH-073, and JWH-250 by GC/MS (786); analysis of 5F-UR-144 and UR-144 by GC/MS, LC-TOF-MS, and 1D- and 2D-NMR (787); an overview of synthetic cannabinoids in South Korea from 2009 to June 2013 (788); analysis of AM-2201, JWH-203, JWH-210 and RCS-4 by LC, high-res MS, LC-QTOF-MS, and NMR (789); correlated results from the analyses of synthetic cannabinoids in Turkey from 2010 to 2012 (790); analysis of JWH-019, JWH-081, JWH-203, and JWH-250 by UHPLC-QTOF-MS (791); analysis of 28 (unspecified) “synthetic cannabinoids” by LC/ESI-MS/MS (toxicological focus) (792); isolation of cis- and trans- CP-47,497-C8 (and others not specified in the abstract) – extraction from plant materials by flash chromatography (793); analysis of azepane isomers of AM-1220 and AM-2233, AM-2233, and URB-597 by LC/MS, GC/MS, “accurate MS,” and NMR (794); isolation and analysis of 1-butyl-3-(2-methoxybenzoyl)indole and the 2-methoxy isomer of RCS-4 by column chromatography and prep-HPLC, followed by GC/MS, ESI-TOFMS, and 1D- and 2D-NMR (795); a review of the analysis of synthetic cannabinoids on botanical materials (796); analysis of unspecified “cannabimimetics” bearing 2,2,3,3-tetramethylcyclopropanecarbonyl moieties by GC/MS, LC/MS, and NMR (797); characterization of some synthetic cannabinoids, derivatives of indole-3-carboxylic acid, by GC-HRMS, UHPLC-HRMS, NMR, and FTIR (798); detection of AB-001, AM-2232, APINACA, N,5-dimethyl-N-(1-oxo-1-(p-tolyl)butan-2-yl)-2-(N'-(p-tolyl)ureido)benzamide, (4-ethylnaphtyl)-AM-2201 (EAM-2201), 5-fluoropentyl-3-pyridinoylindole, 5FUR-144 (synonym: XLR11), 4-hydroxydiethyltryptamine (4-OH-DET), JWH-213, JWH-307, JWH-030, 4-methylbuphedrone, (4-methylnaphthyl)-AM-2201 (MAM-2201), (4-methylnaphtyl)-JWH-022 [synonym: N-(5-fluoropentyl)-JWH-122], N-(4-pentenyl)-JWH-122, UR-144, and URB-754 on plant materials (methods not specified in the abstract) (799); analysis of N-(1-amino-3-methyl-1-oxobutan-2-yl)-1-pentyl-1H-indazole-3-carboxamide (AB-PINACA) and N-(1-amino-3-methyl-1-oxobutan-2-yl)-1-(4-fluorobenzyl)-1H-indazole-3-carboxamide (AB-FUBINACA) by LC/MS, GC/MS, high-res MS, and NMR (800); a pharmacological study of the structural features of synthetic cannabinoids and their in vivo cannabimimetic activity (801); simultaneous determination of JWH-018 and JWH-073 by UFLC (Ultra-Fast LC) (802); analysis of cannabicyclohexanol, JWH-

018, JWH-073, JWH-081, JWH-122, JWH-210, JWH-250, and RCS-4 by GC/MS, LC-QTOF-MS, and HPLC (803);

2014 an overview of the emergence, identification, legislation and metabolic characterization of synthetic cannabinoids in herbal incense products (804); chromatographic and mass spectral studies on 6 1-pentyl-acylindoles (regioisomeric synthetic cannabinoids) (805); analysis and differentiation of substituted 1-alkyl-3-acylindoles (isomeric synthetic cannabinoids) by GC-MS, IR, and some exact mass GC-TOF-MS (806); differentiation of 1-alkyl-3-acylindoles and 1-acyl-3-alkylindoles (isomeric synthetic cannabinoids) by GC MS, and IR (807); a review (808); differences in the GC-EI-MS spectra of JWH-250, JWH-302, and JWH-201 (809); presumptive color-testing of synthetic cannabimimetics by Duquenois-Levine, van Urk, and 2,4-DNPH (810); analysis of AM-2201, JWH-122, JWH-203, JWH-210, and RCS-4 by DART-MS (811); identification and quantification of synthetic cannabinoids by GC/MS and GC/ECD (812); synthesis and biological activities of synthetic cannabinoids (813); structural elucidation, analytical characterization, and identification of [1-(5-fluoropentyl)-1H-indazol-3-yl(naphthalen-1-yl)methanone, naphthalen-1-yl(1-pentyl-1H-benzo[d]imidazol-2-yl)methanone, and 1-(5-fluoropentyl)-1H-benzo[d]imidazol-2-yl(naphthalen-1-yl)methanone by GC/MS, GC/HR-MS, UHPLC-HR-MS, NMR, and FT-IR (814); identification and analysis of indol-3-carboxylates series and indazole-3-carboxylates (novel cannabinoids) by GC/MS, GC-HRMS, UHPLC-HRMS, NMR, and FTIR (815); analysis of the 6 benzoyl-substituted-1-pentylindoles (isomeric synthetic cannabinoids) by GC/MS and FTIR (816); simultaneous determination of 10 synthetic cannabinoids by HPLC (817);

2015 a retrospective survey of synthetic cannabimimetics in Bulgaria 2010-2013 (818); synthesis and SARs of RCS-4 and its regioisomers and C4 homologue (819); identification of 8-quinolinyl 4-methyl-3-(1-piperidinylsulfonyl)benzoate (QMPSB), MAM-1220, and CHM-081 by GC/MS, LC/MS, and NMR (820); synthesis and spectroscopic analysis of analogues of 1H-indol-3-yl(2,2,3,3-tetramethylcyclopropyl)methanone and 1H-indol-3-yl(adamantan-1-yl)methanone by NMR, MS, FTIR, and GC-FTIR (821); quantitation of 32 synthetic cannabinoids (dibenzopyrans, cyclohexylphenols, naphthoylindoles, benzoylindoles, phenylacetylindoles, tetramethylcyclopropylindoles) on plant materials by a validated HPLC/UV method (822); QSARs of 43 cannabimimetic aminoalkylindole derivatives and their metabolites (823); qualitative and quantitative analysis of 2 fluorine containing cannabinoids (XLR-11 and AM-2201) by ¹⁹F-NMR, with comparison against GC/MS (824); the variability of active ingredients in Spice within Alaska as an indicator mechanism for manufacture and distribution (825); rapid screening and quantification of synthetic cannabinoids in herbal products with COSY and TOCSY NMR (826); separation of cannabinoids on 3 different mixed-mode columns (827); an overview and review of synthetic cannabinoids (828); differentiation of the positional isomers of JWH-081 by GC-EI-MS and GC-MS/MS (829); identification and quantification of 5-fluoro-AB-PINACA, AB-CHMINACA, AB-FUBINACA, 5-fluoro-PB-22, 5-fluoro-AMB, MDMB-CHMICA, EAM-2201, and STS-135 by GC/MS (830); identification of synthetic cannabinoids by UHPLC-TOFMS and GC/MS (among 32 solutes, only JWH-018 and CP47,497 are identified in the abstract) (831); synthesis

and characterization of N-(1-amino-3-methyl-1-oxobutan-2-yl)-1-(cyclohexylmethyl)-3-(4-fluorophenyl)-1H-pyrazole-5-carboxamide (3,5-AB-CHMFUPPYCA) and differentiation from its 5,3-regioisomer (832); analysis of ADB-BINACA, AB-FUBICA, ADB-FUBICA, and AB-BICA by LC-HRMS, GC/MS, and NMR (833); identification and analytical characteristics of 5 new synthetic cannabinoids with an indazole-3-carboxamide structure bearing an N-1-methoxycarbonylalkyl group by GC/MS, GC/HRMS, UHPLC-HR-MS/MS, and ¹H and ¹³C NMR (834); a review of synthetic cannabinoids (835); analysis of 1-n-pentyl-3-(1-naphthoyl)indole (JWH-018), three deuterium-labeled analogues, and the inverse isomer 1-naphthoyl-3-n-pentylindole by MS (836); analysis of JWH-018 and its 5 regioisomers by GC/MS (837); separation and detection of cannabicyclohexanol (CCH: cis-isomer), trans-CCH, 5-(1,1-dimethylheptyl)-2-[(1R,3S)-3-hydroxycyclohexyl]-phenol (CP-47497), 5-(1,1-dimethylheptyl)-2-[(1R,2R,5R)-5-hydroxy-2-(3-hydroxypropyl)-cyclohexyl]-phenol (CP-55940), 3-(1,1'-dimethylheptyl)-6aR,7,10,10aR-tetrahydro-1-hydroxy-6,6-dimethyl-6H-dibenzo[b,d]pyran-9-methanol (HU-210), 2-[1R-3-methyl-6R-(1-methylethenyl)-2-cyclohexen-1-yl]-5-pentyl-1,3-benzenediol (CBD), (1-pentyl-1H-indol-3-yl)-1-naphthalenyl-methanone (JWH-018), (1-butyl-1H-indol-3-yl)-1-naphthalenyl-methanone (JWH-073) and 1-(1-pentyl-1H-indol-3-yl)-2-(2-methoxyphenyl)-ethanone (JWH-250) by SFC/MS (838); an overview of illnesses and deaths from abuse of synthetic cannabinoids (839); syntheses and analytical characterizations of 15 N-alkyl-arylcyclohexylamines by GC and HPLC coupled to multiple forms of mass spectrometry, as well as NMR, UV/DAD, and IR (840); characterization of 2 thiazolylindoles and a benzimidazole (potential cannabinoids) by MS, IR, and NMR (841); a review of bioisosteric fluorine in the clandestine design of synthetic cannabinoids (842); identification and quantitation of 5-fluoro-ADB-PINACA and MAB-CHMINACA by HRMS, GC/MS, and LC-MS/MS (843); a study on the fragmentation pathways of JWH-018 and JWH-073 (844); determination and identification of synthetic cannabinoids and their metabolites in different matrices by chromatographic, spectroscopic, and spectrometric methods (845);

2016 differentiation of the 6 regioisomeric dimethoxybenzoyl-1-pentylindoles by EI-MS and FT-IR (846); a study of the fragmentation of 21 synthetic cannabinoids with an iso-Pr group or a tert-Bu group by EI-Quad-MS and positive ESI-TOF-MS (847); analysis of 22 synthetic cannabinoids, and separately of JWH018 and 9 of its positional isomers, by ultra high performance SFC (848); variation in commercial “smoking mixtures” containing third-generation synthetic cannabinoids (849); identification of 6 synthetic cannabinoids by DART - LTQ ORBITRAP (850); identification of APINACA 2H-indazole analogue, AMPPPCA, and 5F-AMPPPCA by LC-QTOF-MS, GC-TOF-MS, and NMR (851); differentiation of JWH-122 and JWH-210 by GC-EI-MS/MS (852); analysis of 5F-AMB and PX-3 by ¹H and ¹³C NMR, HR-MS/MS, and Raman (853); an overview and review of recent international trends in Spice use (854); analysis of the 2-alkyl-2H-indazole regioisomers of synthetic cannabinoids AB-CHMINACA, AB-FUBINACA, AB-PINACA, and 5F-AB-PINACA (possible manufacturing impurities with cannabimimetic activities) by ¹H and ¹³C NMR, GC/MS, and UV/Vis (855); rapid identification of 10 synthetic cannabinoids by DART-MS and NMR (856); use of a QSAR model to determine the affinity of synthetic cannabinoids to the CB1 receptor (857); identification and characterization of ADB-BICA, NNL-1, NNL-2, and PPA(N)-2201 by LC-QTOF-MS, GC/MS, FTIR,

and NMR (858); determination of 8 synthetic cannabinoids by heat assisted sample introduction and dielectric barrier discharge ionization MS (859);

Synthetic Cannabinoids and Cannabimimetics with Other Drugs (except when a minor part of a larger study): **2012** identification of atropine, scopolamine, lysergamide mitragynine, 4-methoxymethcathinone, 3-fluoromethacathinone, JWH-073, JWH-081, JWH-0250, and JWH-0251 in “herbal products” purchased via the Internet in 2009 and 2010 by LC/PDA/MS and GC/MS (860); analysis of CP-47,497 CP-47,497-C8, JWH-018, JWH-073, JWH-200, MDPV, mephedrone, and methylone by UHPLC/TOFMS (861); **2013** a review, including a comparison of the natural and synthetic cannabinoid materials (862); identification of ADB-FUBINACA, ADBICA, AM-2201 4-methoxynaphthyl analog, APICA N-(5-fluoropentyl) analog, APINACA N-(5-fluoropentyl) analog, JWH-122 N-(5-chloropentyl) analog, QUPIC, QUCHIC, and UR-144; N-(5-chloropentyl) analog (alpha-pyrrolidinovalerothiophenone (alpha-PVT) and 3,4-dichloro-N-([1-(dimethylamino)cyclohexyl]methyl)benzamide (AH-7921) also identified) (863); an overview of Psilocybe mushrooms, 5MeO-DIPT, tryptamine, MDMA and related compounds, synthetic cannabinoids, and cannabimimetics (864); **2014** analysis of piperazine derivatives (BZP, MPMP, TFMPP), cathinone derivatives (N-ethylcathinone, buthylone, ethylone, methylone, buphedrone, flephedrone), pyrovalerone derivatives (MDPV, naphyrone), and synthetic cannabinoids (AM-694, JWH-019, JWH-073, JWH-081, JWH-122, JWH-200, JWH-250), by GC-EI-MS (865); determination of AM-2201, JWH-018, JWH-022 JWH-073, JWH-122, JWH-203, JWH-210, JWH-250, HU-210, RCS-4, THC, and various metabolites by UHPLC-MS/MS (866); analysis of cocaine, methylone, 4'-methylethcathinone, 3,4-MDPV, JWH-210, JWH-250, and JWH-203 by ion mobility-TOF-MS (867); analysis of a mixture of diphenidine and 5-fluoro-AB-PINACA (868); **2015** an overview of cannabis vs. synthetic cannabinoids (869); an overview of synthetic cathinones and cannabinoids (870); a review of a major researcher's 50 years of research on cannabinoids, with future-looking comments (871); analysis of synthetic cathinones and cannabimimetic agents by MS, LC/MS, LC-MS/MS, NMR, IR, and DART-MS (872).

1.E – Polydrug A: Mixed or Unrelated Individually Named Compounds or Substances

[Note: Each year in this subsection is separated by a line space.]

2012 analysis of cocaine, heroin, and MDMA by spectral fluorescence (873); use of a modified multiwall carbon nanotubes paste electrode for simultaneous voltammetric determination of morphine and diclofenac in biological and pharmaceutical samples (874); an extended overview and review of “date-rape” drugs (GHB, MDMA, flunitrazepam, and ketamine) (875);

2013 detection of flunitrazepam, ketamine, and MDMA by IMS (toxicological focus) (876); analysis of methoxetamine, 3-methoxyeticyclidine, and 3-methoxyphencyclidine by GC- and CI- MS, NMR, and HPLC-DAD-ESI-MS/MS

(toxicological focus) (877); identification of 1,4-benzobenzodiazepines (clonazepam, flurazepam, alprazolam, midazolam, bromazepam, chlordiazepoxide, lorazepam, and diazepam) and antidepressants (bupropion, sertraline, paroxetine, and fluoxetine) as adulterants in phytotherapeutic dieting formulations by voltammetry (878); differentiation of anorexics (amfepramone, fenproporex, sibutramine), benzodiazepinic anxiolytics (clonazepam, flurazepam, alprazolam, midazolam, medazepam, chlordiazepoxide, diazepam), antidepressants (bupropione, fluoxetine, sertraline, paroxetine), diuretics (hydrochlorothiazide, furosemide, chlortalidone, amiloride, spironolactone), and hypoglycemics (glimepiride, chlorpropamide, glibenclamide) by a solid state electrochemical method (879); analysis of tramadol and morphine by spectrofluorimetry and spectrophotometry (880); determination of morphine, nalbuphine, and "naltrexone drugs" in bulk and pharmaceutical formulations by a kinetic spectrophotometric method (881); determination of tramadol, morphine, nalbuphine and naltrexone analgesic drugs using potassium permanganate and spectrophotometry (882); determination of 13 sedative-hypnotics in health foods (including phenobarbital, estazolam, and diazepam) by HPLC-MS/MS (883); detection of lidocaine, diazepam, and ketamine as adulteration in foodstuffs and beverages by HPLC (884); analysis of methaqualone, saccharin, paracetamol, and phenacetin in illicit drugs by HPLC (885); an overview of the analyses of BZP, mephedrone, JWH-018, TFMPP, sage poet, kratom, fly agaric, kava-kava, and others (886); determination of 4 cathinones (mephedrone, butylone, 4-Me-PPP, and 4-MEC) and 5 tryptamines (5-EtO-DPT, 5-EtO-DALT, 5-EtO-MIPT, 5-EtO-ALCHT, and 5-EtO-2MALET) by ESI-AP-Ion Mobility-TOF-MS (887); identification of kratom, 2C-C-NBOMe, 25I-NBOMe, RH-34 and UR-144, 2-(2,3-dimethoxyphenyl)-N-(3,4,5-trimethoxybenzyl)ethanamine (DMA-NBTOMe), acetylated 25I-NBOMe, acetylated DMA-NBTOMe by GC/MS and NMR (888); analysis of barbital, clozapine, chlordiazepoxide, midazolam maleate, phenobarbital, perphenazine, promethazine HCl, chlormezanone, nitrazepam, amobarbital, oxazepam, secobarbital sodium, estazolam, lorazepam, clonazepam, alprazolam, diazepam, and triazolam by UHPLC with PDA detection (889); analysis of alprazolam, estazolam, clonazepam, diazepam, phenobarbital, midazolam maleate, triazolam, nitrazepam, barbital, secobarbital, chlordiazepoxide, lorazepam, amobarbital, and oxazepam by UHPLC with PDA detection (890); analysis of mephedrone, 5,6-methylenedioxy-2-aminoindane (MDAI), and MDMA by SERS on copper coins coated with deposited silver (891); detection of 6 chemical constituents illegally added into health foods for dieting by UPLC-MS/MS (only sibutramine HCl and phenolphthalein were identified in the abstract) (892); identification of undeclared synthetic drugs (ranitidine, orphenadrine citrate, piroxicam, and dexamethasone) in medicines illegally sold as phytotherapies by diffusion-ordered NMR spectroscopy and HPLC-UV-SPE-NMR (893); the long-term stability of 4-MEC, MDAI, methoxetamine, 5-MeO-DALT, 6-APB, MPA, 5-IAI, MDAT, 2-AI, AMT, 25C-NBOMe, AH-7921, 5-MAPB in blood and plasma, as determined by HPLC/DAD, LC-MS/MS, and UHPLC-Q-TOF-MS (894); determination of dextromethorphan and levomethorphan in heroin by enantioselective HPLC and electronic CD (895); identification of sibutramine HCl, fenfluramine HCl, phenolphthalein, strychnine, ephedrine HCl, and hydrochlorothiazide in health foods with weight reducing properties by TLC and HPLC-MS/MS (896); a survey of 449 "legal highs" seized in Poland between mid-2008 and mid-2011 (including MPDV,

caffeine, butylone, TFMPP, lidocaine, 4-MEC, mephedrone, pFPP, BZP, and MDPBP, and others) (897);

2014 analysis of 4-fluoroamphetamine, methiopropamine, ethcathinone, 4-methylethcathinone, N-ethylbuphedrone, ethylphenidate, 5-MeO-DALT, dimethocaine, 5-(2-aminopropyl)benzofuran, and nitracaine by a Selective Reagent Ionisation-TOFMS (898); trends in Irish street-level heroin and cocaine 2010-2012 (899); terahertz detection of ketamine and ATs (900); an overview of the presence of mephedrone, 4-methylethcathinone, BZP, MDPV, TFMPP, methoxetamine, 4-fluoromethcathinone, 4-methylamphetamine, PMA, methylone, PMMA, naphyrone, alpha-methyltryptamine, butylone, MDAI, desoxypipradrol, D2PM, MPA, synthetic cannabinoids, 2-AI, 5-IAI, 5-MeODALT, MDPBP, 5/6-APB, pentedrone, and pentylone in post-mortem and criminal casework (toxicological focus) (901); analysis of 2-aminopropyl-benzofuran with 4 potential positional isomers, methiopropamine, and 2-(ethylamino)-1-(4-methylphenyl)pentan-1-one by GC/MS and NMR (902); analysis of alprazolam and fluoxetine by UV/Vis (903); a review on detecting residues of chlorpromazine and diazepam in foods (904); identification of ephedrine, caffeine, furosemide, fenfluramine, phenolphthalein, sibutramine, N-desmethyl sibutramine, and N-didesmethyl sibutramine in weight controlling health food by UHPLC/DAD (905); analysis of bromazepam, flunitrazepam, fluoxetine hydrochloride, clozapine, and risperidone by TLC (906); analysis of cocaine, LSD, levamisole, papaverine, and others by MALDI-HRMS, HPLC/DAD, and Quad-MS (907); analysis of cocaine, heroin, methamphetamine, oxycodone, and amphetamine on currency by LC/MS (908); syntheses, characterization, and in vitro metabolism of nitracaine, methoxypiperamide and mephtramine (909); analysis of MDMA and mCPP by CE (910); determination of the stability in solution of 4-MEC, MDAI, methoxetamine, 5-MeO-DALT, 6-APB, MPA, 5-IAI, MDAT, 2-AI, AMT, 25C-NBOMe, AH-7921, and 5-MAPB by HPLC-DAD, LC-MS/MS, and UHPLC-Q-TOF-MS (911)So; analysis of amphetamine, methamphetamine, MDMA, N,N-dimethylamphetamine, PMA, PMMA, BZP, TFMPP, mCPP, and MeOP by DESI-MS (912); analysis of 3-methylmethcathinone, methylone, butylone, 4-methylethcathinone, flephedrone, methylenedioxypropylvalerone, pentedrone, methoxetamine, APINACA, AKB48, benzydamine, meta-chlorophenylpiperazine, 5-MeO-DALT, 5-MeOMIPT, 6-APB, 4-APB, diphenidine, and others, by single quadrupole GC/MS, positive ESI- LC/HRMS, and NMR (913); determination of lidocaine, ketamine, and diazepam in foodstuffs using micellar LC (914); analysis of amphetamine, methamphetamine, caffeine, paracetamol, and theophylline by HPLC (915); identification of the piperazine derivative MT-45 (I-C6), the synthetic peptide Noopept (GVS-111), the synthetic cannabinoid A-834735, 4-methoxy- α -PVP, and 4-methylbuphedrine (analytical methodologies not provided in the abstract) (916); analysis of FUB-PB-22, 5-fluoro-NNEI indazole analog (5-fluoro-MN-18), AM-2201 indazole analog (THJ-2201), XLR-12, 5-fluoro-AB-PINACA, 5-chloro-AB-PINACA, AB-CHMINACA, and 5-fluoro-AMB; DL-4662, α -PHP, 4-methoxy- α -POP, 4-methoxy- α -PHPP, and 4-fluoro- α -PHPP; 2-(2-ethylaminopropyl)-benzofuran (2-EAPB), nitracaine, diclofensine, diphenidine, 1-benzylpiperidine, and acetylfentanyl (analytical methodologies not identified in the abstract) (917); analysis of mixtures of methamphetamine, MDMA, and ketamine by GC/MS and GC/FID (918);

2015 analysis of dextromethorphan, 2-aminoindane, and lidocaine using handheld NIR, Raman, and FTIR/ATR instruments (919); examination of “third hand smoke” from cocaine and methamphetamine as a source of recoverable trace evidence (920); detection of cocaine and ketamine by paper microfluidic devices (921); qualitative, quantitative, and temporal study of cutting agents for cocaine and heroin confiscated in western Switzerland from 2006 to 2014 (analytical methodologies not identified in the abstract) (922); detection of cocaine, phytocannabinoids, nicotine, caffeine, and others in the air by collection on filters with analysis by GC/MSD (923); detection of nicotine, caffeine, cocaine, cannabidiol, and THC on particulates in indoor air (analytical methodology not identified in the abstract) (924); trends from 2002 to 2013 in the diversion and abuse of oxycodone, hydrocodone, hydromorphone, fentanyl, morphine, and tramadol (925); validation of a GC/FID for the quantitation of cocaine and heroin (926); analysis of various NPSs, including “Synthacaine” (purported to be a mixt. of methiopropamine (MPA) and dimethocaine, but instead containing MPA and benzocaine), two positional isomers of (2-aminopropyl)-benzofuran (5-APB and 6-APB), 2-amino-1-(4-bromo-2,5-dimethoxyphenethyl)ethanone (bk-2C-B), and 2-(ethylamino)-1-(4-methylphenyl)pentan-1-one (MEAP) (analytical methodologies not identified in the abstract) (927); analysis of two component mixts. of morphine-papaverine and acridine-papaverine by TLC-IMS (928); identification of brodifacoum, black tar heroin and its impurities (morphine, codeine, noscapine, papaverine, and monoacetylmorphine), crack cocaine, and 1-methylaminoanthraquinone by an atmospheric solid analysis probe interfaced to a linear ion trap-MS (929); analysis of 25H-NBOMe, 25D-NBOMe, 25E-NBOMe, 25I-NBMD, RH34, escaline, 5-DBFPV, 3,4-MDPHP, 3,4-dimethyl-NEB, 3,4-dimethyl- α -ethylaminopentiophenone, 3,4-dimethyl- α -PVP, 4F- α -ethylaminopentiophenone, bk-IVP, bk-IBP, MMXE, 25I-NBOMe, ADB-CHIMINACA, 5F-ADB, and butane-1,4-diol by GC/MS, HRMS, and NMR (930); determination of amphetamine, cocaine, methadone, diazepam, methylphenidate, oxazepam, tramadol, morphine, buprenorphine, and 6-monoacetylmorphine by SERS (931); determination of 22 drugs of abuse and transformation products in airborne particulate matter by pressurized liquid extraction followed by LC-MS/MS (cannabinol, cocaine, and methamphetamine were the most abundant compds; the other 18 compds were not identified in the abstract) (932); detection of THC, methamphetamine, and amphetamine at low ppb level in air using a field asymmetric IMS microchip sensor (933); use of paper microfluidic devices for presumptive identification of cocaine, opiates, ketamine, various phenethylamines, and others (934); detection of phytocannabinoids, cocaine, lidocaine, and nicotine by ESI-FT-ICR-MS (with comparison against the fast blue B colorimetric test (935); use of fluorescent d10 metal complexes for the presumptive identification of cocaine, PCP, diphenhydramine, and benzylpiperazine (936); determination of benzodiazepines and zolpidem in water samples (using polypropylene tubes as single-use and low-cost sorptive extraction materials) (937); determination of phentermine, phendimetrazine, phenmetrazine, fenfluramine, benfluorex, mephentermine, fencanfamine, sibutramine, sildenafil, vardenafil, and tadalafil in food supplements by LC-HRMS (938); analysis of venlafaxine, escitalopram, fluoxetine, candesartan, risperidone, trihexyphenidyl, thioridazine, aripiprazole, and trifluoperazine by UHPLC (939); a review of the published voltammetric and potentiometric methods developed for determination of dextromethorphan and

diphenhydramine (940); analysis of FDU-NNEI, AB-CHMINACA, MN-18, N-OH-EDMA, dimethoxy- α -PHP (analytical methodologies not identified in the abstract) (941); analysis of methamphetamine, morphine, and codeine by a probe ESI-MS with a discontinuous atmospheric pressure interface (942); determination of barbital, phenobarbital, chlormezanone, amobarbital, zopiclone, melatonin, chlorphenamine maleate, clozapine, zaleplone, zolpidem tartrate, oxazepam, nitrazepam, triazolam, clonazepam, midazolam maleate, diazepam, and olanzapine in traditional Chinese medicines and health foods by HPLC (943); determination of carbamazepine, doxepin, diazepam, lorazepam, amitriptyline, temazepam, oxazepam, and alprazolam in an urban water system (analytical method not listed in the abstract) (944);

2016 use of screen-printed electrodes for quantification of cocaine and THC (945); the persistence of illicit drug smoke residues from cocaine and methamphetamine and their recovery from common household surfaces (946); chemical profiling of cocaine and heroin as a tool to decipher the structure and organisation of illicit drug markets (947); a review of the cutting of cocaine and heroin (948); analysis of mephedrone and MDAI by microcrystalline testing and Raman microspectroscopy (949); use of a fluorescence probe for ketamine and methamphetamine detection without pretreatment (950); IMS response of cocaine, heroin, methamphetamine, MDMA, and THC against environmental background levels (951); the vaporization enthalpy and vapor pressure of fenpropidin and phencyclidine (PCP) at $T/K = 298.15$ by correlation GC (952); simultaneous determination of morphine and naltrexone by HPLC (953); analysis of 5-MAPDB, 5-AEDB, MDMA methylene homolog, 6-Br-MDMA, and 5-APB-NBOMe by LC-QTOF-MS, GC/MS, and NMR (954); sorption of ionized pharmaceutical and illicit drugs to a mixed-mode coated microsampler, including amphetamine, amitriptyline, promazine, chlorpromazine, triflupromazine, difenzoquat, 8 basic pharmaceutical and illicit drugs (MDMA, atenolol, alprenolol, metoprolol, morphine, nicotine, tramadol, verapamil, 3 neutral benzodiazepines (diazepam, temazepam, and oxazepam), and diclofenac (955); analysis of a mixt. of cocaine, MDA, and MDMA by single analyzer precursor scanning using an ion trap (956); analysis of the phenethylamine derivative 2-(4-iodo-2,5-dimethoxyphenyl)-N-[(3,4-methylenedioxyphenyl)methyl]ethanamine (25I-NB34MD) and the piperazine derivative 1-(3,4-difluoromethylenedioxybenzyl)piperazine (DF-MDBP) by LC/MS, GC/MS, HRMS, and NMR (957).

2. Instrument Focus

Forensic Chemists must maintain familiarity with updates in current instrumental techniques and become versant in new, improved methods of analysis.

Improved/existing and new technologies are reviewed and applied to both routine and specialized analyses of drugs. In cases where improved performance is observed, case reports are generated for the forensic community.

2.A – Polydrug B: Mixed or Unrelated Groups of Compounds or Substances

Named Groups of Compounds: 2013 analysis of 277 “selected” synthetic cannabinoids and cathinones, amphetamines, natural cannabinoids, opioids, cocaine and other “important drugs of abuse” by UHPLC-HR-TOFMS (toxicological focus) (958); analysis of cathinones, phenethylamines, tryptamines, and piperazines by LC-QQQ-MS/MS in the MRM mode (toxicological focus) (959); 2014 analysis of various phenethylamines, cathinones, synthetic cannabinoids, and tryptamines by IMS (number of compounds not provided in the abstract) (960); an overview and literature review of synthetic cannabinoids and synthetic cathinones (961); a review of the analysis of (unspecified) “psychostimulants” by TLC (962); qualitative analysis of 34 synthetic cannabinoids and synthetic cathinones by GC-triple quadrupole-MS/MS (963); screening and identification of cathinones, synthetic cannabinoids/cannabimimetics, and phenethylamines by UHPLC with DAD and MS detection (964); a review of the analysis of (unspecified) “anesthetics” by TLC (965); identification of 61 different psychoactive substances (predominantly substituted phenethylamines, cathinones, tryptamines and synthetic cannabinoids) by LC-chemiluminescence-nitrogen detection (966); analysis of morphine and a series of adrenergic phenolic amines (not identified in the abstract) by chemiluminescence detection on 3D-printed and CNC milled flow-cells (967); cross-reactivity of 24 phenylethylamines (including 8 cathinone derivatives), 3 piperazines, and 3 tryptamines in commercial enzyme-linked immunosorbent assays (968); chiral analysis of seven benzofurys, four cathinones, two diphenidines, ethylphenidate, methiopropamine, and thiothinone by CE (969); an overview of the appearance and evolution of cannabimimetics and cathinones (970); 2015 characterization of 25I-NB2OMe, 25I-NB3OMe, 25I-NB4OMe, 25I-NB2B, 25I-NB3B, 25I-NB4B, their 5-methoxytryptamine counterparts, and 6 meta-substituted N-benzyl derivs. of 5-methoxytryptamine (CF₃, F, CH₃, Cl, I, SCH₃), by GC/ion trap-MS in both EI and CI modes, LC/DAD, IR, ESI-QTOF-MS/MS, and Triple-Quad-MS/MS (971); analysis of 11 phenethylamines and cathinones by 1H-NMR, COSY, TOCSY, and DOSY (972); an overview of synthetic cannabinoids and designer cathinones (973); regioisomeric and enantiomeric analyses of 24 designer cathinones and phenethylamines using UHPLC and CE with added cyclodextrins (compounds not identified in the abstract) (974); a review of the detection methods (including covering colorimetric detection, immunochem. assays, GC/MS analyses, and LC/MS) for synthetic cannabinoids and cathinones (975); cross-reactivity of 2,5-dimethoxyamphetamines, 2C (2,5-dimethoxyphenethylamines), β -keto amphetamines, substituted amphetamines, piperazines, α -pyrrolidinopropiophenones, tryptamines and PCP analogs on five commercial immunoassay screening kits (976); 2016 separations of barbiturates,

sulfonamides, nucleic bases, and nucleosides on polymethacrylate zwitterionic monolithic micro-columns in 2D-LC (977);

Abused Substances Illegally Added to Licit Pharmaceuticals, Herbal Medications, Health Supplements, and Foodstuffs

(Notes: A) Specific, named compounds are compiled in their individual categories above; B) There are many dozens/hundreds of (highly repetitive) articles pertaining to adulteration of Chinese foods, food seasonings, health care supplements, sexual enhancement aids, Chinese Traditional Medicines, etc.; only a select six of these are included below): **2012** analysis of for anorexigenic, benzodiazepinic, and antidepressant drugs in phytopharmaceuticals by GC/MS (978); **2013** detection of undeclared synthetic drugs in traditional herbal medicines, using LC-MS/MS, GC-MS/MS, and similar techniques (979); standardless ¹H-NMR determination of a “wide range of” pharmacologically active substances in dietary supplements and medicines (only mesterolone is specifically mentioned in the abstract) (980); **2014** detection of 35 illegally added steroid compounds in foods and dietary supplements by LC-MS/MS (981); detection of 29 weight loss compounds in foods and dietary supplements by LC-MS/MS (982); a review of the determination of pharmacologic adulterants in herbal-based pharmaceuticals by CE (983); rapid identification of 22 drugs illegally added into sleep-improving health foods by UHPLC-TOF-MS (984); **2015** simultaneous analysis of 28 narcotic adulterants (not identified in the abstract) used in dietary supplements by LC-MS/MS (985); analysis of 24 sedative-hypnotic drugs (not identified in the abstract) illegally added into health foods, by UPLC-ESI-Q-TOF/MS (986); screening of 24 sedative hypnotics illegally added to “improving sleep” health foods by HPLC-ion trap-MS (987); determination of 36 chemicals added into traditional Chinese medicines and health care products by UPLC-MS/MS (988); substitute reference substance and secondary mass spectral libraries for rapid screening of sedative hypnotic drugs illegally added to Chinese drugs and health products by HPLC-DAD and HPLC-MS/MS (989); an overview and review of alkaloids in foods (990); determination of caffeine and adrenergic stimulants in food supplements by HPLC/DAD (991); identification of chemical substances illegally adulterated in traditional Chinese medicines and health foods by physico-chem. anal., TLC, HPLC, LC/MS, GC/MS, CE, ion mobility chromatog., IR, NIR, Raman, and LC-MS/MS (992); **2016** a comprehensive strategy to detect the fraudulent adulteration of herbs by FTIR and chemometrics, as well as LC-HRMS (993); direct determination of 42 chemical drugs illegally added in herbal medicines and dietary supplement by HPLC-Quadrupole- electrostatic field Orbitrap-HRMS (994); an overview of regulation of dietary supplements in the U.S. and issues of adulteration with phenethylamines (995); detection of low molecular weight adulterants in beverages by DART-MS (996); an overview and review of the adulteration of herbal sexual enhancers and dieting aids (997);

Abused Drugs and Pharmaceuticals in Surface Waters and Municipal Wastewater Streams:

[Note: Each year in this subsection is separated by a line space.]

2012 analysis of sewage in the Brazilian Federal District as a means for estimating cocaine consumption (analytical method not specified in the abstract) (998);

2013 a study of the uncertainty associated with the estimation of community illicit drug consumption via analysis of sewage (999); analysis for mephedrone, methylone, MDPV, BZP, TFMPP, methcathinone, and MDMA in sewage in Adelaide, Australia, by SPE-LC-MS/MS (1000); a review of drugs of abuse in waters and wastewaters: occurrence, analysis, and forensic applications (1001); detection of pharmaceuticals and “food additives” in sewage by SPE-LC-MS/MS (1002); by online-SPE-LC/MS (1003); analysis for 25 different drugs in wastewater by solid phase extraction and GC/MS (1004); detection of illicit drugs in wetlands water by LC/MS (1005); analysis of wastewater in Finland for abused drugs and opioids, using SPE and LC-MS/MS (1006);

2014 identification and quantification of trace concns. of pharmaceuticals (caffeine, prazosin, enalapril, carbamazepine, nifedipine, levonorgestrel, simvastatin, hydrochlorothiazide, gliclazide, diclofenac-Na, and mefenamic acid) in surface waters, by LC-TOF/MS (1007); removal efficiencies of cocaine, amphetamine, methamphetamine, THC-COOH, benzoylecgonine, MDMA, ketamine, heroin, and other drugs at a wastewater treatment plant (analytical methodology not identified in the abstract) (1008); determination of stimulants, hallucinogens and their metabolites, opioids, morphine derivs., benzodiazepines, antidepressants, and others in wastewaters in England (analytical methodologies not identified in the abstract) (1009); population normalization using ammonium in wastewater-based epidemiology, and its application to illicit drug monitoring (benzoyl ecgonine, THC-COOH, cocaine, and 4-hydroxy-3-methoxymethamphetamine are named in the abstract) (1010); use of a cavitand-grafted silicon microcantilever as a universal probe for illicit and designer drugs in water (1011); determination of amphetamines, MDMA, cocaine, opioids, cannabis, and ketamine, and their major metabolites, in urban wastewaters by UHPLC-MS/MS (1012); determination of 1525 micropollutants and transformation products in wastewater by LC-QTOF-MS with an accurate-mass database (1013); survey of the occurrence of pharmaceuticals in Spanish drinking waters (1014); highly sensitive determination of 68 psychoactive pharmaceuticals, illicit drugs, and related human metabolites in wastewater by LC-MS/MS (1015); screening of illicit and licit drugs in waters in Colombia by LC-QTOF-MS (1016); determination of 21 acidic pharmaceuticals and personal care products in the Turia River Basin, Spain by LC/MS-MS/ESI-NI (1017); a selection of papers from the first international multidisciplinary conference on detecting illicit drugs in wastewater (1018); evaluation of illicit and licit drug consumption based on wastewater analysis in Fort de France urban area (Martinique, Caribbean) (1019); determination of nalbuphine, naltrexone, morphine, and tramadol by a bromatometric assay (1020); a review of the analysis of chiral pharmaceuticals in the environment (wastewater) by chiral chromatography coupled with mass spectrometry (1021); a sampling method for detecting analgesics, psycholeptics, antidepressants, and illicit drugs in aquatic environments in the Czech Republic (1022); quantification of (unspecified) target drugs in different wastewater samples by a validated SPE/LC-MS/MS method (1023); the ecotoxicity and contribution to the environmental hazard of pharmaceuticals in hospital wastewater (1024); use of columns containing sand and

undisturbed fine-grained sediments to simulate injection of wastewater contg. caffeine, methamphetamine, and acetaminophen into a septic system, leaky sewer, or landfill (1025); a review of the determination of pharmaceuticals and illicit drugs in waters by LC-HRMS (1026); communal assessment of drugs of abuse and identification of their transformation products by analysis of sewage/wastewater by online SPE-LC-HRMS (1027); estimation of illicit and pharmaceutical drug consumption estimated via wastewater analysis (1028); a review of the occurrence, effects, and methods for detection of antibiotics and illicit drugs in the environment (1029); determination of cocaine, benzoylecgonine, ecgonine methylester, methadone, 2-ethylidene-1,5-dimethyl-3,3-diphenylpyrrolidine, 6-monoacetylmorphine, amphetamine, methamphetamine, ecstasy, mephedrone, methylenedioxypropylamphetamine, 11-nor-9-carboxy-delta-9-tetrahydrocannabinol, ketamine, and norketamine in sewage (analytical method not identified in the abstract) (1030); an overview of international management trend of pharmaceuticals and personal care products in water environments (1031); the transformation products of illicit drugs in the aquatic environment (1032); estimation of amphetamine and methamphetamine use through sewage-based analysis (1033); determination of pharmaceuticals and personal care products in a mesoscale subtropical watershed and their application as sewage markers (1034); comparison of illicit drug use in three selected towns in Slovakia by wastewater analysis (1035); identification of contaminants in water by UHPLC-QTOF-MS (1036); analysis for ethyl sulfate in raw wastewater for estimation of alcohol consumption and its correlation with drugs of abuse in the city of Barcelona, Spain (1037); an overview of organic contaminants in surface water and groundwater in Italy (1038); determination of amphetamines in wastewater by LC-MS/MS (1039); a discussion of the need to develop ethical guidelines for researchers using sewage epidemiol. to monitor drug use in the general population and in specific precincts, including prisons, schools, and workplaces (1040); determination of benzodiazepines, related pharmaceuticals, and metabolites in water by SPE and LC-MS/MS (1041); using biomarkers in wastewater to monitor community drug use (focus on NPSs) (1042); determination of over 400 priority and emerging pollutants in water and wastewater by SPE and LC-TOF-MS (1043); removal efficiencies of amphetamine-type stimulants, cocaine and benzoylecgonine, opioids, codeine, MDA, fentanyl, dihydrocodeine, and heroin at each point of wastewater treatment (analytical methodology not identified in the abstract) (1044); determination of cocaine, benzoylecgonine, propranolol, diclofenac, amitriptyline, carbamazepine, carbamazepine-epoxide, citalopram, metoprolol, carisoprolol, and sertraline in urban streams in Brazil (analytical methodology not identified in the abstract) (1045); systematic screening for common wastewater-marking pharmaceuticals in urban aquatic environments (1046);

2015 occurrence and in-stream attenuation of wastewater-derived pharmaceuticals in Iberian rivers, Spain (1047); determination of 4 benzodiazepines (bromazepam, carbamazepine, diazepam, and nordiazepam) and 4 barbiturates (barbital, pentobarbital, phenobarbital, and secobarbital) in river water and wastewater using SPE followed by LC-(ESI)MS/MS (1048); screening for pharmaceuticals and illicit drugs in wastewater and surface waters of Spain and Italy by UHPLC-QTOF-MS and LC-LTQ-Orbitrap-MS (1049); determination of heroin and methadone in wastewater in Lausanne, Switzerland (analytical methodology not identified in the abstract)

(1050); fast determination of 40 drugs in water (10 effluent wastewater and 10 surface water samples) using large volume direct injection LC-MS/MS (1051); determination of 10 synthetic cannabinoids, cathinones, piperazines and pyrrolidophenones in wastewater by LC-MS/MS (1052); determination of ketamine and mephedrone in wastewater in 17 cities in Italy, by SPE-LC-MS/MS (1053); methamphetamine and ketamine (analytical methodology not identified in the abstract) (1054); an overview and review of determination of contaminants in water by UHPLC/MS (1055); detection of cocaine and benzoylecgonine (and other drugs) in samples collected from three sewage treatment plants in Cyprus by off-line solid phase extn. followed by LC-MS/MS (1056); screening for more than 1,000 licit and illicit drugs and their metabolites in wastewater and surface waters from the Bogota, Colombia area by SPE followed by UHPLC-QTOF-MS (1057); detection of amphetamines, opioids, cocaine, cannabinoids, lysergics, and their corresponding metabolites by SPE-LC-HR-MS (1058); advances towards a universal screening for organic pollutants in waters, by GC-QTOF-MS and LC-QTOF-MS (1059 and 1060); detection of illicit drugs in raw sewage influents by HRMS (1061); chemometric application of pharmaco-signatures in different aquatic systems (1062); determination of methoxetamine, butylone, ethylone, methylone, methiopropamine, PMMA, and PMA in sewage by LC-ESI-MS/MS (1063); the systematic and day-to-day effects of chemical-derived population estimates on wastewater-based drug epidemiology (1064); use of a Fenton-like reaction to remove illicit drugs and pharmaceuticals from wastewater (emphasis on methamphetamine and tramadol) (1065); determination of amphetamine and methamphetamine at 10 wastewater treatment plants by LC-HR-MS/MS (1066); detection of methamphetamine, amphetamine, and codeine in wastewater (analytical methodology not identified in the abstract) (1067); analysis of pharmacologically active compounds in the environment by chiral LC-MS/MS (1068); detection of 4'-methyl- α -pyrrolidinohexanophenone (MPHP), 2-[4-(ethylsulfanyl)-2,5-dimethoxyphenyl]ethanamine (2C-T-2; Rosy), 4-methyl-5-phenyl-4,5-dihydro-1,3-oxazol-2-amine (4-MAR), and 1-(4-methoxyphenyl)-2-propanamine (PMA) in raw sewage by HR-MS (location not identified in the abstract) (1069); linking drugs of abuse in wastewater to contamination of surface and drinking water (17 drugs of abuse, including cocaine, several amphetamines, opioid drugs, and 2 metabolites, benzoylecgonine, and 2-ethylidene-1,5-dimethyl-3,3'-diphenylpyrrolidine (a metabolite of methadone) were investigated; analytical methodology not identified in the abstract) (1070); determination of alcohol and cocaine co-consumption in 2 European cities as assessed by wastewater analysis using LC-MS/MS (1071); wastewater-based epidemiology of stimulant drugs based on analysis of sewage samples from 42 European cities collected daily for one week in March, 2013 (1072); determination of 25 synthetic psychoactive compds., including amphetamine, sympathomimetic substituted amphetamines, synthetic cathinones, and ketamine, in raw wastewater, secondary effluent, and river water by SPE followed by LC-MS/MS (1073); **comparison of wastewater analysis and population surveys for use of methamphetamine, MDMA, and cocaine** (1074); determination of cocaine and benzoylecgonine in the Esmeraldas watershed in Ecuador (analytical methodology not identified in the abstract) (1075); comparison of population surveys with wastewater analysis for monitoring illicit drug consumption in Italy from 2010-2014 (1076);

2016 identification of “a wide range of suspected and unknown compds. in environmental samples” by LC-HRMS (1077); determination of the occurrence and spatial distribution of 158 pharmaceuticals, drugs of abuse, and related metabolites in offshore seawater by SPME and LC-MS/MS (1078); determination of pharmaceuticals in coastal systems using SPE and UPLC-MS/MS (1079); use of wastewater-based epidemiology to estimate consumption of methamphetamine, benzoylecgonine, MDMA, methadone, oxycodone, and hydrocodone (analytical methodology not identified in the abstract) (1080); **analysis of illicit drugs in wastewater to assess the market share held by criminal groups** (1081); determination of amphetamine, methamphetamine, MDMA, and cocaine in 7 locations in Belgium over 2011-2015 (analytical methodology not identified in the abstract) (1082); validation and uncertainties evaluation of an isotope dilution-SPE-LC-MS/MS for the quantification of drug residues in surface waters (including diazepam and MDMA) (1083); determination of cocaine and benzoylecgonine in environmental samples by newly developed sorbent materials (1084); detection of opioid analgesics, amphetamines, cocaine, heroin, stimulants, anesthetics, sedatives, anxiolytics, designer drugs, phosphodiesterase-5 inhibitors, and amphetamine and methamphetamine drug precursors in wastewaters by LC-MS/MS (1085); quantitative analysis of morphine, oxymorphone, hydromorphone, oxycodone, hydrocodone, and THC-COOH in river and wastewater by UHPLC-MS/MS with an API/ESI source (1086); source discrimination of drug residues in wastewater by chiral LC-MS-MS (a case study) (1087); determination of drugs of abuse and alcohol consumption through sewage-based epidemiology among different groups of population on the Greek Island of Lesbos (1088); screening for drugs of abuse in the wastewater in a small college town in Southern Arkansas by GC/MS, GC/FID, and HPLC/MS (1089); screening for NPSs in urban wastewater using HRMS (1090); evaluation of sampling plans for cocaine, methamphetamine, MDMA, and methadone in wastewater (1091); wastewater based epidemiology in Finland (samples analyzed by UHPLC-MS/MS) (1092); estimation of drug abuse in 9 Polish cities by wastewater analysis by HPLC-MS/MS (1093); determination of heroin, cocaine, amphetamine, MDMA, methamphetamine, cannabis, codeine, and methadone in 6 Croatian cities (analytical methodology not identified in the abstract) (1094); correlated results from an Australia-wide wastewater monitoring of cocaine/benzoylecgonine, methamphetamine, and MDMA (analysis by LC-MS/MS) (1095); determination of cocaine, MDMA, and methamphetamine residues in wastewater by LC/MS (1096); common illicit drugs (primarily methamphetamine and ketamine and their metabolites in surface waters) (analytical methodology not identified in the abstract) (1097); removal of psychoactive pharmaceuticals and illicit drugs from wastewaters by zerovalent iron and iron(VI) (1098); determination of cocaine, benzoylecgonine, ephedrine, MDMA, methadone and its metabolite EDDP in Spanish river basins by online SPE-LC-ESI-MS/MS (1099); a review on the stability of illicit drugs in sewers and wastewater samples (1100); analyses for 48 emerging pollutants, including 25 drugs of abuse and metabolites, 17 cytostatic drugs, and 6 iodinated contrast media, in tap water in Madrid, Spain by SPE and LC-MS/MS (1101); effects of time delay between sample collection and extraction of wastewater samples for amphetamine and opioid analysis (1102); quant. analysis of 33 compds in a Brazillian coastal zone., including cocaine and bezoylecgonine, by

LC-MS/MS (1103); detection and quantification of various opioid compounds (primarily heroin and morphine) in urban wastewater in Cookeville, Tennessee by LC-MS/MS (1104); determination of cocaine, methamphetamine, MDMA, amphetamine, codeine, morphine, heroin, fentanyl, oxycodone, methadone, BZP, TFMPP, methcathinone, methylone, mephedrone, MDPV, alpha-PVP, PMA, 25C-NBOMe, 25B-NBOMe, 25I-NBOMe, and cannabis in Adelaide, Australia for up to 4 years between Dec. 2011 and Dec. 2015 (analytical methodology not identified in the abstract) (1105); determination of metabolites of methamphetamine, cocaine, THC, and heroin by LC/MS (1106); determination of amphetamine-like compds., ketamine, cocaine, and opioids in North China (analytical methodology not identified in the abstract) (1107); detection of cocaine in wastewater with DNA-directed immobilization aptamer sensors (1108);

“Novel Psychoactive Substances” (NPSs): 2013 a review of 1320 cases containing one or more of 26 synthetic cannabinoids, 12 designer stimulants, and 5 hallucinogenic-like drugs (1109); an overview of the New Zealand approach to regulated NPSs (1110); **2014** a study of the prevalence and correlates of NPS use amongst a group of regular Ecstasy users in Australia (1111); a review (1112); a review (1113); a report from the European Drug Emergencies Network on their efforts to improve the knowledge of acute drug toxicity of recreational drugs and NPS (1114); an overview of the effects and risks associated with NPSs (toxicological focus) (1115); an overview of legislation against NPSs in Ireland (1116); identification of NPSs by FTIR, Raman, and GC-IR (1117); an overview of the emerging trends in the abuse of NPSs (1118); an overview (1119); detection and presumptive identification of NPSs by a portable NIR spectrometer (1120); the impact of new retail restrictions and product licensing on the regulated legal market for NPS products in New Zealand (1121); an overview of the high variability of active ingredients concentration, mislabelled preparations, and presence of multiple psychoactive substances in NPS products (1122); **2015** wide-range screening of NPSs by FIA-HRMS (1123); detection and characterization of NPSs by IMS (1124); a review (1125); a brief overview of recent trends (1126); rapid screening of 35 NPSs by IMS and DART-QTOF-MS (1127); a study on the prediction of bioactivity of NPSs (1128); detection of NPSs by SERS (1129); an overview of recent developments in the analysis of NPSs (1130); an overview of NPSs and their impact on forensic science (1131); an overview and review, covering years 2013-2015 (1132); **2016** a proposal for a new categorization of NPSs based on neurobiol. mechanisms of action (1133); an overview of how NPSs are studied, produced, marketed, and controlled (1134); screening for 221 NPSs by infrared and Raman (1135); a review on the screening for NPSs by LC coupled with low- and high-resoln. MS (covering PubMed-listed studies from Jan. 2014 to Jan. 2016) (1136); detection of NPSs in street samples by NIR and chemometrics (1137); an update on New Zealand’s legal market for NPSs (1138); an overview of the pharmacology of stimulant and hallucinogen NPSs (1139); a brief overview of the NPS situation in Japan (1140); a brief overview of the analytical challenges posed by NPSs (1141);

“Hallucinogens”, “Hypnotics” (and similar generic terms): 2013 editorial remarks against the global prohibition of psychoactive drugs (1142); **2014** a review of the non-medical use of dissociative drugs (1143); a review of the determination of

of anxiolytics and sedatives by TLC (1144); **2015** sedative-hypnotic and anxiolytic effects of "lotus leaf alkaloid extract" (the exact species of lotus – there are many - was not identified in the abstract) (1145);

“Illicit Drugs” (including “Controlled Substances,” “Drugs of Abuse,” “Illicit Drugs,” “Narcotics,” “Seized Drugs,” and similar generic terms): **2012** “drugs of abuse” by Raman (1146); use of spatially offset Raman to detect “illicit drugs” through opaque plastic containers, colored glass bottles, paper envelopes, and clothes (1147); a review on THz time-domain spectroscopy (including THz spectra for “drugs of abuse”) (1148); detection of “drugs of abuse” using SERS (1149); application of handheld FTIR and Raman spectrometers for detection of “drugs of abuse” (1150); a review of the analysis of “seized drugs” by UHPLC and UHPLC-MS (1151); a short review of recent advances in analysis of “drugs” (and other substrates) by MS (1152); **2013** an evaluation of the results of impurity profiling of “illicit drugs” from different analytical methods and/or from different laboratories (1153); detection of trace amounts of “illicit drugs” on surfaces by direct analyte-probed nanoextraction coupled to nanospray ionization-mass spectrometry (1154); detection of “drugs” concealed inside diffusely scattering packaging, including plastic, paper, and cloth, by spatially offset Raman (1155); analysis of “illicit drugs” by ambient pressure thermal desorption ionization MS (1156); rapid screening for 73 “toxic and harmful substances” in foods by UHPLC/MS, with sample cleanup using the QuEChERS system (1157); an overview and review of the analysis of “illegal drug products” (1158); the effects of solvents on the analysis of “drugs” by ESI-MS (1159); a review of the analysis of “law-evading and illegal drugs” using liq.-liq. extrn. and GC/MS (1160); a review of CE and CEC methods used for analysis of “drugs” in biological matrices (1161); use of a supramolecular sensor array with two fluorescent receptors to detect “addictive OTC drugs” (1162); analysis of “seized drugs” by LC-ESI/MS/MS and AP-MALDI-MS/MS, with comparisons of the two techniques (1163); detection of “illicit substances” and pharmaceutical counterfeits by nuclear quadrupole and magnetic resonance (1164); annual review of “banned substances” (sports doping focus) (1165); an overview of advanced analytical instrumentation and methods for “drugs of abuse” (toxicological focus) (1166); **2014** screening of textiles for “contraband drugs” using portable Raman spectroscopy and chemometrics (1167); an evaluation of the effectiveness of MS, IR, and portable Raman to analyze commonly encountered drug mixts., as well as “legal highs” (1168); screening of “drugs of abuse” using a commercial paper spray system (1169); detection of “abused drugs” by HPLC (1170); an overview of recent trends in the analysis of “emerging drugs of abuse” (1171); analysis of of designer drugs (“bath salts”) by Raman and SERS (1172); a review of new designer “drugs of abuse” (1173); a quantitative structure-toxicity relationship of the aquatic toxicity for various “narcotic pollutants” using the norm indexes (1174); **2015** a comprehensive review of the pyrolysis of “drugs of abuse” (1175); analysis of “drugs of abuse” (naturally occurring psychotropic drugs and new designer drugs) by DART-MS (1176); use of diazonium ions for the presumptive testing of “narcotics” containing an activated aromatic ring (1177); screening for “illicit drugs” by direct-heating HS-SPME with GC/MS (1178); identification of “abused drugs” by GC/FTIR (1179); an overview and review of “drugs of abuse” and their detection methodologies (1180); determination of “illicit drugs” and their metabolites on banknotes by methanol extrn.

followed by LC-MS/MS (1181); a review of alkylsilyl derivatization techniques in the analysis of “illicit drugs” by GC (GHB, amphetamines, opiates, and cannabinoids were mentioned in the abstract) (1182); indirect chiral separation of “new recreational drugs” by GC/MS using trifluoroacetyl-L-prolyl chloride (1183); **2016** results of the Trans European Drug Information (TEDI) project (results for cocaine, ecstasy, and amphetamine, plus comments on NPSs detected between 2008 and 2013) (1184); rapid identification of “seized controlled substances” and related compounds by MS/MS without chromatography (1185); screening of “drugs of abuse” using DART-MS (1186); “forensic drug” analysis by chemical derivatization followed by GC/MS and LC/MS (1187); a survey of the qual. distribution of “drugs of abuse” (mostly NPSs) confiscated in Italy between 2013 and 2015 (1188);

Pharmaceuticals/Counterfeits (with a focus on differentiation of legitimate versus counterfeit products, or for monitoring quality control for legitimate pharmaceuticals; see also a significant number of citations concerning counterfeits under Phosphodiesterase-5 Inhibitors, above): **2012** a review of the detection of counterfeit medications by Raman (1189); **2013** a review of a paper-based test for screening for counterfeits (1190); a general overview of the chromatographic techniques used to characterize counterfeit and illegal pharmaceuticals (1191); an overview of chromatographic and spectroscopic counterfeit detection methods (1192); examination of tablet surfaces by Multimodal DESI-MS imaging to detect counterfeits (1193); detection of counterfeit medications with portable Raman (1194); analysis of pharmaceuticals by Raman (1195); a review on the detection of counterfeit medications, focusing on HPLC and MS, but also discussing color testing, TLC, GC, Raman, NIR, FTIR, and NMR, using antimalarial drugs and sildenafil as illustrative examples (1196); **2014** detection of counterfeit medications with Raman and NIR (1197); confirmational identification of pharmaceuticals via DART-TOF-MS (1198); an overview of pharmaceutical process validation of solid dosage form (1199); **2015** systematic chemical and packaging analysis of counterfeit medications to derive useful intelligence (1200); **2016** an analytical strategy for rapid identification of counterfeit medications (1201); a review of the identification of counterfeit medicines by chemometrics (1202); a comprehensive review on prevalence, detection, and prevention of counterfeit drugs (1203).

2.B – Instrument Focus

General Overviews and Reviews, and articles covering multiple techniques:

2014 an overview of forensic drug analyses, including an analytical road-map (1204); an overview of drug testing, covering chem. testing, chromatog., spectroscopy, CE, immunoassay, and IMS (1205); **2015** a review of miniaturized separation techniques for forensic drugs analysis (including CE, CEC, and nano-LC) (1206);

Color Testing: **2014** the effect of benzene ring substituents on the mechanism of Duquenois Levine test for (phyto-)cannabinoid detection (1207); detection of pharmaceuticals using paper analytical devices (embedded with various color-testing reagents) (1208); **2015** use of presumptive color tests for NPSs (abstract not

available) (1209); the modernization of physical appearance and solution color tests using quantitative tristimulus colorimetry (1210);

Computerized Tomography (CT): 2015 dual-energy CT behavior of heroin, cocaine, and typical adulterants (1211); use of CT (and X-ray, ultrasound, and MRI) to detect body packing (1212);

Electrophoresis (and Related Techniques): 2013 determination of active ingredients and preservatives in pharmaceuticals by CZE (1213); a review of recent advances in electrodriven enantioseparations (listed applications include “pharmaceutical” and “forensic”) (1214); 2014 separation of acidic drugs by CEC using both chlorinated and nonchlorinated polysaccharide-based selectors (1215); a comprehensive overview and review (1216); a review of the application of CE techniques in toxicological analysis (1217); a review of recent method developments and applications of CE/DAD to pharmaceuticals (1218); 2015 a review of electromigrative sepn. techniques in forensic toxicol. (1219);

Gas Chromatography: 2013 forensic applications of GC (1220); 2016 a review of the forensic potential of comprehensive 2D-GC (1221); cleanup of complex matrices (containing drugs) by QuEChERS followed by GC analysis (1222);

Hyperspectral Imaging: 2013 development of a handheld widefield hyperspectral imaging (HSI) sensor for standoff detection of explosive, chemical, and narcotic residues (stated applications include “locating production facilities of illegal drugs”) (1223);

Infrared Spectroscopy: 2013 use of IR spectral imaging for drug quality control (1224); 2014 analysis of varied substrates by FTIR spectroscopic imaging (1225); use of a handheld near IR spectrometer for the classification of 140 different substances, including cocaine, heroin, oxycodone, diazepam, synthetic cathinones, and synthetic cannabinoids (1226);

Ion Chromatography: 2012 ion chromatographic analysis of pharmaceuticals to determine authenticity and adulteration (listed applications include “forensic analysis”) (1227); 2014 a review of ion chromatography-mass spectrometry (1228);

Ion Mobility Spectroscopy: 2014 use of DESI-AP-IMS for drug detection (1229);

“Lab-on-a-Chip” (Microfluidics): 2011 the use of microfluidic platforms for solid form screening of pharmaceuticals by Raman (1230); 2013 an overview of “forensic drug analysis” by microfluidic devices (1231); 2014 enhancement of chemiluminescent detection in microfluidic systems, for anal. of a wide range of compds., including illicit drugs and pharmaceuticals (1232);

Liquid Chromatography: 2012 an overview of good laboratory practices for HPLC (1233); an overview of some of the most recent applications of hyphenated LC techniques for forensic analyses (1234); 2013 quantitative structure-retention relationships models for prediction of HPLC retention time of small molecules (1235);

2014 use of immobilized polysaccharide-based stationary phases for enantioseparation in normal versus reversed phase HPLC (1236); a review of the use of chiral supercritical fluid chromatography for analysis of pharmaceuticals and drugs of abuse (1237); a review of HILIC, discussing the development, basic sepn. mechanisms, stationary and mobile phases, and summarizing its applications in several research fields (1238); **2015** a chemometric approach to improve the accuracy and precision of quantitation in 2D-LC with dual detectors and multivariate curve resolution (1239); simultaneous determination of hydrophobicity and dissociation constant for 161 drugs by gradient RP-HPLC/MS (1240); HPLC method development using structure-based database search, physico-chemical prediction, and chromatographic simulation (1241); **2016** automated screening of reversed-phase stationary phases for small-molecule separations using LC/MS (emphasis on LC) (1242); simulation of elution profiles under gradient elution conditions, with mismatched injection and mobile phase solvents (includes simulated sepn. of selected amphetamines) (1243);

Mass Spectrometry: **2012** the mass spectra of designer drugs (reference text) (1244); **2013** use of Desorption Electro-Flow Focusing Ionization of explosives and narcotics for ambient pressure mass spectrometry (the “narcotics” included cocaine; no others were listed in the abstract) (1245); a review of DART-MS (1246); a review of ultrasensitive MS of organic molecules (listed applications include “forensics”) (1247); a review of ambient MS, including DESI, DART, and extractive ESI (listed applications include “forensic identification”) (1248); an evaluation of standardized software for processing GC/MS data from different instruments (1249); the application of ultra-fast triple quadrupole LC-MS/MS for forensic analysis of “abused drugs” (1250); a review of DESI-MS (listed applications include “illicit drugs”) (1251); evaluation and testing of an alternative search algorithm for compound identification using the Wiley Registry of Tandem Mass Spectral Data, MSforID (1252); mass spectrometry using Matrix Assisted Ionization in vacuum (1253); **2014** recent advances in forensic drug analysis by DART-MS (1254); **2015** a review of surface-assisted laser desorption ionization (SALDI) MS for forensic analysis (1255); a review of forensic mass spectrometry (1256); a wide use target screening system for GC/MS (1257); a new quant. contained-electrospray process for ESI-MS (1258); the use of the partial least squares method to model the positive ESI response produced by small pharmaceutical molecules (1259); a review of the characterization of synthetic and natural product pharmaceuticals by functional group analysis using ESI-ion trap-MS (1260); the use of online chemistry databases to facilitate structure identification (1261); a review of identification criteria and complicating factors for drug confirmation by mass spectrometry (1262); **2016** a review of the applications of ambient mass spectrometry to forensic chemistry (1263); a review of DART-MS (1264); determination of trace palladium in chemical bulk drug by ICP-MS (1265); a review of DART-MS (1266);

Microextraction Techniques: **2013** a review of liquid phase micro-extraction (LPME) techniques used in analysis of Chinese traditional medicines (1267); a review (listed applications include forensic and pharmaceutical) (1268); **2015** a review of SPME techniques (1269); **2016** a review of coupling SPME with ambient

MS (1270); a review of microextraction in forensic toxicology (1271); an overview of microextraction techniques for illicit drug testing (1272);

Microscopy and Microscopic Instrumental Techniques: **2013** comparison between microcrystalline tests performed on microscope slides versus flat capillary tubes (1273); **2014** a review of developments in applications of FTIR microspectroscopy, covering 2005 to 2013 (1274); **2015** use of an FTIR/ATR microscope for detecting analytes in high-interfering matrixes and in products with unknown ingredients (illicit tablets, counterfeit tablets, and unknown powders) (1275);

Nuclear Magnetic Resonance Spectroscopy: **2013** tracking authentic pharmaceuticals by ^2H - and ^{13}C -NMR (1276); **2015** cocaine, MDMA, and “metilona” (possibly methylone?) by “No-D NMR” (i.e., without the use of deuterated solvents) (1277); an overview of a “crime-scene NMR laboratory” (1278); **2016** improving the performance of high-precision qNMR measurements by a double integration procedure (1279); a review of the use of quant. ^1H NMR spectroscopy in drug discovery and development (including a review of the pertinent literature between 1963 and 2015) (1280);

Raman: **2013** Use of THz-Raman accessing molecular structure with Raman spectroscopy for enhanced chemical identification, analysis, and monitoring (especially for discrimination of polymorphs) (1281); **2014** deep Raman detection with 2D correlation analysis for elucidation of a subsurface component under thick powder or packed contents in a bottle (1282); **2016** a review of the applications of SERS in forensic science (1283);

Spectrophotometry: **2014** methods for evaluating the visual limits of color perception are proposed to create common rules for constructing color test scales for visual colorimetric assays (1284); the molecular electron ionization cross - section and λ_{max} in the studies of activities of alkaloids (1285); **2015** defining optimal conditions of colors in 3D space in dependence on gamma values, illumination, and background color (1286); **2016** a review of derivative UV-Vis spectrophotometry (1287);

Stable Isotopes: **2011** forensic applications (reference text) (1288); **2012** a review of the forensic applications of IRMS (1289); **2013** a review of inter-laboratory comparability of stable isotope data (1290); an extensive review of the isotopic anatomies of molecules and minerals (1291); the use of carbon stable isotope ratios in drugs characterization (by IRMS) (1292); global isoscapes for $\delta^{18}\text{O}$ and $\delta^2\text{H}$ in precipitation (1293); **2014** spatial, seasonal, and source variability in the stable oxygen and hydrogen isotopic composition of tap waters throughout the U.S. (1294); **2015** precipitation isotope ($\delta^{18}\text{O}$) zones revealed in time series modeling across Canada and northern U.S. (1295); simple spreadsheet templates for the determination of the measurement uncertainty of stable isotope ratio delta values (1296); **a review of IRMS for source determination** (1297);

Supercritical Fluid Chromatography: 2016 an evaluation of innovative stationary phase ligand chemistries and analytical conditions for the analysis of basic drugs by SFC (1298);

Thin Layer Chromatography (and similar Planar Chromatographic Methods): 2013 an overview, including “forensic applications” (1299);

“Vibrational Spectroscopy” (Raman, mid-, near- and far-IR, and THz spectroscopy): 2012 a review of the use of IR spectroscopy, terahertz spectroscopy and Raman spectroscopy in forensic sciences (1300); a review of sampling techniques for Raman, mid-, near- and far-IR, and THz spectroscopy (1301);

X-Ray Techniques: 2013 the use of energy dispersive X-ray diffraction (ED-XRD) spectra of drugs (and explosives) to detect “body packing” (1302);

Other: 2012 trace determination of metals (copper, zinc, nickel, cobalt, iron, arsenic, antimony, bismuth, vanadium, molybdenum, selenium, and lead) in drugs and pharmaceuticals as N-phenyl[1,2 methane fullerene C₆₀]C₆₁ complexes (1303); **2013** the use of gamma detectors in explosives and narcotics detection systems (1304); a review of microfluidic paper-based analytical devices and micro total analysis systems (1305); the utility of cyclodextrins in analytical chemistry (1306); **2014** a review of the use of acidic potassium permanganate as a chemiluminescence reagent (1307); the use of a chiral diffraction grating to measure the enantiomeric excess of a chiral compound (1308); the application of UV laser-induced solid-state fluorescence spectroscopy for characterization of solid dosage forms (1309); **2015** a review of miniaturized separation techniques (1310); a review of the pyrolysis of drugs of abuse (1311); an overview of emerging hyphenated SEM-EDX (scanning electron microscopy with energy dispersive X-ray spectroscopy) and Raman spectroscopy systems (1312); a review of capacitively coupled contactless conductivity detection (1313); **2016** a review of enhanced performance separations, covering papers published in Anal. Chem. from late 2014 through May 2016 (1314).

3. Miscellaneous Topics

Abuse Deterrent Formulations (see also numerous, specific examples under oxycodone and opiates): **2013** an overview of prescription drug abuse and the need for abuse deterrent formulations (1315); **2014** development and impact of prescription opioid abuse deterrent formulation technologies (1316); the use of prescription opioids with abuse-deterrent technology to address opioid abuse (1317); a review of extended release hydrocodone (1318); the US FDA draft guidance for developing abuse-deterrent opioid analgesics (1319); an overview of anti-drug-abuse measures, including abuse-deterrent formulations (1320); an overview of methods used to reduce abuse potential of commonly abused pharmaceuticals (1321); **2015** an overview of the advance in the R&D of abuse-deterrent opioid analgesics (1322); an overview of abuse-deterrent formulations in countering opioid misuse and abuse (1323); an overview and review of abuse-deterrent formulations (1324); **2016** a comparison of the effectiveness of abuse-deterrent formulations of oxymorphone and oxycodone extended-release drugs (1325); **a review and assessment of the potential impact of abuse-deterrent formulations of prescription opioid analgesics** (1326); an overview of prodrug technology and its application for developing abuse-deterrent opioids (1327); a review (1328); an assessment of extended release abuse deterrent formulations (1329);

Anions and Cations: 2016 a review of the simultaneous separation of cations and anions by CE (1330);

Bacteria: 2014 recovery and identification of bacterial DNA from heroin and methamphetamine (1331); **2016** a discussion of a recent increase in drug abusers in Scotland who have presented with *Staphylococcus aureus* bacteremia with life-threatening complications due to their injection of NPSs (1332); the use of microbe analyses for forensic and criminal investigations (1333);

Canines: 2014 the efficacy of drug detection by fully-trained police dogs varies by breed, training level, type of drug and search environment (1334); treatment and prevention of acute poisoning of drug dogs caused by exposure to methamphetamine, ketamine, and MDMA (1335); **2015** a review of the advances in the use of odor as forensic evidence through optimizing and standardizing instruments and canines (1336);

Clandestine Laboratories – Appraisals and Safety: 2014 an update on the hazards and health effects assocd. with clandestine drug laboratories (1337); an evaluation of the acute and chronic environmental effects of clandestine methamphetamine waste (1338); adsorption and desorption characteristics of methamphetamine, MDMA, and pseudoephedrine in soils (1339); an overview and discussion of home preparations of abused substances (1340); **2015** vehicle-mounted portable mass spectrometry for covert detection of clandestine methamphetamine laboratories (1341); **2016** decontamination of personal protective equipment and related materials contaminated with toxic chemicals (1342);

Degradation of Drugs and Pharmaceuticals: **2014** a review of forced degradation and stability indicating studies of drugs (1343); determination of pharmaceutical impurities and degradation products by NMR (1344); **2015** analysis of degradation products from drugs by a rapid resolution LC-collision energy correlated-MS (1345); **2016** a stability-indicating UPLC-MS/MS assay for 1960's era pharmaceuticals in dosage forms (1346);

Education: **2013** use of forensic science to teach method development in undergraduate analytical laboratories (1347); the use of paper-based diagnostics with high school students to model forensic investigation and colorimetric analysis (1348); **2014** the use of forensic science and simulated crimes in a one-week long "Criminal Camp" to teach the theory and practice of basic concepts in chem., physics, medicine, and biol. (1349); using education to combat "chemophobia" (1350); a course for non-science majors at a college that looks at the chem. behind the crime itself, and the chem. behind the anal. of evidence from the crime (1351); a discussion for the need for forensic science programs to develop job-related skills in their students (1352); an overview of forensic science (1353); initiation and evolution of a forensic chemistry program (1354); the use of forensic chem.-themed activities to introduce fundamental concepts, such as the scientific method, to middle and high school students (1355); an overview of the chemistry behind forensic science (1356); use of presumptive and confirmatory tests using analogs of illicit drugs as an undergraduate instrumental methods exercise (using multiple color tests, GC-MS, and ATR-FTIR) (1357); utilizing the "CSI Effect" in chemistry instruction (1358); a discussion for the need, development, and implementation of an effective continuing forensic science education program (1359); a discussion of the need for robust and rigorous scientific research in academia based on need-based input from forensic practitioners who see the day-to-day issues in their laboratories (1360); an overview of the Forensic Science Education Programs Accreditation Commission's (FEPAC) accreditation program, the FEPAC stds., and the process involved in seeking FEPAC accreditation (1361); careers in forensic chemistry (1362); using *The Poisoner's Handbook* in conjunction with teaching a first-term general/organic/biochemistry course (1363); **2015** a universal internet-based prevention program for ecstasy and NPSs (for teenaged students) (1364); a discussion of the efforts to develop integrated forensic platforms that allow for the forensic investigation of human biol. traces, identification of illicit drugs, and the study of digital evidence (1365); a case study review of a problem based learning approach used to educate and train young forensic scientists through the use of six sigma investigative tools (using hydrolysis of cocaine to benzoylecgonine at various pHs as the teaching example) (1366); **2016** a performance task case study (misconduct) for teaching data analysis and critical thinking (1367); using a "Drug of the Week" approach to educate chemistry students about prescription drugs and their abuse (1368); use of a variety of small scenes using doll house furniture to educate criminal justice majors (1369);

Immunoassays: **2014** a review of the practical aspects of immunoassays and their application in clin. chem. for anal. of medicines and drugs of abuse (1370); the use cheminformatics to predict cross reactivity of "designer drugs" to their currently available immunoassays (1371);

Impurities and Impurity Profiling: **2012** a review of detection techniques for trace pharmaceutical impurities (1372); **2013** an overview (1373); a review of impurities in pharmaceuticals (1374); comparison of CCC and prep-HPLC for separating minor impurities in drugs (1375); **2014** analysis of impurities in drugs by LC-MS (1376); an overview of impurity profiling of pharmaceuticals (1377); a compendium of techniques for the analysis of pharmaceutical impurities, including TLC, HPTLC, HPLC, UPLC, GC., flash chromatog., SFC, CE, MECC, UV/Vis, IR, NMR, MS, LC/MS, LC-MS/MS, LC/NMR, HPLC/DAD-MS, HPLC/DAD/NMR/MS, UHPLC-MS, UHPLC-MS/MS, and chemometrics (1378); analysis and impurity identification in pharmaceuticals (1379); an overview of the impurity profiling methods for pharmaceuticals per current U.S. Pharmacopoeia guidelines (1380); **2015** method development for impurity profiling using SFC and comparing 6 different stationary phases (1381); an overview and review of recent advances in pharmaceutical impurity profiling (1382); a review of impurity profiling of drugs since 2010 (1383); development of an achiral SFC method with UV and MS detection for impurity profiling of drugs (1384 and 1385); an overview and review of impurity profiling of pharmaceuticals (1386); a review of impurity profiling, covering TLC, HPLC, HPTLC, LC-MS-MS, LC-NMR, LCNMR-MS, GC-MS, and LC-MS (1387); a review of impurity profiling of drugs (1388); **2016** an analysis of ionic interactions when characterizing 9 different stationary phases for drug impurity profiling with SFC (1389);

Inhalants: **2015** an overview of the abuse of nitrous oxide (1390);

Labelling and Packaging: **2012** examination of counterfeit labels on pharmaceuticals by IR and Raman (1391); **2013** a study on the effects of common drug packaging materials on nondestructive detection of contents by Raman spectrometry (1392); **2014** quantitative analysis of torn and cut duct tape physical end matching (1393); detection of counterfeit blister packaging by FTIR and chemometric methods (1394); a review of the identification of stamp impressions, including by microscopy, computer-assisted artificial identification, and anal. methods (UV-Vis, fluorometry, IR spectroscopy, Raman spectroscopy, TLC, LC, GC, and MS) (1395); **2015** evaluation of drug packaging by DSC (1396); determination of ethylene dichloride in drug packaging material made of polyethylene dichloride by headspace GC/ECD (1397);

Legal Issues: **2014** a regulatory perspective on the abuse potential evaluation of novel stimulant drugs in the U.S. (1398); **an effort to develop objective scientific methods to quantify and define the important "substantially similar" structural parameter used in several laws** (1399); **2015 a proposal for objective scientifically-derived measures of molecular structural similarity** (1400);

Precursors: **2013** impurity profiling of sassafras oils by GC×GC-TOF-MS (1401); **2014** a brief overview of the precursors for drugs of abuse (1402); determination of safrole in ethanol extract of nutmeg (*Myristica fragrans* Houtt) using RP-HPLC (1403);

Quality Assurance: **2013** use of a software tool (“Drugs WorkBook”) for the quantification of illicit drugs (1404);

Sampling Plans: **2013** a study of particle size of amphetamine, heroin, cocaine, and herbal cannabis and its influence on mass reduction (1405); a general sampling plan for the quant. instrumental anal. of heroin, cocaine, amphetamine, cannabis resin, MDMA tablets, and herbal cannabis (1406); a new sampling plan which focuses on sample heterogeneity (from ENFSI) (1407);

Sensors (Biological and Instrumental): **2013** a review of biological organisms as volatile compound detectors (stated applications include illicit drugs) (1408); **2014** assessing the potential of metal oxide semiconducting gas sensors for illicit drug detection markers (1409); use of a parasitic wasp as a biosensor for cocaine (1410); a review of biosensors in forensic analysis (1411); **2015** detection of illicit drugs by trained honeybees (1412);

Soil: **2012** forensic examination (reference text) (1413); **2014** by elemental analysis (1414); use of visible microspectrophotometry and FTIR/ATR for examination of soils for trace evidence (1415); **2016** protocols for soil examinations (1416);

Surveys of Drug Use: **2014** a comparative evaluation of whether computer survey technology improve reports on alcohol and illicit drug use in the general population (1417); an overview of 4 different systems utilized in Australia for monitoring drug use (1418); the use of internet snapshot surveys to enhance understanding of the availability of 2 NPSs (4-methylaminorex and 4,4'-dimethylaminorex) (1419); a survey of pharmacological cognitive enhancement among university students in the UK and Ireland who were abusing “smart drugs” (modafinil, methylphenidate, or Adderall) (1420); **2015** a measure of the “interest” in MDPV, methylone, 4-MEC, 4-HO-MET, MXE, 6-APB, AH-7921, and 3-MMC before and after its scheduling in Sweden (1421); an update on the Pistoia Alliance Controlled Substance Compliance Service Project (1422); a discussion and evaluation of the contents, the destinations, and the sources of 960 postal items seized by the Swiss customs authorities at the Swiss border between 2013 and 2014 (1423);

Other: **2013** collection of trace chemicals from diverse surfaces by use of strippable coatings (1424); the practical relevance of pattern uniqueness in forensic science (1425); **2014** a review of resolution by fractional crystallization of diastereomeric salts (1426); determination of water in active pharmaceutical ingredients using ionic liquid HS-GC and two different detection protocols (not identified in the abstract) (1427); reducing the complexity of an agent-based local heroin market model (1428); the examination of trace physical evidence and artificial materials (1429); use of an online database of chemical compounds for the purpose of structure identification (1430); **2015** a discussion of the comparison processes and evaluation systems that form a forensic intelligence framework, advocating scientific decision criteria and a structured but flexible and dynamic architecture (1431); an overview of ingestion of illicit drugs by “parachuting” (1432); the use of DNA sequencing analyses of the fungal diversity found in dust samples for geo-sourcing (1433); an assessment of the

toxicity of the refill liquids for electronic cigarettes (based on the presence of micro-organisms, diethylene glycol, ethylene glycol, hydrocarbons, ethanol, aldehydes, tobacco-specific nitrosamines, and solvents) (1434).

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Forensic Chemistry

Toxicology, 2013-2016

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1. Introduction

Forensic toxicology plays a vital role in scientific investigation or evaluation of the role of drugs or poison in cases with medico-legal consequences. The advancement of scientific field brings both challenges like emergence of new psychotic substances (NPS), as well as rapid growth of toxicology field. A large number of articles published in forensic toxicology every year thrive its development, including the innovations of laboratories round the world for solving continuous challenges in toxicology; the pursue of better scientific methods, practices and instrumentations for detection of drugs in biological specimens with higher reliability; and untiring efforts of worldwide toxicologists in establishing a concrete foundation for toxicology interpretations.

This review collected relevant publications in forensic toxicology since last review in 2013, covering the progress over the past 3 years from mid-2013. As a continuation of previous reviews, this article is divided into two parts, namely "Current Toxicology Issues" and "Advances in Toxicological Analysis".

2. Current Toxicological Issues

2.1 Driving Under the Influence

Driving under the influence of alcohol (DUI) and drugs continues to be a global concern. Numerous resources have been put into research for preventive measures. In this review, we summarize studies on prevalence of driving under the influence of alcohol and drugs, their impairments on driving, toxicological examination, alcohol pharmacokinetics and calculation as well as legislation regarding legal limits.

2.1.1 Survey on Prevalence of Driving Under the Influence of Alcohol & Drugs

Surveys on prevalence of driving under the influence of alcohol and drugs help to evaluate the effectiveness of preventative measures. These studies were conducted worldwide through analyzing data from traffic offence and accidents [1-26] as well as from roadside testing [27-34]. Most of these surveys revealed that apart from alcohol, cannabis and sedative-hypnotics were respectively the illegal and medicinal drugs significantly involved in driving under the influence.

2.1.1.1 Alcohol related traffic offence and accidents

Study on alcohol related traffic accidents in Australia showed an inverse relationship between monthly alcohol-related traffic crash rate and random breath testing (RBT) rate [1]. The findings suggested that as the number of RBTs conducted increases, the number of drivers willing to risk being detected for drink driving decreases.

Zhang *et al.* analysed the traffic violations data in Guangdong Province of China [2] and revealed that several factors associated with a significantly higher probability of both speeding and drunk driving were identified, particularly male drivers, private vehicles, the lack of street lighting at night and poor visibility. Another study on the alcohol-positive drivers involved in nonfatal traffic crashes in Shanghai [3] indicated that the vast majority of drivers were male. Both the mean blood alcohol concentration (BAC) and mean age of the male drivers were higher than female drivers. Moreover, most of the alcohol-related traffic crashes occurred in the evening and single-vehicle crashes involving cars had the highest percentage of occurrence.

The role of alcohol in fatal traffic accidents in Croatia was studied [4]. The results of the study showed that alcohol remained one of the main contributing factors of traffic accidents and victims of traffic accidents were mostly male drivers.

A 5-year overview of DUI in Italy was studied and alcohol consumption still remained a crucial factor in road accidents [5]. Another research concerning DUI offenses in Midwestern state [6] revealed that higher levels of alcohol risk were found in rural than urban DUI offenders. The results of the study indicated that rural DUI offenders had a significantly greater risk of heavy alcohol use when compared to urban DUI offenders.

The prevalence of alcohol consumption, helmet use and head trauma in cycling accidents in Germany was studied [7]. The cyclist and cycling accident characteristics associated with alcohol consumption and helmet use were established. Training initiatives on helmet protection should be encouraged as cyclists not wearing a helmet were more likely to have consumed alcohol.

2.1.1.2 Drugs related traffic offence and accidents

Driving under the influence of drugs (DUID) is a global traffic safety and public health concern. Many studies have been carried out to determine the trend and characteristics on the prevalence of drugs. The trends in drug use on drug impaired drivers in fatal traffic accidents was conducted [8]. The results showed that there was a general increase of prevalence of drug usage and the largest increases in broad drug categories were narcotics, depressants and cannabis. Similar observations were also found in other studies [9 - 11]. All the findings indicated that drug use was associated with a significantly increased risk of fatal crash involvement.

Many studies on the drug-related traffic accidents showed that the common illicit drugs found in the impaired drivers were cannabis, amphetamines, opioids, cocaine and benzodiazepines. The prevalence of cannabis increased significantly amongst other illicit drugs [12 - 14]. Studies on the newly emergence drugs found in the impaired drivers were done in different countries and these drugs included synthetic cannabinoids [15, 16], pregabalin [17], and methiopropamine [18]. The contribution of the drugs to the driving impairment is still need to be studied.

The prevalence of recent use of illicit drugs among truck drivers was studied by Peixe *et al.* [19]. The results revealed that the use of amphetamines and cocaine were common among professional truck drivers transporting grain loads. The impact of opioid analgesics on crash

responsibility in truck drivers was examined [20] and the prevalence of opioid analgesics in fatal crashes was found to be low.

Prescription drugs of abuse from the impaired drivers is on the rise [21, 22]. Sedative-hypnotics were commonly encountered by drivers apprehended for DUI in Finland [23]. Study in DUI cases in Finland in 2009 to 2011 revealed that temazepam was present in over half of the cases, along with other benzodiazepines such as midazolam and nitrazepam, and the non-benzodiazepine hypnotics: zopiclone and zolpidem. The prevalence of psychoactive prescription drugs was also found to be strongly associated with DUI [24, 25]. The use of prescription drugs with illicit drugs and/or alcohol can further increase the risk in impaired driving [10, 12, 26]. Out of the cases analyzed between 2009-2012 in Netherland, 94% (2842/3038) of the cases were detected medicinal and/or illicit drugs [12]. Medicinal drugs were found in 33% of the blood samples, with the highest prevalence for anxiolytics. In 86% of the cases illicit drug-positive results were obtained, with the highest prevalence for cannabis. Statistically significant associations between drug use and road traffic crash involvement were found, and that simultaneous use of two or more psychoactive drugs was associated with higher road traffic crash risk [10]. A retrospective 10-year study (2001-2010) road-traffic crashes when the driver had amphetamine and/or methamphetamine (MA) in autopsy blood in Sweden was conducted [26]. Amphetamine was present in the blood of 106 drivers (3.9%) either alone or together with other psychoactive substances (e.g. alcohol, cannabis, diazepam, alprazolam, etc.). Many of the victims (75%) had been arrested previously for use of illicit drugs or DUI.

2.1.1.3 Roadside testing for impaired drivers

Studies on roadside breath testing have been done to investigate the drink driving characteristics of impaired drivers [27]. A study revealed that the prevalence for DUI was considerably high at non-typical checkpoint hours [28]. Other studies indicated that driving after consumption of alcohol was highest during night-time hours of weekend days, followed by during day-time hours on weekend days, especially early in the morning and early in the evening [29, 30]. Moreover, the rate of alcohol-impaired drivers was higher for men than for women and it showed an increasing pattern with age [30]. The profile of women detected drink driving was studied and revealed that higher BACs were more common among younger women [31].

Roadside surveys on impaired drivers using breath testing and saliva screening were done in different countries. Amphetamines, cocaine, tetrahydrocannabinol (THC), benzodiazepines and zopiclone were the common detected drugs in saliva [32, 33] and some of these drugs were present as multiple drugs. Benzodiazepines (3.9%) and cocaine (3.8%) were the most frequently detected drugs in saliva in a study from Porto Alegre, Brazil. It is found that younger drivers in Australia were more likely to test positive for cannabis whilst older drivers were more likely to test positive for MA [34].

2.1.2 Effects of Alcohol & Drugs on Driving

Studies on driving impaired by alcohol and drugs help policy makers in deciding scopes of specified drugs and their legal limits. A study on driving impairments from social drinking concluded that driving and cognitive performance both showed dose-dependent alcohol impairment and showed strong placebo effects on ratings of subjective intoxication [35]. A review on driving offenses created by the intake alcohol and other drugs and their effects on driving behavior was published [36].

Being two most prevalent substances, the combined effects of alcohol and cannabis were studied with results indicated an increase performance impairment from cannabis-alcohol combinations [37, 38, 39]. In a study of simulated drives, participants with blood THC

concentrations of 8.2 and 13.1 µg/L during driving increased standard deviations of lateral position (lane weave, SDLP) similar to 0.05 and 0.08 g/210L breath alcohol concentrations. Cannabis-alcohol SDLP effects were additive rather than synergistic [37]. Drivers who had been tested for both drugs and alcohol after involvement in a fatal crash in the US (1991-2008) were examined [38]. The prevalence of THC and alcohol in car drivers involved in a fatal crash has increased approximately five-fold from below 2% in 1991 to above 10% in 2008. Drivers who were positive for both agents had greater odds of making an error than drivers positive for either alcohol or cannabis only. A study showed that vaporization is an effective THC delivery route [39]. The significantly higher blood THC and 11-Hydroxy-Δ⁹-THC (11-OH-THC) C_{max} values with alcohol possibly explained increased performance impairment observed from cannabis-alcohol combinations.

The effects of THC on response rate were compared to levels of THC and its metabolites in the blood of monkeys, indicated that blood levels do not predict behavioral or physiological effects of THC with different patterns of exposure [40]. A study on cannabis' psychomotor, neurocognitive, subjective and physiological effects in occasional and frequent smokers showed significant differences between the two types of smokers, suggesting tolerance development [41]. The analysis of variance between living (DUID cases) and deceased drivers' cannabinoid concentrations showed that 11-OH-THC and 11-nor-Δ⁹-tetrahydrocannabinol-9-carboxylic acid (THC-COOH) concentrations are not statistically different between the two groups, but that THC concentrations are statistically different, making it difficult to directly correlate postmortem with antemortem THC concentrations between living and deceased drivers [42].

In view of the gained popularity of synthetic cannabinoids, a number of studies on their impairments on driving were reported. A driver with symptoms of under the influence of drug was detected with UR-144 with concentration of 14.6 ng/mL together with its pyrolysis product in the blood [43]. Twelve cases of suspected impaired driving were discussed in which the drivers who subsequently tested positive for synthetic cannabinoid drugs (JWH-018, JWH-081, JWH-122, JWH-210, JWH-250, AM-2201) underwent a psychophysical assessment. The most consistent indicator was a marked lack of convergence. In all cases where a Drug Recognition Expert (DRE) officer evaluated and documented impairment (10 cases), it was attributed to the DRE cannabis category [44]. In another review related to 58 suspected impaired driving cases that were positive for AB-CHMINACA and AB-PINACA, the range of blood concentrations was 0.6->10 ng/mL for AB-CHMINACA (N=33) and 0.6-41.3 ng/mL for AB-PINACA (N=25) [45]. Overall, several physiological indicators varied from those typically observed with marijuana use. Slurred speech, confusion, lack of coordination/dexterity and lethargy were commonly observed. In a Norwegian study on concentrations of APINACA, 5F-APINACA, UR-144 and its degradant product in blood samples from impaired drivers, 5F-APINACA was found in one driver and three drivers was detected with both APINACA and 5F-APINACA in blood with concentrations from 0.24 to 24.5 and 0.9 to 6.5 µg/L, respectively, and UR-144 in two cases in concentrations of 0.22 and 0.47 µg/L. The data was compared to previous reported concentrations of other synthetic cannabinoids [46]. Cases of driving under the influence of various substances including a synthetic cannabinoid receptor agonist XLR-11 [47, 48], UR-144 [48] were reported with focus on sign of impairment and toxicology analysis.

A case of driving under the influence of methoxydiphenidine was reported with serum concentration determined to be 57 ng/mL [49]. The subject presented with amnesia, out-of-body experiences, bizarre behavior, and decreased motor abilities. In a drug driving case involving methoxetamine, the case sample was found to contain polydrugs with 10 ng/mL of methoxetamine detected in whole blood [50]. Several cases of individuals driving under the influence of synthetic phenethylamines focusing on analytical results and signs of

impairment were reported [51]. Consumption of synthetic phenethylamines can lead to impairments similar to those observed after the use of, for instance, amphetamine or 3,4-methylenedioxy-N-methylamphetamine (MDMA). A literature search on the psychomotor, cognitive, visual and perceptual functions of ketamine related to safe driving was conducted and an overview of ketamine and its congeners' clinical pharmacology issues, recreational psychoactive effects, and identification in biological specimens was also provided [52]. A population-based epidemiological study concerning the association of sedating/nonsedating antihistamines and fatal traffic accidents indicated that the risk of fatal traffic accident of those driving under the influence of sedating antihistamines was 1.61 (0.38 to 6.77, $P = .51$) times the risk of those without medication [53]. In a controlled study on the effects of dextromethorphan (DXM) on driving performance, a one-time dose of DXM 120 mg did not demonstrate driving impairment on the STISIM® Drive driving simulator or increase SFST failures compared to guaifenesin 400 mg [54]. A review on the association between kava (a herbal anxiolytic) use and motor vehicle crashes was published [55]. While no statistically significant adverse changes attributable to kava were found, there was weak evidence of slowed reaction time but one study was found to be significantly impaired when kava was consumed with alcohol.

2.1.3 Detection of Alcohol

Breath and blood are well recognized sample matrices for testing impairment by alcohol. A number of studies on detection of alcohol in these matrices were reported.

An article reviewed historical development, physiological principles and practical application of breath-alcohol analysis using in forensic science and legal medicine was published [56].

An evaluation on the accuracy of handheld pre-arrest breath test instruments (PBT) as a predictor of the evidential breath alcohol test results concluded that when maintained and operated by trained personnel, the PBT provided a reasonable estimate of the evidential test result. These results would be of value in evidential hearings seeking to admit the PBT results in drunk driving trials [57].

In view of the defense on positive breath alcohol test results due to the ingestion of homeopathic mother tinctures, the alcoholic content of three homeopathic mother tinctures and their ability to produce inaccurate breath alcohol was studied. The results indicated an observation period of 15-20 minutes prior to breath alcohol testing eliminated the possibility of false-positive results [58].

A comparison of breath alcohol concentration (BrAC) with BAC where BrAC test was to be administered, without, however, delaying the collection of the blood sample using real-life condition on drink-driving cases from the district of the Middle Hessian Police Headquarters suggested a conversion factor of 2.1‰ l/mg to German legislature as a new statutory value [59].

The effect of long-term room temperature storage on the stability of ethanol in whole blood specimens was investigated. After 5.6-10.5 years of room temperature storage, seven samples initially negative for alcohol remained negative while all samples initially positive for ethanol demonstrated a decrease in BAC over time with a statistically significant difference in loss observed based on blood sample volume whether or not the tube had been previously opened [60]. Tubes that were not previously opened and were more than half full demonstrated better BAC stability with 89% of these tubes demonstrating a loss of BAC between 0.01 and 0.05 g/dL.

2.1.4 Detection of Drugs

Urine, blood and oral fluid (OF) are the common sample matrices for toxicological analysis of specimens in DUID. Typical analyses involve drug screening by various immunoassay methods and then confirming and quantitation by gas or liquid chromatography-mass spectrometry.

2.1.4.1 Urine & Blood

Evaluation on immunoassay methods were undertaken included in investigation on cross-reactivity profiles of six benzodiazepines not included in the manufacturer's instructions (3-hydroxy-flunitrazepam, 7-amino-nitrazepam, brotizolam, delorazepam, pinazepam, α -hydroxy-midazolam) to EMIT® II Plus for urine testing [61]. The study showed that pinazepam, delorazepam and brotizolam are the most reactive molecules, while the other ones present a very low cross-reactivity. The use of EMIT® II Plus 6-AM immunoassay as an immunoassay screening test on 6-acetylmorphine (6-AM) for very recent heroin consumption in urine and blood was evaluated [62]. Based on the concordance between the results of the 6-AM immunoassay versus the LC-MS/MS, the sensitivity of the 6-AM assay was calculated as 100% and 95% for urine and blood respectively, with a specificity and accuracy of 100% for both biological samples.

Liquid chromatography-tandem mass spectrometry (LC-MS/MS) and ultra high performance liquid chromatography tandem mass spectrometry (UHPLC-MS/MS) are widely applied in drug confirmation and quantitation. A method for determination of delta-9-tetrahydrocannabinolic acid A, a marker for differentiating between the intake of prescribed THC medication and cannabis products, in blood or plasma using LC-MS/MS was established [63]. Another method to quantify 56 new psychoactive substances in blood and urine using LC-MS/MS, including amphetamine derivatives, 2C compounds, aminoindanes, cathinones, piperazines, tryptamines, dissociatives and others was developed [64]. A method for the confirmation of 22 metabolites from 11 parent synthetic cannabinoids by LC-MS/MS was reported [65]. The suitability of multi-analyte calibration approach in the analysis of authentic samples containing only one or two analytes was studied using a validated LC-MS/MS method [66]. Comparison of approximately 60 samples to a former gas chromatography mass spectrometry (GC-MS) method showed good correlation. The newly validated method was successfully applied to more than 1600 routine samples and 3 proficiency tests.

A reversed phase UHPLC-MS/MS method was developed for the quantitative analysis of the anti-epileptic compounds carbamazepine, carbamazepine-10,11-epoxide, gabapentin, lamotrigine, levetiracetam, oxcarbazepine, 10-OH-carbazepine, phenobarbital, phenytoin, pregabalin and topiramate in whole blood [67]. Using 0.1 mL sample volume with a simple protein precipitation with acetonitrile and methanol, the limits of quantification (LOQs) varied from 0.064 to 1.26 mg/L in blood. Another UHPLC-MS/MS method was established for the determination of zopiclone and zolpidem in whole blood for use in DUID and autopsy cases [68]. With the use of 0.1 mL of blood, the run time was 4.5min and the lower limits of quantification (LOQs) for zopiclone was 0.19 ng/mL and 1.10 ng/mL for zolpidem.

2.1.4.2 Oral Fluid

2.1.4.2.1 On-site Tests Development

Oral fluid (OF) has become a valuable matrix for toxicological analysis in DUID, because of easy and non-invasive collection procedures. The use of OF drug testing devices offers the ability to rapidly obtain a drug screening result at the time of a traffic stop. An evaluation of two such devices, the Dräger Drug Test 5000 with a seven drug panels (amphetamine, MA, cannabinoids, opiates, cocaine, benzodiazepines and methadone) and the Affiniton DrugWipe with five drug panels (amphetamine/MA, cannabinoids, opiates, cocaine and benzodiazepines) was conducted [69]. The Dräger Drug Test 5000 (DDT5000) and

DrugWipe returned overall sensitivities of 51 and 53%, and positive predictive values of 93 and 63%, respectively. The most notable difference in performance was the DDT5000's better sensitivity in detecting marijuana use. Both devices failed to detect benzodiazepine use. The ability of another OF screening device-the DrugWipe5S(®) to detect recent THC use in chronic cannabis smokers was also studied [70]. The DrugWipe5S(®) was positive just after smoking (90%); however, sensitivity rapidly decreased within 1.5 hours (50%). The performance of the Rapid STAT(®), DrugWipe5/5+(®) and Dräger DrugTest(®) 5000 on-site OF devices was evaluated with random OF specimens from car drivers in North Rhine-Westphalia (Germany) [71]. During an 11-month period, testing of 1,212 drivers showed that OF devices still show a lack of sensitivity (MA, benzodiazepines) and specificity (THC). A pilot study to test four substance use screening devices developed in Germany under local South African conditions and assess their utility for detecting DUID as part of the standard roadblock operations of local law enforcement agencies was conducted. OF was screened for drugs as per the standard calibrated cut-offs of all four devices. After alcohol, amphetamine, MA and cocaine were the most common drugs of impairment detected. [72].

2.1.4.2.2 Confirmatory Laboratory Tests Development

Various confirmation methods for prevalent drugs in driving under the influence were developed, with the methods on cannabinoids dominated. The quantification of THC, its metabolites, 11-hydroxy-THC and THC-COOH and other natural cannabinoids including tetrahydrocannabinol, cannabidiol, and cannabigerol in 1mL of OF [73] by LC-MS/MS was reported. An UHPLC-MS/MS method and a solid phase microextraction-GC-MS (SPME-GC-MS) method were also developed for the confirmation of THC in OF of small volume (100 µL) [74]. THC concentrations ranged from traces below the LOQ (2 ng/mL) up to 690 ng/mL. Another method for quantification of THC-COOH in human fluid collected with the Quantisal and Oral-Eze devices by GC-MS/MS was developed [75]. Extracted analytes were derivatized with hexafluoroisopropanol and trifluoroacetic anhydride and quantified by GC-MS/MS with negative chemical ionization. This method was capable to quantify THC-COOH in the concentration range of 10 - 1000 ng/L.

For other common drugs of abuse, a high-speed matrix assisted laser desorption/ionization (MALDI)-triple quadrupole-tandem mass spectrometry method for the determination of 3,4-methylenedioxymethamphetamine (MDMA) in OF was developed [76]. With MALDI omitting chromatographic separation, very short analysis times of about 10 s per sample were possible. A procedure for the simultaneous analysis of 20 illicit drugs, belonging to the classes of cocaine, amphetamines, natural and synthetic opioids and hallucinogens, by LC-MS/MS was reported, in which, the sample preparation was based on microextraction by packed sorbent (MEPS) [77]. LOQs ranged from 0.5 to 30 ng/mL.

The relationship of drug concentrations between OF and whole blood was evaluated for amphetamines, opioids, cocaine and metabolites, THC, benzodiazepines and for other psychoactive medicines using validated gas and liquid chromatography-mass spectrometric (LC-MS) methods [78]. Due to large variation seen in the study, it is suggested that drug findings in OF should not be used to estimate the corresponding concentrations in whole blood (or vice versa).

2.1.4.3 Other Matrix

A study on cannabinoids in exhaled breath following controlled administration of smoked cannabis by LC-MS/MS was reported. The results showing that breath may offer an alternative matrix for identifying recent driving under the influence of cannabis, but with sensitivity limited to a short detection window (0.5-2 hours) [79].

2.1.5 Alcohol Pharmacokinetic & Calculations

Widmark's equation is frequently used in the conversion of BAC to the amount of alcohol taken and vice versa. The equation continues to be a subject of research. Recently, two studies on the uncertainty measurement of Widmark's equation had been reported [80, 81]. It is noted that formulae for calculating the coefficient of variation in Widmark Factor Formulation were derived by Serale and showed that the coefficient of variation is not some fixed percentage but must be calculated in each case [80]. Gullberg offered further discussion on the issue and commented that the method of root sum square by summing the coefficients of variation to obtain combined uncertainty is not correct, since Widmark's equation is not strictly multiplicative. The equation has both additive and multiplicative terms, and therefore the method of error propagation is the correct application [81]. The resulting expanded measurement uncertainty for routine BAC determinations in a laboratory was determined to be 8% together with associated issues such as the role of measurement uncertainty in compliance assessment; the topic of the zero-alcohol limit from the forensic toxicology point of view; and the role of significant figures and rounding errors on measurement uncertainty and compliance assessment being discussed [82].

The differences between the measured BAC and the estimated concentration by Widmark's equation in elderly persons were reported [83]. The measured maximum BACs of the elderly participants were found significantly higher than the target BAC and that the calculated Widmark factors showed a high coefficient of variation, suggesting a tendency to an elevation of the actual BAC with increasing age.

The effect of the body mass index on the volume of distribution of ethanol was evaluated. It was found that the volume of distribution decreased with increasing body mass index for both sexes, suggesting fixed values for the volume of distribution of 0.7 L/kg and 0.6 L/kg for men and women, were mainly suited to judge underweight or normal weight people, but not obese persons [84].

Variation of breath alcohol elimination rate was studied as a function of age, gender, and drinking practice, factorially combined. Mean elimination rates (g/210 L/h) were found to be higher for women (N=84, M=.0182, SD=.0033) than for men (N=84, M=.0149, SD=.0029); higher for heavy drinkers (N=56, M=.0176, SD=.0038) than for light and moderate drinkers combined (N=112, M=.0160, SD=.0032); and higher for older subjects (51-69 years, N=42, M=.0180, SD=.0038) than younger subjects (19-50 years, N=126, M=.0161, SD=.0033). None of the two-way interactions (age \times gender, age \times drinking practice, gender \times drinking practice) or the three-way interaction (age \times gender \times drinking practice) was statistically significant [85].

Two studies performed by the same group on intra-individual and inter-individual variation in breath alcohol pharmacokinetics were reported [86, 87]. Participants of both sexes underwent serial breath alcohol concentration measurements after alcohol consumption. One study examined the short term variation (on two consecutive occasions, early evening and again the following morning) [86] while the other studied on three visits [87]. Widmark factors were determined to be 0.73-0.77 (short term) vs. 0.71-0.81 (three visits) in males and 0.61-0.64 (short term) vs. 0.59-0.68 (three visits) in females. Elimination rate was higher in the morning than evening in both males (7.4 vs. 5.7 μ g/100 mL/h) and females (6.9 versus 5.8 μ g/100 mL/h) while average elimination rates in males (5.3 μ g/100 mL breath/h, range 4-7.7) and females (5.6 μ g/100 mL breath/h, range 4-7) were not significantly different in three visits.

A comparison of breath-alcohol screening test results with venous BAC in suspected drunken drivers in Finland was carried out. Results indicated that with the assumption of a blood-breath alcohol ratio of 2260, reading from breath-alcohol test was lower than the

actual BAC by 15% on average and there was considerable uncertainty if screening test was used to estimate venous BAC. As a whole, the roadside breath-alcohol screening instruments worked well for the purpose of selecting drivers above the statutory limit [88].

2.1.6 Legal Limits on Drink & Drug Driving Law

2.1.6.1 Blood Alcohol Limits

Global report on Road Safety released by the World Health Organization indicated that 176 countries have a national drink-driving law in place, but only 134 of these are based on BAC limits (or equivalent breath alcohol concentrations). 84 countries have a drink-driving law based on BAC with a limit of less than or equal to 0.05 g/dL. Such laws are much more likely among high income countries than middle or low-income countries. As young and novice drivers are at a much-increased risk of road traffic crashes, 35 countries apply limits less than or equal to 0.02 g/dL for this high-risk group. As commercial drivers involved in drink-driving have more serious outcomes, 46 countries have set legal BAC limits for commercial drivers at less than or equal to 0.02 g/dL [89].

2.1.6.2 Drug Level Limits

There are a wide variety of psychoactive substances that have the potential to adversely affect driver behaviour. Lack of scientific evidence on the links between drug levels, impairment and crash risk for many drugs makes it difficult to set threshold limits for each substance. As a result, objective measures akin to BAC limits are largely lacking in most countries' laws on drug-driving. While 159 countries have national legislation prohibiting drug-driving, most of these laws do not define what substances are considered to be drugs. Some countries cite specific substances in their drug-driving laws by applying "zero tolerance", which simply reinforces laws relating to the illegal possession and consumption of drugs [89]. A handful of countries, however, include a list of drugs in their road traffic laws. For example, Luxembourg prohibits driving under the influence of cannabis, amphetamine, MA, morphine and cocaine. Other countries have moved towards specifying limits of drugs where threshold levels for crash risk have been established. For example, in the United Kingdom 8 generally prescription (6 benzodiazepines, methadone and morphine) (in risk based approach) and 8 illicit drugs (benzoylecgonine, cocaine, THC, ketamine, lysergic acid diethylamide, MA, MDMA and 6-monoacetylmorphine (6-MAM) (in zero tolerance approach) were added into new regulations that came into force in England and Wales on 2 March 2015. Regulations on amphetamine (in risk based approach) came into force on 14 April 2015 [89, 90].

Other studies on establishing legal limits were reported. A review on the state of drug-impaired driving in Canada indicated that driving after drug use is commonplace and is now more prevalent among young people than driving after drinking [91]. It is concluded that the government should establish per se limits for the most commonly used drugs, enforceable through a system of screening and evidentiary tests similar to the model used in Victoria, Australia. A review examined major considerations when developing threshold THC concentrations and specifics of legal THC limits for drivers adopted by 7 states in the US and other countries was reported [92]. With the exception of Iowa, all of these states list legal THC thresholds in whole blood in their respective drugged driving laws while 4 states have also set legal THC and/or THC-COOH limits in urine. Sixteen countries in Europe have set legal non-zero THC concentrations at or above which drivers are prosecuted for driving under marijuana. Unlike the US, no European countries have set legal THC and/or THC-COOH limits in urine. Another study discussed the proposed per se approach legislation in the UK on driving under the influence of cannabis with the consideration against current scientific evidence [93]. There is a significant dose-related decrement in driving performance following cannabis use which is much worse when cannabis and alcohol are

detected together. Patterns of use are important when interpreting blood concentration data and undoubtedly make setting thresholds for drug-driving legislation difficult.

2.2 Workplace Drug Testing (WDT)

Substance abuse in workplace is a major concern especially in the industry where safety is of utmost importance. Drug testing programs are established to help achieve a drug-free work environment, promote fair competition in sport, facilitate harm minimization and rehabilitation programs, better manage patient care by clinicians and service law enforcement authorities. Thus, guidelines from various organizations have been developed and updated to safeguard the public. Substance Abuse and Mental Health Services Administration (SAMHSA) proposed to revise the Mandatory Guidelines for Federal Workplace Drug Testing Programs (Guidelines) for urine testing and the proposal was published in May 2015 [94]. The proposed revision revised the initial and confirmatory drug test analytes to include prescription medications (*i.e.* oxycodone, oxymorphone, hydrocodone and hydromorphone) in federal drug-free workplace programs add methylenedioxyamphetamine (MDA) and methylenedioxyethylamphetamine (MDEA) as initial test analytes raise the lower pH cutoff from 3 to 4 for identifying specimens as adulterated. European Guidelines for Workplace Drug Testing in Urine from European Workplace Drug Testing Society (EWDTS) was updated in 2015 [95]. The recommended substances and maximum cut-off concentrations for tests in urine have been revised: for screening, cocaine metabolites from 300 to 150 g/L; methadone or metabolite of 300g/L revised to EDDP 100 g/L (or methadone 300g/L); for confirmation, cocaine metabolite (benzoylecgonine) from 150 to 100 g/L and buprenorphine or metabolites from 5 to 2 g/L. Also, recommended cut-off concentration of methaqualone was removed from the list.

WDT has been a focus in many countries. In Italy, Rosso *et al.* [96] examined how effective WDT is executed among the group of professional drivers. The method of WDT execution and drinking pattern of drivers are being investigated. Of 497 anonymous questionnaires collected, 21.4% drivers revealed that they have consumed alcohol at work. Drivers with high seniority or operating on international routes are more likely to consume alcohol at work. The author concluded that the current mode of WDT execution in Italy might not be effective.

Workers could be subject to hair drug testing if their urines are found positive for WDT. WDT in Italy as studied by Vignali *et al.* included two levels of monitoring: a first stage drug testing on urine samples and a second involving both urine and hair analysis for those workers who tested positive at the first level [97]. The study revealed second-level surveillance of WDT, which includes hair analysis, is more effective in identifying illicit drug use. In Canada, a ruling from the Supreme Court governs employer to test employees for alcohol and drug use in workplace. The request from employers must be based on a reasonable cause. Hurley [98] found that compared to mandatory testing, a policy of reasonable cause testing can significantly lower the probability of wrongly classifying a non-drug users to be drug users due to testing inaccuracy and human error. In Norway, Edvardsen *et al.* [99] examined the prevalence of alcohol and drugs use and their influence in workplace among a group of health professionals by analyzing data from self-reported questionnaires and OF. Alcohol was not detected in any of the samples. Ethyl glucuronide (EtG), a specific alcohol metabolite, was found in 0.3% of the collected samples. Illicit drugs and medicinal drugs were identified in 0.6% and 7.3% of the samples, respectively.

2.2.1 Approaches & Methodologies

WDT sees a high volume of disputes due to the consequences of failing a test. Positive results of WDT are being challenged in different ways. Numbers of research have focused on testing the validity of these challenges.

2.2.1.1 “Poppy seed defense” of Opiates

A glucuronide metabolite (designated 'ATM4G') which is originated from a tertiary amide compound, resulted from manufacturing byproducts from the synthesis of illicit heroin by reaction of acetic anhydride with the alkaloid impurity, thebaine, to produce compounds with a 2-(N-methylacetamido)ethyl side-chain was identified, and may be used as a marker of 'street' heroin administration to differentiate poppy seed ingestion and use of heroin [100]. In the study, urine samples taken from 22 known heroin users were positive for morphine but negative for MAM using a cutoff concentration of 10 ng/mL. These samples were further analyzed using LC-MS/MS to identify the marker compounds which were present in 16 out of 22 samples. The absence of MAM while present of the metabolite marker provides additional evidence for identifying heroin users in the argument of “poppy seed defense”.

In another study, Smith *et al.* [101] measured how much morphine and codeine concentration in urine after consuming poppy seeds of known opiate content. Two 45 g oral poppy seed doses, separated by eight hours and each containing 15.7 mg morphine and 3 mg codeine, were given to participants (N=22). Participants (N=22) provided 391 urine specimens over 32 hours following dosing; 26.6% and 83.4% were positive for morphine at 2000 and 300 µg/L GC-MS cutoffs, respectively. For the 19 subjects who completed the study, morphine concentrations ranged from <300 to 7522 µg/L with a median peak concentration of 5239 µg/L. No specimens were positive for codeine at a cutoff concentration of 2000 µg/L, but 20.2% exceeded 300 µg/L, with peak concentrations of 658 µg/L (284-1540).

2.2.1.2 Passive inhalation of Cannabis Smoke

Cone and coworker conducted a series of studies to explore exposure of non-smoker to secondhand cannabis smoke systematically to re-evaluate a popular defense argument against cannabis abuse: passive inhalation of cannabis smoke; six non-smokers were seated alternately with experienced smokers for three exposure sessions each with 1 hour duration [102-104]. No presumptive positives occurred for urine from non-smokers at 100 and 75 ng/mL; a single positive occurred at 50 ng/mL; and multiple positives occurred at 20 ng/mL. Maximum THC-COOH concentrations by GC-MS for non-smokers ranged from 1.3 to 57.5 ng/mL [102]. A total of 27 specimens out of 250 were found having a confirmatory concentration that exceeds 15ng/mL, a cutoff concentration for THC-COOH in urine, suggested by SAMHSA. THC-COOH concentrations generally increased with THC potency [103], but exposure under ventilated conditions resulted in much lower blood cannabinoid levels [104]. Minor physiological and subjective drug effects were reported and the psychomotor ability and working memory of non-smoker were slightly impaired after the exposure, similar to the effect of active cannabis smoking [103]. OFs and blood specimens from non-smokers and smokers were also collected and analyzed [104]. Positive tests for THC up to 3 hours following the exposure for OF and blood from non-smokers were obtained by LC-MS/MS with limit of quantitation (LOQ) = 0.5 ng/mL. No THC-COOH (LOQ = 0.02 ng/mL) in OF from nonsmokers was detected. These experiments mimic an extreme exposure condition in which the smoke was clearly visible and causes eye irritation. Thus, the authors suggested that although many variations such as length of exposure and potency of cannabis use in real scenario are unknown, it is very unlikely for cannabis smoke in less extreme conditions to cause a positive result in testing and behavioral change.

2.2.1.3 Distinguishing Coca Leave chewing from Cocaine abuse

The practice of coca leaves chewing, which is legal in some counties such as Argentina and Bolivia, complicates the identification of cocaine abuser in WDT. Rubio *et al.* investigated the possibility of using hygrine and cuscohygrine as markers to distinguish coca leave chewing from cocaine abuse and developed a GC-MS method to analyze these markers

qualitatively. 24 urine samples of coca leaves chewer with different habit of chewing and occupations together with 38 urine samples of cocaine abusers from forensic cases in Spain and Argentina were investigated [105]. Hygrine and cuscohygrine were identified qualitatively in urine specimens of all coca leaves chewers while none of them were found in those of cocaine abusers. The instability of cuscohygrine, which will degrade to hygrine rapidly, makes quantification of the marker compounds difficult and warrants that identification of these markers in urine should be done sooner rather than later. The study was extended in discrimination between chewing coca leave (26 Argentinean coca chewers) and the abuse of cocaine (22 German cocaine users) using hair analysis [106]. It was found that the ratio of cuscohygrine, cinnamoylcocaine and ecgonine methyl ester to cocaine ratios could serve as criteria for the discrimination between both groups with the means and medians 5-fold to 10-fold higher for coca chewers and a low overlap of the ranges between both groups.

2.2.1.4 Medicinal Use of Amphetamines

People who tested positive for amphetamines abuse may argue that the result is due to prescription or over-the-counter medications such as intranasal decongestants containing *L*-methamphetamine (*L*-MA). Huestis and co-workers examined the potential of commonly used immunoassays to distinguish legitimate use of intranasal decongestants from amphetamines abuse and confirmed the results using an enantiomer-specific GC–MS with >99% purity of R-(–)- α -methoxy- α -(trifluoromethyl)phenylacetyl derivatives and 10 μ g/L LOQs [107]. No D-amphetamine or D-MA was produced in urine from 22 healthy adults following controlled Vicks VapoInhaler administration, an intranasal decongestants containing *L*-MA, at manufacturer's recommended doses. Concentrations of *L*-MA in urine from two participants can exceed 250 μ g/L, the cutoff concentration of confirmatory test suggested by SAMHSA. The median *L*-MA maximum concentration was 62.8 g/L (range: 11.0–1,440). Rewarded by the high efficiency of initial immunoassay screening (>97%), an enantiomer-specific confirmation for amphetamine positive specimens suspected of inhalation of Vicks VapoInhaler would not be frequent.

2.2.1.5 Urine Authenticity

Diluted, substituted, or adulterated urine specimen in WDT is considered invalid. Urine providers may argue that legitimate reasons such as medical condition attributes to the invalidity. Holden *et al.* [108] examined the concentration of creatinine of toxicology urine samples and concluded that creatinine under 20 mg/dL does not necessarily imply a sample adulteration. Another study on the effect of creatinine normalization of drug values from diluted urine sample also pointed out the importance of creatinine normalization in the improvement of first-level WDT [109].

2.2.2 Analytes & Matrices

Although urine is usually the specimen of choice and the focus on WDT guidelines, guidelines related to alternate specimens are surfacing. A proposal from SAMHSA [110] aimed to establish scientific and technical guidelines for the inclusion of OF specimens in the Mandatory Guidelines for Federal Workplace Drug Testing Programs (Guidelines) was published. The Department of Health and Human Services (HHS) projected that approximately 7% (or 10,500) of the 150,000 specimens tested per year will be OF specimens. Information related to hair specimen analysis including specimen collection, preparation and cutoffs are publicly consulted [111]. Similar guidelines from EWDTs for testing OF and hair are also updated recently [112-113]. While cut-off concentrations in screening tests in OF are more or less the same, the updated EWDTs guidelines required a lower cut-off concentrations for confirmation: amphetamines from 30 to 15 ng/mL; benzodiazepines from 10 to 3 ng/mL; opiates from 40 to 15 ng/mL; buprenorphine or metabolites from 5 to 1 ng/mL and propoxyphene or metabolites from 40 to 5 ng/mL. For

testing in hair, minor changes in recommended cut-off includes: i) THC concentration from 0.05 to 0.1 ng/mg (screening) and 0.05 ng/mg to 0.05 ng/mg (confirmation) and THC-COOH 0.2 pg/mg to 0.0002 ng/mg (confirmation); and ii) addition of cut-off concentrations for methadone and buprenorphine (ng/mg): 0.2 and 0.01 respectively for screening; 0.2 (methadone), 0.05 (EDDP); 0.01 (for both buprenorphine and norbuprenorphine) for confirmation. Biological matrices other than urine are gaining weight in WDT.

2.2.2.1 Urine for workplace drug testing

Urine remains the most popular and appropriate testing matrix for WDT. Although a number of techniques can be used for drug testing in urine, immunoassay is the most common screening technique. It is rapid, simple and relatively cost effective. In recent years, designer piperazines are emerging novel psychoactive substances (NPS) with few high-throughput screening methods for their identification. Huestis and co-workers continued to contribute to the evaluation/development of methods in detection of piperazines, cathinones and synthetic cannabinoids [65,114,115]. The evaluation of the use of biochip array technology (BAT) immunoassay as a high-throughput screening method for identification of phenylpiperazines and benzylpiperazines in 20,017 randomly collected urine workplace specimens was reported [114]. All presumptive positive specimens (N= 840) and randomly selected presumptive negative specimens (N=206) were analyzed and confirmed by a liquid chromatography high-resolution mass spectrometry (LC-HRMS) with LOQ of 2.5 or 5 µg/L. They found that the performance of the Randox BAT immunoassay was improved when antibody cutoffs were raised. Besides, determination of trazodone in confirmative method is recommended because screened and confirmed positive piperazine result could be due to legitimate use of trazodone. Like the designer piperazines, synthetic cathinone, is also lack of a high-throughput immunoassay screen. The evaluation of the performance of the Randox Drugs of Abuse V (DOA-V) Biochip Array Technology (two synthetic cathinones antibodies: Bath Salt I (BSI) targets mephedrone/methcathinone and Bath Salt II (BSII) targets MDPV/ MDPBP) in detection of 28 synthetic cathinones in 20,017 authentic military urine specimens, that the presumptive positive specimens were confirmed by LC-MS/MS was reported [115]. The immunoassay method was fully validated with sensitivity, specificity, and efficiency of 100%, 52.1%, and 53.0% obtained at manufacturer's proposed cut-offs (BSI 5 µg/L, BSII 30 µg/L). Performance improved if cut-off concentrations increased (BSI 7.5 µg/L, BSII 40 µg/L). Parent synthetic cannabinoids are rarely detected in urine, the most common matrix employed in WDT. A comprehensive and optimized urine quantitative LC-MS/MS method for identification of synthetic cannabinoid markers in authentic and randomly collected urine specimens was developed [65]. 20,017 randomly collected US military urine specimens were screened with a synthetic cannabinoid immunoassay yielding 1432 presumptive positive specimens with 290 specimens confirmed to be positive using a qualitative synthetic cannabinoid LC-MS/MS method. The five most predominant metabolites were JWH-018 pentanoic acid (93%), JWH-N-hydroxypentyl (84%), AM2201 N-hydroxypentyl (69%), JWH-073 butanoic acid (69%), and JWH-122 N-hydroxypentyl (45%). The study improved the interpretation of synthetic cannabinoid urine test results and suggested suitable urine markers of synthetic cannabinoid intake.

Traditional abused drugs such as opiates play a relevant role in forensic toxicology and their assay in urine or blood is usually performed for example in WDT or toxicological investigation of drug impaired driving. The recent work by Chericoni *et al.* [116] described two new validated methods for detecting morphine, codeine and 6-MAM in human hydrolysed and unhydrolysed urine using a single step derivatisation (using propyl chloroformate) in aqueous phase, followed by liquid-liquid extraction and GC-MS to detect the derivatives. The validated methods were applied to real case samples.

Previous reports revealed poor performance in identifying drugs of abuse users through first-level WDT based on urine samples. Crespi *et al.* studied the effect of creatinine normalization of drug values from diluted urine samples (creatinine levels ≤ 20 mg/dL) on the prevalence of drug user and the independent procedure-related predictors of positivity and dilution [109]. The workers' urine samples were collected at the workplace or at their certified laboratory and analyzed for drugs of abuse by immuno-enzymatic method according to the Italian WDT law between 2008 and 2012. Detectable drugs of abuse concentrations lower than the positive cutoff values were normalized based on mean levels of urinary creatinine. Detectable concentrations of drugs were confirmed by GC-MS. Seventeen out of 23 diluted urine samples with detectable concentrations of cannabinoids or cocaine were found positive after urine creatinine normalization.

2.2.2.2 Oral Fluid for workplace drug testing

Oral fluid (OF) is a new biological matrix for clinical and forensic drug testing, offering non-invasive and directly observable sample collection reducing adulteration potential, ease of multiple sample collections, lower biohazard risk during collection, recent exposure identification, and stronger correlation with blood than urine concentrations. A review of the current knowledge of OF cannabinoids including evaluating pharmacokinetic properties, detection windows, and correlation with other biological matrices and impairment from field applications and controlled drug administration studies was published [117]. Onsite screening technologies, confirmatory analytical methods, drug stability, and effects of sample collection procedure, adulterants, and passive environmental exposure were also reviewed. OF research over the past decade demonstrated that appropriate interpretation of test results required a comprehensive understanding of distinct elimination profiles and detection windows for different cannabinoids, which were influenced by administration route, dose, and drug use history. Thus, cut-off criteria, collection/analysis procedures and storage conditions tailored to each of the drug testing program should be established. A number of OF cannabinoids research on passive environmental exposure, drug use history, donor physiological conditions, oral cavity metabolism needed to better understand mechanisms of cannabinoid OF disposition as well as expand OF drug testing applicability were ongoing as so to build scientific basis for OF testing.

The detection of THC-COOH in OF offers the advantage of documenting active consumption, as it is not detected in cannabis smoke. Analytical challenges such as low (ng/L) THC-COOH OF concentrations hampered routine OF THC-COOH monitoring. A sensitive and specific LC-MS/MS quantification method without lengthy derivatization procedures for THC, its metabolites (11-OH-THC and THC-COOH) and other natural cannabinoids (tetrahydrocannabivarin (THCV), cannabidiol (CBD), and cannabigerol (CBG)) has been developed and validated by Desrosiers *et al.* [73]. All analytes were monitored in positive mode atmospheric pressure chemical ionization (APCI) with multiple reaction monitoring. LOQs were 15 ng/L THC-COOH and $0.2 \mu\text{g/L}$ for all other analytes.

Oxycodone (OC) is recommended to be included as an analyte tested in the proposed SAMHSA's Mandatory Guidelines for Federal Workplace Drug Testing Programs using OF Specimens. The study by Cone *et al.* [118] demonstrated the time course of OC and metabolites, noroxycodone (NOC), oxymorphone (OM) and noroxymorphone (NOM), in near-simultaneous paired OF and whole blood (BL) specimens by LC-MS/MS. A single dose of OC 20 mg controlled-release was administered to 12 healthy subjects followed by specimen collections for 52 hours. The period of detection for OF exceeded BL by ~ 2 -fold at similar cutoff concentrations. At a 1 ng/mL cutoff for OF, the mean detection time was 34 hours for OC and NOC.

In Australia, it is a requirement of workplace OF drugs of abuse testing that drug recovery from collection devices should be verified by an accredited laboratory. Recovery data are used in conjunction with collection volume imprecision data and uncertainty of measurement to provide an estimation of drug concentration in neat OF. Hall *et al.* [119] reported a recovery study of drugs of abuse by using Drager DCD5000 Oral Fluid Collection Device. Based on the results obtained from the device using isopropanol, the overall drug's recovery from the device was found to be acceptable, ranged from 86 to 102%, when the increased collection volume of the swab was taken into account. However, recovery of a metabolite of methadone, (2-ethylidene-1,5-dimethyl-3,3-diphenyl-pyrrolidene (EDDP), was only 45%. It suggested that urine was more suitable sample if methadone therapy was being monitored.

2.2.2.3 Hair for workplace drug testing

Strano-Rossi *et al.* [120] has developed an UHPLC-MS/MS to analyze several classes of drug in hair (30 mg), pertaining to different chemical classes (synthetic cannabinoids, synthetic cathinones, ketamine, piperazines and amphetamine-type substances-ATS) with lower limit of detections (LODs) varied from 2 pg/mg to 20 pg/mg. The method is successfully applied in real forensic cases including WDT with the detection of various synthetic cannabinoids (JWH-018, AM694, JWH-081 etc), Methylendioxypropylone (MDPV) and 4-Methylethcathinone (4-MEC), ketamine, pseudoephedrine and cathine. Nieddu *et al.* [121] validated an LC-MS/MS method to simultaneously quantify 11 phenethylamines, which are banned in Italy, in hair. In vivo study using this method on the effect of hair pigmentation on retaining the targeted phenethylamines is consistent with the literatures that black hair accumulates drugs more easily than grey or white hair.

Agius *et al.* [122] presented a screening method to detect drugs of abuse in hair and in urine using LUCIO-direct ELISA kits covered six classes of drugs including amphetamines, cannabinoids, cocaine, opiates, methadone, and benzodiazepines. A large quantity of authentic hair (N = 9000) and urine (N = 33,262) samples is used to validate the immunoassay tests against the guidelines of German driving licence re-granting medical and psychological assessment, which have lower cut-offs values than conventional WDT guidelines. For almost all screening tests in hair and urine, the area under the curve were greater than 0.8 from the plot of receiver operating characteristics, indicating good to excellent performance.

The effect of cosmetic treatment on hair drug and alcohol testing is evaluated in non-treated hair (n = 9488) and cosmetically treated hair (n = 1026) [123]. Statistically, the positive rate of drugs (THC, cocaine, amphetamine) and EtG has no significant difference in treated and non-treated hair. Differences in concentration range detected have been observed for the effect of cosmetic treatment only at high drug concentrations. These observations warrant the analytical and forensic values of treated hair in drugs and alcohol testing, particularly for testing that is against cut-off values and quantitative results are not required. Kidwell *et al.* [124] revealed that some products increased the MA and cocaine concentrations in all hair types (Caucasian, Asian, and African). Certain ethnic hair products can replace moisture as a diffusion medium, thereby increasing the susceptibility to contamination over 100-fold compared to petroleum-based products.

2.2.2.4 Nail for workplace drug testing

Nail is a biological matrix recently used in toxicology, which is keratinized and produced in the germinal matrix and thickened by the nail bed. Nail samples are believed as a suitable alternative to hair due to the similar window of detection and non-invasive sample collection [125-126]. Drug testing of nail samples collected from high-risk cases during a 3-year period of time in the US observed with positivity rates: amphetamine and MA (14% out of total of 7799), cocaine and related analytes (5% out of total of 7787), oxycodone (15.1%),

hydrocodone (11.4%), THC-COOH (18.1%). Out of 3,039 samples, 756 were positive (24.9%) for EtG [126]. Moreover, less cosmetic treatment, no growth cycle and pigment, and ready availability even under different medical conditions such as alopecia and reception of chemotherapy are the remarkable advantages over the use of hair. Toenail is sometimes more appropriate for drug monitoring than fingernail in term of the risk of contamination [127].

The concentrations of THC-COOH in fingernail clippings were over 4 times greater than that in the corresponding hair specimens in a study conducted by Jones *et al.* [128]. Both studies proposed that nail is an alternative to hair for monitoring drugs of abuse.

The use of EtG in fingernail was studied. Berger *et al.* monitored the concentrations of EtG in fingernails and hair from 606 undergraduates (age of 18 to 25) by LC-MS/MS and assessed by participant interview using the time-line follow-back method [129]. The study suggested that it may have potential for EtG in fingernail as a quantitative indicator of alcohol use. A study conducted by Karanfil *et al.* [130] has collected hair and nail specimens from 16 people taking alcohol. Results showed that EtG concentrations were 1.33-65.67 (+/- SD16.57) ppb in hair specimens and 4.27-225.03 (+/- SD 59.77) ppb in nail specimens. Hair EtG concentrations were correlated with nail EtG concentrations ($r=0,808$, $p<0.001$) and can be useful in people without hair.

2.3 Drug-facilitated Sexual Assault (DFSA)

In case such as Drug Facilitated Sexual Assault (DFSA), victims frequently delay or do not report the crime and extended delays in sample collection will lower the probability of drug detection. To address these difficulties encountered in the toxicological analysis of exhibits in connection with DFSA, a book focusing on selection of biological sample and interpretation of drug found in connection with Drug Facilitated Crimes (DFC) was published [131]. It covered the pharmacology and pharmacokinetics of drug commonly encountered in DFC. A number of cases were also listed as references. A study of 264 patients who consulted a sexual assault center in Trondheim, Norway over an 8-year period from July 2003 to December 2010 was reported [132]. The study showed that alcohol was the most commonly found drug in DFSA cases. Interestingly none of the report cases tested positive for GHB. In another study of urine samples of sexual assault victims submitted to University of Miami toxicology laboratory for analysis in 2013 [133], it was found that methylone has become a popular drug encountered in these cases. Of the forty-five urine samples submitted, 13% were positive for methylone.

2.3.1 Blood & urine Analysis for DFSA

Traditionally, urine and blood were the specimens of choice for the analysis of DFSA cases and a number of methods were developed in the past three year for the analysis of drugs implicated in DFSA cases in urine [134,135,136,137] and in blood [138]. The analysis of zopiclone in biological sample associated with DFSA was problematic because their degradation in blood was very fast. A method was developed and validated by Nilsson *et al.* for the analysis of zopiclone and its major metabolites in urine [139]. The results showed that formation of 2-amino-5-chloropyridine occurred at elevated pH and/or temperature by degradation of zopiclone and its major metabolites. A new dispersive solid phase extraction method coupled with LC-MS/MS was also developed for the analyses of drugs in blood samples. A method for determination of drugs of abuse and benzodiazepines in blood was validated by Anzillotti *et al.* which employed a modified "QuEChERS" procedure followed by UHPLC-MS/MS analysis [140]. LODs of the method were 0.5 ng/mL for all benzodiazepines tested while for drugs of abuse LODs varied from 0.05 to 2 ng/mL. This method was applied on a number of DFSA cases and cocaine and benzodiazepines were detected in the blood samples of these cases.

2.3.2 Hair Analysis for DFSA

In DFSA, it was not rare for the delay between incidents and the sample collection time to exceed several days or weeks. In such situation, hair was generally the only matrix able to establish the involvement of drugs in crime owing to its long detection window. In the past three years, numerous methods were developed for the detection of a single drug or multiple drugs in hair specimen. Miyaguchi *et al.* has reported a method for quantification of 13 major sedative-hypnotics in human hair by LC-HRMS [141]. This method required a smaller amount of sample (5 mg of hair sample) and the LOQ ranged from 1 pg/mg to 4 pg/mg. Maublanc *et al.* has developed a multianalysis method for the simultaneous detection and quantification of 35 psychotropic drugs in hair [142]. These 35 psychotropic drugs have already been implicated in DFC. LODs and LOQs ranged between 0.5 to 5 pg/mg depending on the analytes, except for lorazepam (10 pg/mg). This method has been successfully applied in 24 suspected DFC cases. Nieddu *et al.* presented a simple procedure for the simultaneous quantification of 11 phenethylamines in hair [121]. The LODs and LOQs of this method ranged from 0.03 to 0.07 ng/mg and from 0.09 to 0.20 ng/mg, respectively. Segmental analysis makes it possible to explore the consumption history and to distinguish chronic usage from a single exposure. An UPHLC-MS/MS method was developed and validated by Jakobsson *et al.* for quantification of amphetamine, MA, 3,4-methylenedioxyamphetamine and MDMA in hair sample [143]. The method was applied to samples from a controlled study of amphetamine intake as well as forensic hair samples previously analyzed with UHPLC-TOF-MS screening method.

The abuse of GHB and its implication in cases of suspected DFSA was of keen interest to forensic toxicology laboratories. Jagerdeo *et al.* described a procedure which combined liquid extraction, solid phase extraction, chromatographic separation and LC-MS/MS for detection and identification of GHB in hair [144]. This procedure has been applied for a survey of hair specimens from individual with no known exposure to exogenous GHB in order to determine a cut-off value for GHB in hair. A wide range of endogenous GHB levels were observed in these hair samples (from less than 1.2 to 4.4 ng/mg) and it overlapped with the GHB concentrations reported for hair samples obtained from individual suspected of having been exposed to exogenous sources of GHB. Bertol *et al.* developed and fully validated a method for the detection of GHB in human hair by LC-MS/MS after liquid extraction [145]. This method has been applied to hair sample of 30 non-GHB user to determine the basal level of GHB in hair and showed no significant difference in black (2.34 ± 1.54 ng/mg), blonde (2.51 ± 1.36 ng/mg) and dyed (2.65 ± 0.99 ng/mg) hair samples. The segmental analysis of hair samples (5mm/segment) collected one month and two months after the oral administration of a single dose of GHB in 12 healthy volunteers allowed the calculation of two ratios (4.45:1 and 3.35:1, respectively), a lower value than the ratio 10:1 provided by United Nations Office on Drugs and Crime (UNODC) was recommended the extent of GHB elevation for a positive identification of GHB intake. Controlled dose studies of drug concentration in hair after administration would improve our understanding of interpretation of drug found in hair. Shi *et al.* has reported a GC-MS/MS method capable of quantifying the endogenous level of GHB in human head hair [146]. This method has been applied for determination of the endogenous GHB in hair samples of 66 drug-free Chinese donors. The mean male endogenous GHB level was 2.95 ng/mg (0.92-4.91 ng/mg, n=35), while the mean female level was 0.77 ng/mg (0.28-1.95 ng/mg, n=31).

Cui *et al.* has reported a study on the segmental hair analysis after a single dose of zolpidem [147]. Hair was collected one month after administration of 10mg dose of zolpidem to 20 volunteers and analyzed by UHPLC-MS/MS after alkaline digestion in 0.1 M of NaOH. Zolpidem concentration was found in the range of 135.0-554.6 pg/mg. This result was controversial as revealed by Kintz and co-workers [148] by collecting data from six

laboratories of zolpidem in hair after a single exposure in 49 cases, the drug was found in the range of 0.5–47 pg/mg, with a mean measured value of 11.3 pg/mg, and has sparked a series of discussion. Shima *et al.* documented a sensitive method for detection of zolpidem in a 2 cm strand of hair [149]. This method combined a newly established LC-MS/MS procedure in conjunction with a one-pot pulverization extraction method. The detection limit of zolpidem was 50 fg/2-cm single hair. Fifteen strands of hair sample from one volunteer, who took a single oral dose of 10 mg zolpidem tartrate 35 days earlier, were analyzed and zolpidem was detected in 14 hairs. The amounts of zolpidem detected in each positive 2-cm segment of single hair ranged from 27 to 63 pg (average 43 pg).

Xiang *et al.* has published a review on the drug concentration in hair in control study as well as in reported DFC [150]. In this review hair concentrations of 35 psychoactive drugs given in 20 controlled dose studies were presented and compared to 25 different drugs detected in reported case work. Those concentrations reported in DFC were mostly similar to or higher than those reported in controlled studies. The factors affecting interpretation of segmental hair results including hair color, growth rates, sample preparation and surface contamination are discussed. Maublanc *et al.* reported the finding of prazepam in hair of a DFSA victim [151]. Prazepam was rarely included in toxicological assays in blood because it was known to be totally and rapidly metabolized to nordiazepam, oxazepam and 3-hydroxyprazepam after oral intake. This finding raised question about the methods that did not look for prazepam in hair and in particular about those methods that used prazepam as internal standard. Not only these methods would not detect prazepam in hair, they would also underestimate the concentration of other benzodiazepines that was present in the hair sample.

2. 4 New Psychoactive substances

A large number of new psychoactive substances (NPS), also called "legal highs" or "designer drugs", have emerged in recent year. NPS can be chemically classified as phenethylamines, amphetamines, synthetic cathinones, piperazines, pipradrols/ piperidines, aminoindanes, benzofurans and tryptamines [152]. Synthetic cannabinoids are another group of novel substances which all act as agonists at the cannabinoid CB1 receptor similar to THC but are chemically diverse. According to the report of UNODC, a total of 541 NPS were reported up to December 2014 [153]. In 2014, 101 NPS were reported to European Monitoring Centre for Drugs and Drug Addiction (EMCDDA) of which 30 were synthetic cannabinoids [154].

2.4.1 Synthetic Cannabinoids

Synthetic cannabinoids, also known as "spice" or "herbal high", were initially synthesized in the early 1960s following the discovery of the structure of THC [155] and began to appear on the market in 2004 [156], are formulated to interact with endogenous cannabinoid receptors in brain to produce psychoactive cannabis-like effect [157]. It was the most frequently discovered NPS between 2005 and 2013 according to the finding of the EU early warning system (EWS) run by the EMCDDA [154, 158] and the number of synthetic cannabinoids detected through the EWS continues to grow from 9 reported in 2009 to 30 reported in 2014 with a total of 134 synthetic cannabinoids having been notified to the EMCDDA as of December 2014 [154]. Synthetic cannabinoids can be chemically classified into naphthoylindoles, benzoylindoles, phenylacetylindoles, adamantylindoles, cyclophenols and other miscellaneous group. Five synthetic cannabinoids (JWH-018, JWH-073, JWH-200, CP47,497 and CP-47,497 C8 homologue) has been scheduled as schedule 1 controlled substances by the United States Drug Enforcement Administration (DEA) in March 2011 [159] and more were scheduled later.

2.4.1.1 Studies of Synthetic Cannabinoids Metabolites

Hegstad *et al.* reported the detection times of carboxylic acid metabolites of JWH-018 and JWH-073, first generation of synthetic cannabinoids. The mean elimination half-life in urine were found to be 14.0 (range 4.4-23.8) days and 9.3 (range 3.6-16.8) days for CN-JWH-018-COOH and CN-JWH-073-COOH, respectively. It was suggested that the urine specimens could be positive for JWH-018-COOH and JWH-073-COOH respectively for more than 6 weeks and 3 weeks [160].

Strano-Rossi *et al.* performed a pilot study on the main metabolic reactions for four synthetic cannabinoids: JWH-015, JWH-098, JWH-251 and JWH-307. Rat liver slice samples were analyzed by liquid chromatography-quadrupole time of flight (LC-Q-TOF) and identification of metabolite was executed using Mass-MetaSite™ software. The experimental findings found to be in good accordance with the silico prediction of main metabolic reactions performed using MetaSite™ software [156]. The MetaSite software was also used in the metabolite profiling study of N-(1-amino-3-methyl-1-oxobutan-2-yl)-1-pentyl-1H-indazole-3-carboxamide (AB-PINACA) and 5-fluoro-AB-PINACA (5F-ABPINACA) by Wohlfarth *et al.* Metabolites obtained using human liver microsomes as well as authentic urine specimens from AB-PINACA cases were analyzed by LC-HRMS and the data were analyzed with MetaolitePilot using different data processing algorithms. 23 metabolites of AB-PINACA and 18 metabolites of 5F-AB-PINACA were identified. Similar metabolic profiles were found in the two authentic urine specimens from suspected AB-PINACA and those obtained using human liver microsomes. AB-PINACA was predominantly hydrolyzed to AB-PINACA carboxylic acid, carbonyl-AB-PINACA, and hydroxypentyl AB-PINACA, likely in 4-position. The most intense 5F-AB-PINACA metabolites were AB-PINACA pentanoic acid and 5-hydroxypentyl-AB-PINACA. The *in vitro* half-life for AB-PINACA was found to be 18.70.4 min and the intrinsic clearance found to be 35 mLmin⁻¹kg⁻¹. For 5F-AB-PINACA, the *in vitro* half-life was 35.93.0min with intrinsic clearance of 18 mLmin⁻¹kg⁻¹ [161]. Metabolites of THJ-018 (1-naphthalenyl(1-pentyl-1H-indazol-3-yl)-methanone) and THJ-2201 ((1-(5-fluoropentyl)-1H-indazol-3-yl)(naphthalen-1-yl)methanone), indazole analogs of JWH-018 and AM-2201, were studied by incubating 10 mol/L THJ-018 and THJ-2201 in human hepatocytes for 3 hours. TripleTOF® mass spectrometer was used to identify the metabolites after incubation and MetaSite was used to perform the silico metabolite predictions [162]. Thirteen THJ-018 metabolites were detected, with the major metabolic pathways being hydroxylation on the N-pentyl chain and further oxidation or glucuronidation. For THJ-2201, 27 metabolites were observed, predominantly oxidative defluorination. MetaSite prediction matched well with THJ-018 hepatocyte metabolites but underestimated THJ-2201 oxidative defluorination.

AM-2201 structurally differ from that of JWH-018 in the presence of a fluorine atom in the pentyl chain and has higher binding affinities to CB receptors than JWH-018. Jang *et al.* identified major urinary metabolites of AM-2201 in rat and human urine using commercially available metabolites of JWH-018 and AM-2201 by LC-MS. The results of the study showed that the presence of 6-indole hydroxylated metabolites of each drug and N-4-hydroxy metabolite of AM-2201 was found to contribute to the decisive differences in the metabolic patterns of the two drugs. In addition to the concentration ratio of the N-(5-hydroxypentyl) metabolite to the N-(4-hydroxypentyl) metabolite of JWH-018 may be used as a criterion to differentiate between AM-2201 and JWH-018 abuse [163].

In an *in vitro* study on phase 1 metabolism of N-(1-adamantyl)-1-pentyl-1H-indol-3-carboxamide (APICA) and its fluorinated analog N-(1-adamantyl)-1-(5-fluoropentyl)-1H-indole-3-carboxamide (STS-135) shown that dihydroxylated and trihydroxylated species are preferred metabolites for the detection of APICA abuse whereas monohydroxy metabolite is better in case of STS-135 [164]. The phase 1 and phase 2 metabolism pathway *in vitro* study of STS-135 with the use of human hepatocytes was also performed by Gandhi *et al.*

The *in vitro* half-life of STS-135 was found to be 3.1 ± 0.2 min with intrinsic clearance of 208.8 mLmin⁻¹kg⁻¹ [165].

MAM-2201, a fluorinated JWH-122 analog, was first reported in 2012 as an ingredient in herbal mixtures in Korea. Jang *et al.* performed an *in vitro* study of major MAM-2201 and JWH-122 metabolite using human liver microsomes and compared the results with those of urine specimens from suspected MAM-2201 or JWH-122 user. The studies demonstrated that N-5-hydroxylated JWH-122 metabolite and N-4-hydroxylated JWH-122 metabolite were the primary metabolite of MAM-2201 and JWH-122, respectively. And the author proposed that the relative concentration of the two metabolites could be a key factor to prove MAM-2201 or JWH-122 abuse [166].

In addition to the above studies, identification of metabolites of N-(1-adamantyl)-1-pentyl-1H-indazole-3-carboxamide (AKB-48) [167,167], 5F-AKB-48 [167,169], 1-pentyl-8-quinolinyl ester-1H-indole-3-carboxylic acid (PB-22) [170], 1-(5-fluoropentyl)-8-quinolinyl ester-1H-indole-3-carboxylic acid (5F-PB-22) [170] and RC S-8 [171], AB-CHMINACA [172] were also reported.

2.4.1.2 Analysis of Synthetic Cannabinoids

Validation method for the detection and quantification of JWH-018, JWH-018- N-pentanoic acid and JWH-018N-(5-hydroxypentyl) in blood and urine using solid-phase extraction (SPE) followed by UPLC-MS/MS analysis [LOD (0.08-0.14 ng/mL), and LOQ (0.10-0.21 ng/mL)] was reported [173]. Thaxton *et al.* reported GC-MS analysis of JWH-018 and five regioisomeric 1-naphthoyl substituted 1-n-pentylindoles. The use of a capillary column containing a trifluoropropyl methyl polysiloxane (Rtx-200) stationary phase was found to prove excellent resolution of the six compounds [174]. Scheidweiler and Huestis reported a validated method of simultaneous quantification of 20 synthetic cannabinoids and 21 metabolites, and semi-quantification of 12 alkyl hydroxy metabolites of the synthetic cannabinoids in human urine specimens by LC-MS/MS [175]. A library-based screening procedure for 46 synthetic cannabinoids in serum using LC-ion-trap-MS system was developed by Huppertz *et al.* The search algorithm matches retention time, MS and MS²/MS³ spectra. High-temperature ESI, IonBooster™ was used in the method and proved to enhance the MS intensities of several groups of analytes. The analysts LODs in serum range from 0.1 ng/mL to 0.5 ng/mL [176]. Ma *et al.* demonstrated the use of a miniature mass spectrometer for the analysis of synthetic cannabinoids in trace powders as well as in blood and urine samples. Extraction spray ionization was applied to the discontinuous atmospheric pressure interface (DAPI)-MS system for good sensitivity and stable spray signals. 5 L of blood or urine samples was required for the analysis of five selected synthetic cannabinoids, JWH-018, JWH-081, AM-2201, RCS-4 and XLR-11, the LOQs for the synthetic cannabinoids in blood and urine were determined to be between 10 and 20 ng/mL [177].

2.4.2 Synthetic cathinones

Synthetic cathinones, commonly known as "bath salt" are -ketone amphetamine compounds derived from cathinone, a naturally occurring phenylalkylamine found in leaves of khat. They began to appear first in European recreational drug market in the mid-2000s [178,179,180,181]. According to the American Association of Poison Control Centers (AAPCC), there were 2,697 reported cases of human exposures to bath salts in 2012 and the number of cases dropped to 522 in 2015 and 166 in 2016, through May 31 [182]. Synthetic cathinones are usually consumed by snorting drug powder or oral use. And there was a recently phenomenon of injection of synthetic cathinones [183]. The Drug Enforcement Administration has issued final order to temporarily schedule 10 synthetic cathinones (4-methyl-N-ethylcathinone (4-MEC); 4-methyl- α -pyrrolidinopropiophenone

(4-MePPP); alpha-pyrrolidinopentiophenone (-PVP); 1-(1,3-benzodioxol-5-yl)-2-(methylamino)butan-1-one (butylone); 2-(methylamino)-1-phenylpentan-1-one (pentedrone); 1-(1,3-benzodioxol-5-yl)-2-(methylamino)pentan-1-one (pentylone); 4-fluoro-N-methylcathinone (4-FMC); 3-fluoro-N-methylcathinone (3-FMC); 1-(naphthalen-2-yl)-2-(pyrrolidin-1-yl)pentan-1-one (naphyrone); and alpha-pyrrolidinobutiophenone (-PBP)) into schedule I pursuant to the temporary scheduling provision of the Controlled Substances Act on 28 February 2014 [184].

A simultaneous quantitative method for analysis of 24 synthetic cathinones and 4 metabolites in urine by LC-HRMS was developed by Concheiro *et al.* Two product ions of acceptable abundance and specificity were utilized for identification and quantification in the method. 0.25 mL of sample was required for the analysis with LOQ of 0.5 - 1 g/L and linearity from 0.5 - 1 to 100 g/L. The author also checked for the stability of analytes in urine and found that most synthetic cathinones were stable at 4°C for 72 hours and after 3 freeze-thaw cycles (N = 27), but many (N = 19) were not stable at room temperature for 24 hours except for metabolites of buphedrone, ephedrine, 4-methylephedrine, diethylcathinone, 3',4'-Methylenedioxy-alpha-pyrrolidinopropiophenone (MDPPP), 3',4'-Methylenedioxy-alpha-pyrrolidinobutyrophenone (MDPBP), -PVP, 4-MePPP and MDPV [185]. Concheiro *et al.* also reported a method for the simultaneous quantification of 8 piperazines, 4 designer amphetamines and 28 synthetic cathinones and 4 metabolites in urine by LC-HRMS. The method required 100 L of urine sample subjected to solid phase cation exchange extraction (SOLA SCX). Data were acquired in full scan and data dependent MS² mode. Compounds were quantified by precursor ion exact mass, and confirmed by product ion spectra library matching, taking into account product ions' exact mass and intensities. LODs were 2.5 g/L for all compounds, except 1-(4-Bromo-2,5-dimethoxybenzyl)piperazine (2C-B-BZP), 3,4-Dimethylmethcathinone (3,4-DMMC), 5-(2-Aminopropyl)-2,3-dihydrobenzofuran (5-APDB), 6-(2-aminopropyl)benzofuran (6-APB), Dibenzylpiperazine (DBZP) and methiopropamine, which were 5 g/L [186].

Paul *et al.* reported a method for the analysis of new stimulant designer drugs, such as phenethylamine, amphetamine, cathinone and piperazine derivatives, and common drugs of abuse in urine by using LC-HRMS-Q-TOF. The sample was prepared by using 200 L urine sample with a fast and convenient salting out liquid-liquid extraction procedure. Repetitive measurement of extracted analytes at 30 g/L and 60 g/L showed that all analytes, except for cathinone, were found to be stable within 24 hours at 10°C, whereas cathinone was found only stable up to 5 hours. Freeze/thaw stability at 25 g/L and 50 g/L was verified by a three consecutive freeze/thaw cycle (frozen for 24 hour and thawed for at least 1 hour) and almost all analytes, except for cathinone, showed no significant decrease in their concentration [187].

An analytical method for the determination of 37 new designer drugs, including cathinones, hallucinogenic phenethylamines and piperazines by UPLC-TOF-MS was reported by Pasin *et al.* 100 L whole blood was required for the analysis and the run time was 15 minutes. The LOQs for most analytes were 0.05 mg/L with exceptions such as cathinone, benzedrone, bupropion, 4-Bromo-2,5-dimethoxyphenethylamine (2C-B), 2,5-Dimethoxyphenethylamine (2C-H) and 1-(p-Fluorophenyl)piperazine (pFPP) having LOQs of 0.1 mg/L and the linear range was 0.05 - 2 mg/L for all analytes [188].

Minakata *et al.* used matrix-assisted laser desorption ionization (MALDI)-quadrupole time-of-flight-mass spectrometry (Q-TOF-MS) for the determination of -pyrrolidinopentiothiophenone (PVT), 4'-fluoro-a-pyrrolidinopentiophenone (F-PVP), 4'-methyl-a-pyrrolidinohexanophenone (MPHP), -pyrrolidinoheptanophenone (PV8), -pyrrolidinoctanophenone (PV9), 4'-fluoro-a-pyrrolidinoctanophenone (F-PV9), a-

pyrrolidinopropiophenone (PPP), 4- MePPP, -PBP, and (-PVP) in human blood. The LOD was reported to be 1 ng/mL with quantification range of 2 – 100 ng/mL using 20 L of blood [189, 190].

2.4.3 Substituted Phenethylamines

2.4.3.1 NBOMes

Phenethylamine substances included the 2C family of hallucinogens (2C-B, 2C-I and 2C-C), which are phenethylamines with methoxy group substitutions at positions 2- and 5-; and NBOMes are N-2-methoxy-benzyl substitutes 2C class of hallucinogens, often with a halogen (chlorine, bromine or iodine) at 4- position.

Method for the determination of 4-chloro-2,5-dimethoxyphenethyl-N-[(2-methoxyphenyl)methyl]ethanamine (2CC-NBOMe or 25C-NBOMe) and 2-(4-iodo-2,5-dimethoxyphenyl)-N-[(2-methoxyphenyl)methyl]ethanamine (25I-NBOMe) in human serum using SPE for sample preparation followed by LC-MS/MS with 2-(2,5-dimethoxyphenyl)-N-(2-methoxybenzyl)ethanamine (25H-NBOMe) as the internal standard was developed by Poklis *et al.* The calibration curves were linear over the investigated concentration range, 30 to 2000 pg/mL, with LODs of 10 pg/mL for both compounds. Both 2CC-NBOMe and 25I-NBOMe were considered stable (quality control samples within 20% of the target values) [191]. Johnson *et al.* reported a validated method and the analyte stability of N-(2-methoxybenzyl)-2,5-dimethoxy-4-bromophenethylamine (25B-NBOMe), 25C-NBOMe, 2-(2,5-dimethoxy-4-methylphenyl)-N-(2-methoxybenzyl)ethanamine (25D-NBOMe), 25H-NBOMe, 25I-NBOMe and 2-(2,5-Dimethoxy-4-ethylthiophenyl)-N-[(2-methoxyphenyl)methyl]ethanamine (25T2-NBOMe or 2CT2-NBOMe) via LC-MS/MS in whole blood, plasma and urine with LOQs of analytes ranged from 0.01 to 0.02 ng/mL. Stability experiment of the sample extracts demonstrated that 25I-NBOMe was the most deteriorated compound over the 7-day storage period with an RSD of 39% in the whole blood extract following storage [192]. And the author concluded that the sample extracts should be analyzed following extraction and that storing them for numerous days prior to analysis is not advised. Determination of nine 25-NBOMes, including 25H-NBOMe, 2CC-NBOMe, N-(2-fluorobenzyl)-2-(4-iodo-2,5-dimethoxyphenyl)ethanamine (25I-NBF), 25D-NBOMe, 25B-NBOMe, 2-(2,5-dimethoxy-4-(methylthio)phenyl)-N-(2-methoxybenzyl)ethanamine (2CT-NBOMe), 2-(4-iodo-2,5-dimethoxyphenyl)-N-[(2,3-methylenedioxyphenyl)methyl]ethanamine (25I-NBMD), 2-(2,5-dimethoxy-3,4-dimethylphenyl)-N-(2-methoxybenzyl)ethanamine (25G-NBOMe) and 25I-NBOMe in urine specimen by LC-MS/MS was also reported by the author. The NBOMe derivatives were rapidly extracted from the urine specimens by the use of FAST™ SPE columns with a verified linear range of 1 to 100 ng/mL. The NBOMe derivatives were found to be stable under condition of specimen handling in the laboratory with the exception of 2CT-NBOMe [193]. The author also identified fifteen 25I-NBOMe metabolites in phase I and II mouse hepatic microsomal preparation, and analysis of two human urine samples from 25I-NBOMe-intoxicated patients and found that 5-O-desmethyl-2-I-NBOMe and 2-O-desmethyl-5-I-NBOMe were the best urinary biomarkers for detection of 25I-NBOMe use. As 2-O-desmethyl-5-I-NBOMe has a greater abundance in urine, it is recommended to be the primary biomarker in urine screening for 25I-NBOMe use [194]. Another study on the metabolism of 25I-NBOMe in human and rat urine using GC-MS, LC-MSⁿ, and LC-HRMS/MS was reported by Caspar *et al.* The study showed that 25I-NBOMe was extensively metabolized with O-demethylation, O,O-bis-demethylation, and hydroxylations as predominant pathways and several CYP isoenzymes were involved in formation of the main metabolites [195].

2.4.3.2 Other New Phenethylamines & Benzofurans

Boatto *et al.* developed and validated an LC-MS/MS method for the quantification of four new phenethylamines, 4-bromo-2,5-beta-trimethoxyphenethylamine (BOB), 4-methyl-2,5-beta-trimethoxyphenethylamine (BOD), 3,4-methylenedioxy-beta-methoxyphenethylamine (BOH), and 4-methyl-2,5-dimethoxy-beta-hydroxyphenethylamine (BOHD), in plasma. The plasma samples were cleaned up by SPE with 2,3-dimethoxyphenethylamine- d_3 as internal standard. The LOD and LOQ of this method ranged from 2.23 to 5.95 ng/mL and from 7.4 to 15.5 ng/mL, respectively with tested linear range of 10-500 ng/mL [196].

An investigation of in vitro drug metabolites in human of 7 designer phenethylamines by LC-Q-TOF-MS was performed by Lai *et al.* The phenethylamines included para-methoxyamphetamine (PMA), para-methoxymethamphetamine (PMMA), 4-methylthioamphetamine (4-MTA), and four benzofuran designer drugs, N-methyl-benzodioxolylbutanamine (MBDB), benzodioxolylbutanamine (BDB), 5-(2-aminopropyl) benzofuran (5-APB) and 6-(2-aminopropyl) benzofuran (6-APB). The results showed that the in vitro metabolism pathway of the selected phenethylamines preferentially proceeded via O-dealkylation as the major pathway followed by N-dealkylation, and to a much smaller extent, oxidation of unsubstituted carbon atoms and deamination [197].

Welter *et al.* identified the phase I metabolite of 5-APB and its N-methyl derivative N-methyl-5-(2-aminopropyl)benzofuran (5-MAPB) in acetylated rat urine by GC-MS and/or LC-HR-MSⁿ and identified the phase II metabolites by LC-HR-MSⁿ. The main metabolite of 5-APB was 3-carboxymethyl-4-hydroxy amphetamine and the main metabolites of 5-MAPB were 5-APB (N-demethyl metabolite) and 3-carboxymethyl-4-hydroxy methamphetamine [198]. The plasma concentrations determined in six clinical cases ranged from 5 to 124 µg/L for 5-MAPB and from 1 to 38 µg/L for its N-demethyl metabolite 5-APB. The author also studied the metabolism of 6-APB and its N-methyl derivative N-methyl-6-(2-aminopropyl)benzofuran (6-MAPB) by identifying the metabolites in rat urine and human liver preparations using GC-MS and/or LC-HR-MSⁿ. Besides The main metabolite of 6-APB was found to be 4-carboxymethyl-3-hydroxy amphetamine while main metabolites of 6-MAPB were found to be 6-AOB (N-demethyl metabolite) and 4-carboxymethyl-3-hydroxy methamphetamine [199]. Furthermore, a differentiation of 6-APB and 6-MAPB in urine from their positional isomers 5-APB and 5-MAPB was successfully performed after SPE and heptafluorobutyrylation by GC-MS via their retention times.

2.4.4 Synthetic tryptamines

Tryptamines contain an indole structure joined to an ethyl amine group. Natural tryptamines such as psilocin and psilocybin in “magic mushroom”, bufotenine and its derivative 5-methoxy-*N,N*-dimethyltryptamine (5-MeO-DMT) are hallucinogenic substances [200, 201]. Recently, there are synthetic tryptamines, such as 5-methoxy-*N,N*-Diallyltryptamine (5-MeO-DALT), appear in the recreational drug market [201, 202]. Michely *et al.* identified phase I and phase II metabolites of *N,N*-Diallyltryptamine (DALT) and 5-MeO-DALT in rat urine and in pooled human liver microsomes using LC-HRMS/MS. The result showed that CYP2C19, CYP2D6, and CYP3A4 were mainly involved in the metabolism of DALT and 5-MeO-DALT. The most abundant targets were a ring hydroxy metabolite of DALT, the N,O-bis-dealkyl metabolite of 5-MeO-DALT, and their glucuronides. GC-MS, LC-MSⁿ, and LC-HRMS/MS were then used to detect the abundant metabolites of DALT and 5-MeO-DALT in rat urine and only main metabolites of DALT can be detected by GC-MS [203].

2.4.5 Synthetic Opioids

2.4.5.1 AH-7921

AH-7921 is one of the NPS required risk assessment by the EMCDDA's Scientific Committee. It is a structurally atypical synthetic opioid analgesic invented in the mid-1970s and firstly identified in a sample purchased from an internet retailer in July 2012 [204].

Wohlfarth *et al.* studied the metabolism of AH-7921 by incubating AH-7921 with human liver microsome and analysing by LC-Q-TOF-HRMS/MS. And the data were analyzed with MetabolitePilot™ (SCIEX) using multiple data processing algorithms and the presence of AH-7921 and metabolite was confirmed in the urine case specimen. The author concluded that the desmethyl, di-desmethyl AH-7921 and the glucuronidated metabolites are suitable analytical targets for documenting AH-7921 consumption [205].

2.4.5.2 Lefetamine Derivatives

N-ethyl-1,2-diphenylethylamine (NEDPA) and N-iso-propyl-1,2-diphenylethylamine (NPDPA) were confiscated in Germany in 2008. They are derivatives of lefetamine, an opioid analgesic being synthesized in Japan in the 1940s and became widely abused later. Wink *et al.* studied the phase I and phase metabolism of NEDPA, NPDPA and lefetamine in rat urine using GC-MS, LC-MSⁿ and LC-HRMS/MS [206-207]. Application of a 0.3 mg/kg body weight dose of NEDPA or NPDPA, corresponding to a common lefetamine single dose, revealed that only the metabolites could be detected, namely N-deethyl-NEDPA, N-deethyl-hydroxy-NEDPA, hydroxy-NEDPA, and hydroxy-methoxy-NEDPA or N-de-iso-propyl-NPDPA, N-de-iso-propyl-hydroxy-NPDPA, and hydroxy-NPDPA [206]. Similarly, after a therapeutic lefetamine dose, the bis-nor, bis-nor-hydroxy, nor-hydroxy, nor-di-hydroxy metabolites could be detected [207]. The author also studied the involvement of the ten most important human cytochrome P450 (CYP) isozymes in the N-desalkylation of lefetamine, NEDPA and NPDPA [208].

2.4.5.3 MT-45

MT-45, a synthetic opioid, is one of a series of 1-(1,2-diphenylethyl)piperazine analgesics invented in early 1970s. It was first detected in a powder by Swedish customs in October 2013 and later came one of the compound risk assessment by the EMCDDA's Scientific Committee [209]. Within a nine-month period between November 2013 and July 2014, a total of 28 deaths where the presence of MT-45 in biological samples was analytically confirmed have been reported by Sweden. In 19 of the deaths MT-45 was reported as either the cause of death or contributing to death (even in presence of other substances).

2.4.6 Amphetamine Analogues

2.4.6.1 Camfetamine

Camfetamine (N-Methyl-3-phenyl-norbornan-2-amine), developed and patented as an analeptic by Merck in 1961, is a stimulant drug with effects similar to amphetamine. It was first reported to be used as a recreational drug in May 2011 [210]. The metabolism of camfetamine in rat urine was studied by Maurer *et al.* with the use of GC-MS and LC-HR-MSⁿ and the hydroxyl-aryl camfetamine and the corresponding glucuronide was found to be the most abundant metabolites [211].

2.4.6.2 N-Methylamphetamine

4-methylamphetamine is a synthetic phenethylamine stimulant originally studied as an appetite suppressant in 1950s, but its development was abandoned for unknown reason and was never made commercially available. 4-methylamphetamine was one of the NPS monitored by EMCDDA. According to the risk assessment report of EMCDDA on January 2014, there have been a total of 20 non-fatal cases of acute 4-methylamphetamine toxicity or detection of 4-methylamphetamine in drug-related offences from 5 European countries (Belgium, France, Hungary, Sweden and the United Kingdom), and a total of 21 fatalities from 4 countries (Belgium, Denmark, the Netherlands and the United Kingdom) where 4-methylamphetamine was detected in post-mortem samples reported [212]. The metabolism and detectability of 4-methylamphetamine and its isomers, 2-methylamphetamine and 3-

methylamphetamin, were studied by Maurer *et al.* in rat urine using GC-MS and LC-HR-MSⁿ [213].

2.4.6.3 2-Methiopropamine

1-(thiophen-2-yl)-2-methylaminopropane (2-methiopropamine) is an analogue of MA. It was first synthesized in 1942 and first detected as a recreation drug in Europe in January 2011. An acute toxicity case associated with analytically confirmed recreational use of methiopropamine was reported [214]. Toxicological analysis of the urine of the patient detected methiopropamine at a concentration of 400 ng/mL. The metabolism of amphetamine (CFA), three methylphenyl-amphetamines (2-MA, 3-MA, and 4-MA), 2-methiopropamine (2-MPA), and 5-(2-aminopropyl)benzofuran (5-APB), 6-(2-aminopropyl)benzofuran (6-APB, so-called 'benzofury') as well as their N-methyl derivatives 5-MAPB and 6-MAPB, as well its chemistry, pharmacology, toxicology and toxicokinetics was discussed by Welter *et al.* With the exception of 5- and 6-APB, these NPS were extensively metabolized by N-demethylation and/or hydroxylation [215]. For urinalysis, the unchanged drugs and/or the nor metabolites are the main targets.

2.4.7 Ketamine Derivative: Methoxetamine

Methoxetamine is a 3-methoxy, N-ethyl derivative of ketamine. It was first identified in the UK in November 2010 [216] and was one of the drugs monitored by EMCDDA. According to the risk assessment report dated May 2014 of EMCDDA, a total of 120 non-fatal intoxication and twenty deaths associated with methoxetamine have been reported [217]. Mever *et al.* studied the metabolism and detectability of methoxetamine in rat and human urine by GC-MS and LC-HR-MSⁿ and concluded that metabolic pathways of 4-MEC could be proposed: reduction of the keto group, N-deethylation, hydroxylation of the 4-methyl group followed by further oxidation to the corresponding 4-carboxy metabolite, and combinations of these steps. [218].

2.4.8 Designer Benzodiazepines

Designer benzodiazepines such as diclazepam, flubromazepam, pyrazolam, clonazolam, which have not been approved for medicinal use in any country, started to appear in the market online in recent years [219]. Immunochemical tests applied in clinical settings and drug rehabilitation detect most of the designer benzodiazepines with sufficient sensitivity. However, the mass spectrometric methods needed for confirmation do not regularly cover the latest designer benzodiazepines, due to lack of reference materials. Pyrazolam, is a new benzodiazepine found to be sold in online shop as research chemical in 2012 [220]. Moosmann *et al.* has reported the characterization, metabolism and pharmacokinetics of pyrazolam, flubromazepam and diclazepam [220-222]. The metabolism and pharmacokinetics of the designer benzodiazepines were studied by analysis serum and urine sample of volunteer who has taken the drug. For pyrazolam with 1 mg dose intake by one of the authors, the elimination half-life was found to be about 17 hours and the parent compound was detected with the described LC-MS/MS method in serum for more than 50 hours and in urine for approximately 6 days [220]. For flubromazepam with 4 mg dose intake, the approximate elimination half-life was found to be 106 hours and the parent drug could be detected by LC-MS/MS for 23 days in serum [221]. For diclazepam with 1 mg intake, the approximate elimination half life was 42 hours with metabolites delorazepam, lorazepam and lormetazepam which can be detected in urine by LC-MS/MS for 6, 19, and 11 days, respectively. In serum, the consumption could be proven between 99 hours post-intake targeting the parent compound and up to 10 days targeting the metabolite delorazepam. [222]. Huppertz *et al.* also reported the characterization of clonazolam, deschloroetizolam, flubromazolam and meclonazepam and identification of their in vitro metabolite produced by human liver microsomes through Q-TOF analysis [223].

2.4.9 Immunoassay Cross Reactivity of NPS

2.4.9.1 Synthetic Cannabinoids

Immunoassays could provide an inexpensive, sensitive and rapid screening for drugs in biological samples. A study on (-)-(-1)-hydroxy, carboxy and 6-hydroxyindole metabolite of JWH-018 and JWH-073 in urine samples showed that synthetic cannabinoid ELISA kit with LC-MS/MS for confirmation found no false negative results among a total of fifty-two urine sample tested [224]. Huestis and co-workers evaluated several immunoassay kits for the detection of synthetic cannabinoids based on testing of authentic urine samples and a validated LC-MS/MS method for 29 synthetic cannabinoids and metabolites [225,226,227]. The evaluation based on analysis of 2443 authentic urine samples using immunoassay HEIA K2 Spice kit which targeted JWH-018 N-pentanoic acid metabolite in urine demonstrated good performance at 5 and 10 µg/L cutoffs as compared to the LC-MS/MS method. JWH-073 N-butanoic acid had the highest cross-reactivity of the 74 compounds evaluated, and three synthetic cannabinoid metabolites, JWH-073 N-(butanoic acid), JWH-073 N-(4-hydroxybutyl) and JWH-073 N-(3-hydroxybutyl) were the most cross-reactive. No interference found from 102 investigated compounds except a mixture containing 1000 g/L each of buprenorphine/ norbuprenorphine produced a positive result above the author proposed cutoff (5 g/L) but below the manufacturer's recommended cutoff concentration (10 g/L). The lowest cutoff (5 µg/L) exhibited the highest sensitivity (92.2%) and efficiency (97.4%) while the highest (20 µg/L) cutoff gave the greatest specificity (99.7%) [225]. Another evaluation study using Neogen SPICE ELISA kit targeting JWH-018 N-pentanoic acid in urine was also reported. 2469 authentic urine samples were analyzed by the Neogen immunoassay and LC-MS/MS. Sensitivity, specificity, and efficiency results with the 5µg/L cut-off were 79.9%, 99.7%, and 97.4% and with the 10µg/L cut-off 69.3%, 99.8%, and 96.3%, respectively. And cross-reactivity was shown for 18 of 73 synthetic cannabinoids markers evaluated [226]. The performance of NMS ELISA kit targeting JWH-018 N-(5-hydroxypentyl) metabolite was also carried out [227]. Results of 2492 urine samples analyzed by the ELISA and the LC-MS/MS method showed sensitivity, specificity, and efficiency results of 83.7%, 99.4% and 97.6% and 71.6%, 99.7% and 96.4%, with the 5 and 10 µg/L urine cutoffs, respectively. No interferences were present from 93 common drugs of abuse, metabolites, co-administered drugs, over-the-counter medications or structurally similar compounds, and 19 of 73 individual synthetic cannabinoids (26%), exhibited moderate to high cross-reactivity to JWH-018 N-(5-hydroxypentyl) metabolite. Synthetic cannabinoids including JWH-200, JWH-073 N-(3-hydroxybutyl) metabolite, JWH-073 N-(4-hydroxybutyl) metabolite, JWH-019 N-(6-hydroxyhexyl) metabolite and AM-2201 N-(4-hydroxypentyl) metabolite, showed significant cross-reactivity [227]. Comparison of the same Neogen immunoassay kit with LC-QTOF-MS was conducted by Kronstrand *et al* [228]. It was concluded that there was fairly high cross-reactivity with MAM-2201 and JWH-122 metabolites for the immunoassay, but there was no cross-reactivity with the UR-144 metabolites at all [228]. A study of the detection of UR-144 and XLR-11 in human urine using ELISA kit targeting pentanoic acid metabolite of UR-144 was conducted by Mohr *et al*. The assay has significant cross-reactivity (% cross-reactivity 50% or higher) with UR-144-5-OH, UR-144-4-OH and XLR-11-4-OH metabolites, but <10% cross-reactivity with the parent compounds, and no measurable cross-reactivity with other synthetic cannabinoids and their metabolites at concentrations of <1,000 ng/mL. Analysis of 90 positive and negative control urine samples for UR-144, XLR-11 and its metabolites tested versus LC-MS/MS revealed accuracy, sensitivity and specificity to be 100% at the assay's cutoff of 5-ng/mL relative to the pentanoic acid metabolite of UR-144 [229].

2.4.9.2 Amphetamine Analogs

Cross-reactivities of 39 new amphetamine designer drugs on three abuse drug urinary screening tests (Screen[®]7, SureStep[™], InstAlert[™]) were studied by Nieddu *et al*. It showed that only 9 of the 39 amphetamine designer drugs tested could be detected by one

of the immunoassay tests for detection of amphetamine, MA and MDMA [230]. An evaluation study on the cross-reactivity of five commercially available immunoassay reagent kits for 94 designer drugs on a Roche/ Hitachi Modular P automated screening instrument was performed. The drug concentration used to determine cross-reactivity for each assay was 100 g/mL. It was found that 80 of the designer drugs gave positive result on at least one of the five commercial immunoassays evaluated with the positive rate of 19% for Microgenics DRI® Ecstasy enzyme assay, 20% for Microgenics DRI® Phencyclidine enzyme assay, 39% for Lin-Zhi Methamphetamine enzyme immunoassay, 43% for Siemens/Syva EMIT®II Plus Amphetamines assay and 57% for CEDIA® DAU Amphetamine/Ecstasy assay [231]. Fourteen designer drugs generated a negative result on all five assays. Of the compounds that screened negative on all kits evaluated, most had either large structural substituents connected to the amine nitrogen or groups added to the aromatic ring.

Swortwood *et al.* also evaluated cross-reactivity of 30 designer drugs, including 24 phenylethylamines (with 8 cathinone derivatives), 3 piperazines and 3 tryptamines, with 16 different ELISA reagents. Cross-reactivity towards most drugs was <4% in assays targeting amphetamine or MA [232]. Compounds such as MDA, MDMA, ethylamphetamine, and α -methyltryptamine demonstrated cross-reactivities in the range of 30-250%. When tested against the Randox Mephedrone/Methcathinone kit, cathinone derivatives demonstrated cross-reactivity at concentrations as low as 150 ng/mL.

Krasowski *et al.* have applied two-dimensional (2D) molecular similarity analysis to designer amphetamine-type stimulants and synthetic cannabinoids, the author proposed that the similarity calculations can be used to more efficiently decide which drugs and metabolites should be tested in cross-reactivity studies, as well as to design experiments and potentially predict antigens that would lead to immunoassays with cross-reactivity for a broader array of designer drugs [233].

Holler *et al.* studied the interference of methiopropamine, 4-fluoramphetamine, 4-fluoromethamphetamine and 4-methylamphetamine with immunoassays and GC-MS confirmation analysis utilizing three derivatization procedures, R(-)- α -methoxy- α -trifluoromethylphenylacetyl chloride (R-MTPAC), heptafluorobutyric anhydride (HFBA) and chlorodifluoroacetic anhydride (ClF2AA). Significant cross-reactivity was observed with all the four compounds on the Syva Emit II Plus Amphetamines and Roche KIMS Amphetamines II immunoassays. 4-MA spiked at 250 ng/mL produced a positive response for the Roche AMP kit as did 4-FMA at 750 ng/mL. Methiopropamine found to show significant cross-reactivity in immunoassay, yielding positive response at 900 and 4,000 ng/mL for Syva and Roche AMP (amphetamine kits), respectively [234].

2.4.9.3 Designer Benzodiazepines

The cross-reactivity of phenazepam, etizolam, pyrazolam, flubromazepam, diclazepam and its metabolite delorazepam with the Immunalysis®Benzodiazepines ELISA kit were studied by O'Connor *et al.* The result showed that the benzodiazepines are possible to be detected with the ELISA kit with cross-reactivity found to be 90, 107, 86, 84, 79 and 80% for phenazepam, etizolam, pyrazolam, flubromazepam, diclazepam and delorazepam, respectively when calibrators were prepared in blank blood [235]. Moosmann *et al.* found high cross reactivity of flubromazepam (4 mg intake) in urine samples using AxSYM® 4602 (FPIA) instrument and cobas® 8000 instrument (turbidity assay) but the used assay seemed not to be sufficient for safe detection of the applied dose [221]. Only one of the immunochemical assay was capable of detecting intake of 1 mg diclazepam [222] and pyrazolam (1 mg intake) was not be able to be detected in most of the tested samples [220].

2.5 Case report related to New Psychoactive Substances

2.5.1 Synthetic cannabinoids

Hasegawa *et al.* reported a death case with 5-fluoro-ADB and MAB-CHMNACA were detected in the post-mortem specimens, while 5-fluoro-ADB-PINACA was detected only in the stomach contents [236-237]. They also reported another case of which AB-CHMINACA, 5-fluoro-AMB and diphenidine were detected in the post-mortem specimens of a 30-year-old male deceased [238]. Bechonick *et al.* reported four fatal cases with 5F-PB-22 detected in concentration range of 1.1 – 1.5 ng/mL in the post-mortem blood (PM Blood) [239]. A fatal case of NNEI (an analog of the synthetic cannabinoid JWH-018) poisoning is reported by Sasaki *et al.* [240]. The concentrations of the aforesaid synthetic cannabinoids in blood were summarized below:

Drug	Concentration in blood (ng/mL)	Decedent Age/Sex	Ref.
5-fluoro-ADB	--a	34/M	[236]
MAB-CHMNACA	6.05 - 10.6 ^a		[237]
5-fluoro-ADB-PINACA	--b		
AB-CHMINACA	Below LOQ ^a	30/M	[238]
Diphenidine	715 ^a		
AB-CHMINACA	--c		
5F-PB-22	1.1 ^d	17/M	[239]
5F-PB-22		1.5 18/M	[239]
5F-PB-22		1.5 19/M	[239]
5F-PB-22	1.3 ^{d, e}	27/M	[239]
NNEI	0.64-0.99 ^a	~20/M	[240]

Remarks: ^a detected in other tissues; ^b detected in stomach only; ^c detected in adipose tissue only; ^d other drugs also detected; ^e the sample tested was serum taken about 7 hours before death

For the cases with other tissues examined [236-238, 240], quantitation of all compounds were achieved in adipose tissues, and some of the drugs (5-fluoro-ADB, 5-fluoro-AMB, NNEI) in adipose tissue were comparatively higher than other specimens; the high lipophilicity typical of synthetic cannabinoids was considered as the contributing factor. This suggested the adipose tissue as a choice of specimen when synthetic cannabinoid(s) involvement is suspected [238, 240].

In the case of NNEI, the deceased's hair was divided into segments and analysed for the level of NNEI. Considering the detection of NNEI in the segment of hair representing a period before NNEI available in Japan, the author suggested that the NNEI in the washed hair could be due to external contamination by the smoke of the herbal blend containing NNEI [240].

2.5.2 Synthetic Cathinones

The synthesis of mephedrone was first described by Saem de Burnaga Sanchez in 1929 [241]. In a 3-year review of NPS in casework published by Elliott and Evans indicating mephedrone is the most frequent NPS appear in their casework between January 2010 to December 2012 [242], although the world drug report 2015 of the UNODC showed that from the annual prevalence data, there is a decrease in consumption of mephedrone from year 2010/11 to 2012/13 in the United Kingdom [153].

3,4-DMMC (3,4-dimethylmethcathinone) first appeared in Hungary in 2010. Usui *et al.* reported a quantitative analysis of 3,4-DMMC in blood and urine in a fatal case using LC-MS/MS [243].

Methylone (3,4-methylenedioxy-methcathinone) is a synthetic derivative of cathinone. It was first synthesized by Jacob and Shulgin in 1996 as an antidepressant and antiparkinsonian drug. Two fatal cases associated with methylone were reported by Barrios *et al.* [244] and McIntyre *et al.* [245].

Ethylone (3,4-Methylenedioxy-N-ethylcathinone, also MDEC, bk-MDEA) is a newer N-ethyl form of methylone. The DEA National Forensic Laboratory Information System (NFLIS) first informed ethylone positive case in second half of 2011 [246]. Two studies on ethylone related fatal cases were reported by Lee *et al.* [246] and McIntyre *et al.* [247]. The concentrations of the aforementioned synthetic cathinones in blood are summarized in the table below:

Synthetic Cathinone	Concentration (ng/mL)	Decedent Age/Sex, cause of death or case history	Ref.
3,4-DMMC	27000*	~30/M, syringe & packet marked "LOOP3" found nearby	[243]
Methylone	3100 ^{a,*}	21/M, respiratory distress	[244]
	3400 ^a	19/F, drowning	[245]
Ethylone	<25 ^{b,*}	23/M, gunshot wound of chest	[246]
	2,572 ^{c,*}	29/M, multiple gunshot wounds	[246]
	--*	25/M, alprazolam, cocaine & heroin intoxication	[246]
	1,837	18/M, gunshot wound of chest	[246]
	38 ^{e,*}	27/M, undetermined	[246]
	137 ^{c,*}	24/M, blunt impact; contributing ethylone & ethanol intoxication	[246]
	1,617*	24/M, hanging	[246]

Remarks: * ethylone found in urine; ^a THC was also detected; ^b alprazolam detected; ^c ethanol detected; ^d alprazolam, benzoyllecoginine, morphine detected in blood; ^e diphenhydramine, tramadol detected; ^f alprazolam, morphine, THC detected

2.5.3 Pyrovalerone Analogs

MDPV (3,4-methylenedioxy-pyrovalerone) was first identified in a seizure in November 2008. A total of 525 non-fatal intoxications and 108 fatal cases associated with MDPV were

reported to the EU Early Warning System according to the EMCDDA risk assessment report [248].

-PVP (-Pyrrolidinovalerophenone) is sympathomimetic drugs related to cathinone first described in a patent literature in 1963. 106 deaths associated with the use of -PVP were reported by seven member states according to the EMCDDA Europol Joint Report [249]. Post-mortem distribution of -PVP and its metabolite, OH--PVP, in body fluids and solid tissues in a fatal poisoning case was determined by LC-MS/MS and reported by Hasegawa *et al.* [250]. Another fatal case involving -PVP and 2-(methylamino)-1-phenylpentan-1-one (pentedrone) was reported by Sykutera *et al.* [251]. -PBP (-pyrrolidinobutiophenone) was detected by Wurita *et al.* in an autopsy case, in which the direct cause of death was judged to be subarachnoid hemorrhage. Caffeine, acetaminophen, and allylisopropylacetylurea were also detected in the blood [252]. An autopsy case related to 4-MeOPBP (4'-methoxy--pyrrolidinobutiophenone) was first reported by Shintani-Ishida *et al.* The plasma and urine sample of the deceased were analysis using LC-TOF-MS with MSE [253]. Hasegawa *et al.* reported the post-mortem distribution of PV9 (1-phenyl-2-(pyrrolidin-1-yl)octan-1-one) in solid tissue collected from a 18-year-old female [254]. Kudo *et al.* reported a fatal case involving PV9, 4-methoxy PV9 and 1-(4-methoxyphenyl)-2-(pyrrolidin-1-yl)heptan-1-one (4-methoxy PV8) together with diphenidine, nitrazepam, 7-aminonitrazepam, flunitrazepam, 7-aminoflunitrazepam, triazolam and -hydroxytriazolam [255]. The findings of the synthetic cathinones in blood, urine, brain, liver ad kidney are summarized in the table below:

Drug	Concentration (ng/mL or ng/g)					Decedent Age/Sex, [Ref]
	Blood	Urine	Brain	Liver	Kidney	
α -PVP	654	11,200	518	681	1,580	41/M, [250]
OH- α -PVP	364	5,300	592	1,080	972	
α -PVP	901	--	120	2,601	462	28/M, [251]
OH- α -PVP	185	--	91	2,264	94	
Pentedrone	8794	--	13,248	100,044	22,102	
α -PBP	55.2	906	41.4 ^a	82.6	140	21/M, [252]
4-MeOPBP	9500 ^b	12,000	--	--	--	35/M, [253]
PV9	180	20 ^c	212	377	907	18/F, [254]
PV9	743	1,340	--	--	--	[255]
4-methoxy PV9	261	360	--	--	--	
4-methoxy PV8	2,690	3,870	--	--	--	

Remarks: ^a Cerebrospinal fluid was used; ^b Plasma was used; ^c Antemortem urine

2.5.4 Synthetic -opioid Agonist: AH-7921

AH-7921 (3,4-dichloro-N-[(1-dimethylamino)cyclohexylmethyl] benzamide) was a compound being developed in 1970s and it is one of the NPS being monitored by EMCDDA. A total of six non-fatal intoxication and 15 death associated with AH-7921 were reported by Sweden, the United Kingdom and Norway in the EMCDDA Europol Joint Report [204]. AH7921 blood

levels from a total of 12 death cases reported by Karinen *et al.* [256], Kronstrand *et al.* [257] and Vorce *et al.* [258] were summarized in the table below. In the case reported by Vorce *et al.* [258], the concentration distribution of AH-7921 in the post-mortem specimens was also studied.

Decedent Age/Sex, [Ref]	concentration in blood (ug/mL or g/g)	
	AH-7921	Other drugs detected
~20/M, [256]	0.43 ^a	2-FMA (0.0069), 3-MMC (0.0021), Codeine (0.42), Codeine-6-glucuronide (0.77), acetaminophen (19)
young/F, [256]	0.33 ^a	Methoxetamine (0.64), phenazepam (1.33), Etizolam (0.27), diazepam (0.046), nordiazepam (0.073), oxazepam (0.018), 7-aminonitrazepam (0.43)
27/M, [257],	0.81 ^b	Gabapentin (10)
26/M, [257]	0.99 ^b	Amphetamine (4.7), Aripiprazole (0.16), Dehydroaripirazole (0.06)
24/M, [257]	0.03 ^c	Ethanol (0.11 g/L), Paroxetine (0.03)
45/M, [257]	0.20 ^b	Alimemazine (0.08), Codeine (0.12), Diazepam (0.03), Nordazepam (0.09), Tramadol (0.64), O-Desmethyltramadol (0.04), Paracetamol (5), Pyrazolam (positive)
26/M, [257]	0.30 ^b	Alprazolam (0.003), Hydroxyzine (0.09), Zopiclone (0.02), Promethazine (0.1), Desmethylpromethazine (0.1), Paracetamol (4), Pyrazolam (positive)
34/M, [257]	0.08 ^b	Ethanol (0.17 g/L), N-Ethyl-norketamine (0.01)
27/M, [257]	0.16 ^b	Amphetamine (0.04)
25/M, [257]	0.35 ^b	3-Methylmethcathinone (positive)
22/M, [257]	0.43 ^b	Bupropion (0.40), Hydroxybupropion (0.53), diazepam (0.12), Nordazepam (0.17), Pregabalin (12), Mirtazapine (0.10), Desmethylmirtazapine (0.02)
19/M, [258]	9.1 ^a	--

Remarks: ^a Peripheral blood; ^b femoral blood; ^c heart blood

2.5.5 Synthetic Amphetamine Analogues

2.5.5.1 Methiopropamine

After being first detected in 2011 in Finland, several methiopropamine intoxication and fatal cases were reported. The WHO Expert Committee on Drug Dependence Thirty-sixth Meeting methiopropamine critical review report mentioned 21 cases of methiopropamine intoxication (15 cases detected in urine and 5 cases detected in blood) reported in Sweden during 2013 [259]. The review report also mentioned three fatal cases involving methiopropamine, two of them occurred in January 2012 in the United Kingdom and one of

that reported in Sweden with methiopropamine concentration determined to be 1.3 g/g in femoral blood [259]. Lee *et al.* reported an acute toxicity case associated with methiopropamine. Methiopropamine was detected in the urine of a 27-year-old woman at a concentration of 400 ng/mL, other drugs detected in the urine including morphine (100 ng/mL), JWH-018 and JWH-019 [214]. Another methiopropamine related fatal case in Australia was reported by Anne *et al.* Methiopropamine was in the peripheral blood of the 29-year-old deceased male at a concentration of 38 mg/L 3 days after death [260].

2.5.5.2 2,5-Dimethoxy-4-Chloroamphetamine (DOC)

DOC is a substituted alpha-methylated phenethylamine. It was first synthesized by Alexander Shulgin in 1991. A death case attributed to DOC alone was reported by Barnett *et al.* The decedent was a 37-year-old male known to be a MA abuser. DOC concentration level in iliac blood, urine, liver and brain were found to be 377 ng/mL, 3,193 ng/mL, 3,143 ng/g and 683 ng/g, respectively [261]. A non-fatal case of a patient who took a street-bought hallucinogenic drug thought to be LSD was found to be related to DOC as detected by LC-HRMS was reported by Burish *et al.* [262].

2.5.6 Ketamine Derivative: Methoxetamine

Methoxetamine (MXE), a ketamine derivative, was first identified in September 2010 in UK. A total of 120 non-fatal intoxication and 20 death associated with MXE have been reported to the Early Warning System according to the EMCDDA risk assessment report [263]. Several literature reporting fatal cases involving MXE in recent years are summarized in the table below:

Decedent Age/Sex	Concentration in blood (µg/mL)		Cause of death, [Ref]
	MXE	Other findings	
29/M	5.8	--	--, [264]
31/M	0.32	Amphetamine	--, [265]
29/M	Detected	Methadone, EDDP, mirtazapine	Drug overdose, [266]
25/M	Detected	Alcohol, dihydrocodeine	Drowning, MXE ingestion, [266]
17/M	Detected	Alcohol	Drowning, MXE ingestion, [266]
43/M	0.89	Methiopropamine, 6-APB	Methiopropamine & MXE toxicity, [266]
20/M	0.22	--	Drowning, [266]
27/F	Detected	6-APB	6-APB & MXE ingestion, [266]
41/M	--*	Methiopropamine, MDA, alcohol	Ischaemic heart disease, [266]
27/M,	0.03	Amitriptyline, benzoylecgonine, diazepam, MDMA, MDA, THC	Mixed drug toxicity, [266]

* Detected in urine only

2.5.7 Tryptamine derivatives and related

2.5.7.1 5-(2-Aminopropyl)indole (5-IT)

5-IT is a synthetic derivative of indole and is a positional isomer of alpha-methyltryptamine. Its synthesis was first described by Hofmann and Troxler in 1963 and started to appear in the European drug market in late 2011 [267]. 24 fatal cases associated with 5-IT was mentioned in the EMCDDA report on the risk assessment of 5-IT [267]. 15 of which were occurred in Sweden and reported by Kronstrand *et al.* [268]. Seetohul and Pounder also reported four fatalities involving 5-IT. The table below shows findings in these cases [268, 269]:

Decedent	Conc. in femoral blood (ug/g or mg/L)	Cause of death, [Ref.]	
Age/Sex	5-IT	Other drugs detected	
23/M	18.6	AM2201, 4-APB (present in urine)	5-IT, 4-APB intoxication, [268]
31/M	2.3	Hydroxyzine, Etizolam	5-IT, etizolam intoxication, [268]
24/M	2.4	Zopiclone, Ethylphenidate, Ritalinic acid	Undetermined, [268]
28/M	3.8	Levitiracetam	Epilepsy, [268]
20/M	1.1	Benzoylcegonine, THC, Pentedone	Sudden cardiac death + drug intake, [268]
31/M	5.2	7-Aminoclonazepam, Perphenazine, Ethylphenidate, Ritalinic acid, Methylphenidate	5-IT, ethylphenidate intoxication, [268]
33/M	4.2	--	Probable 5-IT intoxication, [268]
31/M	1.6	Alimemazine, Desmethyralimemazine, Ethylphenidate, Ritalinic acid	5-IT, ethylphenidate intoxication, [268]
29/M	0.7	Pregabalin (pleural effusions)	5-IT, pregabalin intoxication, [268]
28/M	2.5	Carisoprodol, Meprobamate, 7-Aminoclonazepam	5-IT intoxication, [268]
55/M	2.1	Sertraline, Venlafaxine,	5-IT intoxication, [268]
30/M	2.1	Methylphenidate, Ritalinic acid	Probable 5-IT intoxication, [268]
40/M	1.0	--	5-IT intoxication, [268]
24/M	1.1	Benzoylcegonine, MDMA, MDA	5-IT, MDA, MDMA & cocaine intoxication, [268]
28/M	1.7	Ethylphenidate	Undetermined, [268]
25/M	0.8	MDMA	--, [269]
25/F	0.9	6-APB	--, [269]
22/M	0.4	6-APB	--, [269]
25/F	0.3	MDMA, MDA, Amphetamine, 4-methyl-N-ethylcathionine	--, [269]

2.5.7.2 4-MeO-PCP & 4-HO-MET

McIntyre *et al.* reported a fatal case related to 4-MeO-PCP (4-Methoxyphencyclidine) and 4-HO-MET (4-Hydroxy-N-methyl-N-ethyltryptamine). The decedent is a 54-year-old man. The 4-MeO-PCP concentrations in peripheral blood, central blood, liver, vitreous, urine and gastric contents were determined to be 8.2 mg/L, 14 mg/L, 120 mg/kg, 5.1 mg/L, 140 mg/L and 280 mg, respectively. The author suggested a 'moderate' propensity for 4-MeO-PCP post-mortem redistribution. 4-HO-MET was identified in the peripheral blood but not quantified. In addition, venlafaxine (0.51 mg/L), olanzapine (0.42 mg/L), lorazepam (<0.05 mg/L) and hydroxyzine were also detected in the peripheral blood [270].

2.5.7.3 NBOMe

Thirty-two non-fatal intoxications and four fatal cases associated with 25I-NBOMe were reported in the EMCDDA report on risk assessment of 25I-NBOMe [271]. Several fatal reported cases associated with the NBOMe series were summarized below:

Decedent Age/Sex	[Ref.]	Concentration in blood (ng/mL or ng/g)			
		25I-NBOMe	25B-NBOMe	25C-BOMe	25HBOMe
23/F,	[272]	~28	--	~0.7	~1 a, *
15/M,	[273]		--	--	-- b, *
22/M,	[273]	--	--	0.81	-- b, *
		--	--	0.6	-- a *
18/M,	[274]	--	1.59	--	-- c, *
16/M,	[274]	19.8	--	--	-- c, *
21/M,	[275]	Detected	--	--	-- c, *
15/F,	[275]	Detected	--	--	-- c, *
19/M,	[276]	0.405	--	--	-- d, *
~20/M,	[277]	--	0.177	0.018	-- e, *
		--	0.36	0.039	-- f, *

Remarks: * Other drugs also detected; ^a post-mortem blood; ^b ante-mortem blood; ^c post-mortem heart blood; ^d a peripheral blood; ^e Right heart cavity post-mortem blood; ^f Left heart cavity post-mortem blood

2.6 Accreditation and Quality Assurance

2.6.1 Accreditation

The Subcommittee on Forensic Science (SoFS) of the US has released the report "Strengthening the Forensic Sciences" which represents the first set of research findings and conclusions relating to laboratory accreditation, certification of forensic science and medicolegal personnel, proficiency testing, and ethics [278]. The report mentioned that several states had passed legislation mandating accreditation and other forms of oversight of forensic science service providers, but relevant legislation and other oversight requirements varied greatly from state to state and among local jurisdictions. As a result, there are still a significant portion of Federal laboratories and publically funded forensic

crime laboratories unaccredited. The SoFS strongly recommended all forensic science service providers, as well as medical examiner/coroner offices, forensic units, and part-time and private forensic science entities, with the goal that these entities become accredited under appropriate International Organization for Standardization (ISO)-based standards and any supplemental requirements or standards specific to forensic science.

The Scientific Working Group for Forensic Toxicology (SWGTOX) of the National Institute of Standards and Technology (NIST) has published the Revision 1 of "Standard on the Accreditation of Forensic Toxicology Laboratories" [279]. The standard is intended to reflect a minimum standard of practice and to provide specific requirements applicable to the accreditation of forensic toxicology laboratories. It also provides direction to all accreditation bodies working in this field. All sub-disciplines of forensic toxicology provided by the laboratory that are within the scope of SWGTOX must be included in the accreditation. Each sub-discipline has unique characteristics that must be assessed. These sub-disciplines include human performance toxicology (e.g. DFC, DUI or DUID), postmortem toxicology, non-regulated employment drug testing, court ordered toxicology and general forensic toxicology for other toxicology performed for a legal purpose in a variety of biological specimens.

2.6.2 Method Validation

The minimum standards of practice for validating analytical methods in forensic toxicology had been updated and published by the SWGTOX [280]. The changes included the replacement of "accuracy" with "bias" and introduction of "working range" in definition; addition of statement of conducting validation in a manner similar to casework; providing instruction on preparation of fortified matrix samples used in validation; removal of "optional parameters" (recovery and roustness) from the document; addition of requirement of management review and approval of validation etc.

Several essential aspects of method validation in bioanalysis were reviewed [281]. The authors considered that the most controversial parameters (LOQ, robustness and matrix effect) were studied and the definitions and methodology proposed by the different regulatory bodies were compared. The authors found that robustness assay is not yet mandatory according regulatory bodies, and that experimental design emerges to be a very powerful tool for this aim. The Accuracy Profile approach seemed to be the way to estimate LOQ, intergrating trueness, precision, risk and linearity; and that the methodology proposed by Matuszeski *et al.* appears to be adequate to determine the presence or absence of matrix effect.

The available validation strategies for small molecules metabolites present in biological samples had been reviewed by Naz *et al.* [282]. Although different approaches have been made by some research groups in order to define the validation parameters with some accepted ranges after following established methodologies, validation strategies for non-targeted approach are not well defined at present. The authors have discussed in details on the available validation strategies that are being used and some steps are recommended to consider during a non-targeted metabolomics method development.

During method validation, many experiments are required and the steps are highly demanding in time and consumables. In order to avoid the difficult task of performing too many experiments, the Youden test which makes use of fractional factorial designs, has been proved to be a very effective approach. The main advantage of Youden test is the fact that it keeps the required time and effort to a minimum as only a limited number of determinations have to be made by using combinations of the chosen investigated factors.

Eftichia *et al.* has briefly discussed the typical applications of this robustness test found in literature covering a wide variety of sample matrices including biofluids [283].

2.6.3 Uncertainty of Measurement

The American Society of Crime Laboratory Directors/Laboratory Accreditation Board (ASCLD/LAB) has published the guidance to laboratories that must achieve compliance with the ASCLD/LAB Policy on measurement uncertainty as they prepare for and maintain ASCLD/LAB-International accreditation [284]. The guidance document covers the introduction to measurement uncertainty and a general process for the estimation of measurement certainty.

The importance of the uncertainty of measurement in the method validation is described by Zilli [285]. The author provides the analyst with an easy to use step-by-step guide to calculate the uncertainty of measurement in implementing a new analytical method. One of the methods is the bottom-up approach which considers all the possible sources of variability and then to sum up all of them in the final calculation. The other method is the top-down approach which makes use of a collaborative trial and processing the resulting statistics. Both methods imply different advantages and drawbacks. The analyst should choose the one fits best to his requirements.

During the method validation of the analysis of caffeine and its major metabolites in human plasma samples for clinical study related to caffeine in sports medicine, relevant estimates of combined standard uncertainty are computed to obtain uncertainty functions, which allow to determine the measurement certainty as a function of the concentration of the analyte [286]. The great advantage of both uncertainty function and uncertainty profile is the development of a continuous model that enables easy calculation of the standard, expanded and relative expanded uncertainty at any concentration along the validation range.

The uncertainty of measurement has been evaluated for the quantification of THC-COOH and its glucuronide conjugate in urine samples using LC-MS/MS [287]. The sources of uncertainty are identified and classified into four main contributions including standard preparation, calibration curve, method precision and bias. The bottom-up approach was used to estimate of the uncertainty of measurement. The main contribution to the overall uncertainty is due to extraction procedure and calibration curve construction.

3. Advances in Toxicological Analysis

3.1 Development of High Resolution LC-MS Techniques

Quantitative analysis of drugs in biological samples using LC-MS/MS is predominately performed in current years. These LC-MS/MS assays use selective reaction monitoring (SRM) mode in multi-analyte quantitation. SRM employs selective monitoring of precursor-product ion transitions, and multiple transitions are now used routinely in practice to improve specificity of analysis. Although SRM methods are sensitive and specific, their main drawback is that other analytes not included in the method are undetected, even if present at high concentration. Moreover, a retrospective analysis of the sample cannot be conducted for the post-target analysis of other relevant analytes not included in the initial scope.

Recently, non-targeted drug screening can be achieved using high-resolution MS (HRMS) with different mass analyzers including TOF and Orbitrap. HRMS can acquire full-spectrum data which can provide specificity based on accurate mass analysis and has the capability of retrospective analysis of non-targeted analytes. The recent advances in HRMS instrumentation have improved its quantitative capability, enabling both highly specific screening and accurate quantitation. A review on high-resolution MS approaches gave an

overview of publications from January 2011 until March 2014 for the quantitation of drugs and related compounds in biological samples using LC-MS with TOF or Orbitrap. [288]

The identification of synthetic cannabinoids and their metabolites in blood [289] and urine [167, 290] have been done by LC-Q-TOF-MS. The Q-TOF-MS has the single MS and tandem MS modes for data acquisition. The single MS mode provides easy drug screening by accurate mass measurement of target ions and the tandem MS mode provides confirmation or structure estimation by reliable assignment of product ions. The LC-Q-TOF-MS techniques can also be applied for target screening of abused drugs and toxic compounds [291, 292] and designer drugs (cathinones, hallucinogenic phenethylamines and piperazines) [188] in blood, and sport drugs in urine [293]. Furthermore, the LC-Q-TOF-MS has demonstrated its potential in toxicological screening of drugs and metabolites [187, 294, 295].

The liquid chromatography coupled with high resolution and high mass accuracy Orbitrap mass spectrometry can provide full-scan acquisition with good sensitivity for detection of a large number of target and non-target compounds. This technique is especially well suited for wide-scope screening in forensic toxicology. Compounds including drugs of abuse [289], novel psychoactive stimulants [186], cardiovascular drugs [296], mycotoxin [297], pregabalin [298], pesticides [299] and rodenticides [300] have been successfully analysed. A method of toxicological screening of 616 compounds in three biological matrices (*i.e.* authentic serum, urine and whole blood) using turbulent flow chromatographic system coupled with an Orbitrap mass spectrometer was also developed [301]. Identification was based on retention time, accurate mass, isotopic pattern and presence of specific fragments.

3.2 Extraction/Sample treatment Techniques

With the improved sensitivity of instruments, sample treatment simplifications, such as dilute and shoot, **Quick, Easy, Cheap, Effective, Rugged and Safe** (QuEChERS) protocols, extraction of dried blood spots, were feasible. The advancement in materials science, the development of new materials and miniature fabrication techniques also nourished the advancement of microextraction techniques. Both extraction simplifications and microextractions allow a great potential for automation of extraction for analytes from complicated biological matrices required in toxicology.

3.2.1 Extraction Simplifications

3.2.1.1 Dilute and Shoot

Urine drug testing is a noninvasive sampling whereas drugs and metabolites are usually present in high concentrations and relatively long detection windows. Solid-phase extraction (SPE) is frequently used for sample cleanup in recent LC-MS/MS method reports of urine drug testing. However, SPE involves laborious procedures and has inconsistent recoveries for all analytes, especially when many analytes with a wide variety of physical and chemical properties are measured simultaneously. Recently, dilute injection or dilute-and-shoot LC-MS/MS has been successfully employed as an alternative technique in urine drug testing [302, 303]. Dilution was reported to be done with an internal standard [304] or 5-fold with deionised water. Dilution can minimize the sampling procedure and matrix effect. Moreover, there will be no loss of analytes during sample preparation.

Tsai *et al.* has reported the screening and confirmation of drugs of abuse and metabolites in urine by dilute-and-shoot LC-Q-TOF-MS [134]. Using this method, 62 abused drugs and metabolites could be accurately determined with high confidence. The dilute-and-shoot LC-MS/MS method has also been employed in the urine screening of drugs of abuse [305], opioids [306], prescribed drugs for chronic pain management [307], melatonin and cortisol [304], and for tolvaptan [308] and stimulants and narcotics [309] in doping control analysis.

Other than urine, the dilute and shoot LC-MS method was also developed for quantitation of cannabinoids [310] and opioids [311] in OF. These applications illustrated that the dilute-and-shoot method can provide a simple but robust solution in urine drug testing.

3.2.1.2 QuEChERS

Anastassiades first published in 2003 for analysis of pesticide residues in produce, has also been used in forensic toxicology these years. Cappiello and co-workers has reported the analysis of benzodiazepines in beverage using QuEChERS for forensic application such as drug-facilitated crimes [312]. It has been also used for analysis of organophosphorus pesticides in stomach contents of postmortem animals [313], insecticides, methomy and aldicarb in guinea pig blood and brain tissue [314], and 125 pesticides in human whole blood and gastric contents in forensic applications, [315]. QuEChERS has also adopted in analysis of drugs of forensic interest in blood, including benzodiazepines and abused drugs [140, 316, 317], as well as study of designer drugs in postmortem samples of fatal cases [237, 243, 252, 255]. A therapeutic drug, ivabradine, intoxication case was also reported using QuEChERS extraction followed by LC-MS analysis [318].

3.2.1.3 Extraction of Dried Blood Spots (DBS)

Drug analysis of DBS had been used in clinical analysis since the 1960s. There has been increasing interests in DBS analysis, which has advantages such as very small volume of sample required, easy sampling, easy storage, better sample stability and simple sample treatment for analysis. Review articles on validations of bioanalysis [319], therapeutic drug monitoring (TDM) using DBS [320] and the use of mass spectrometry for DBS analysis [321, 322] summarized the progress and development of DBS analysis, and also discussed about various analytical challenges. The earliest reported DBS-based assay validations only cover the general bioanalytical validation parameters mentioned by the Food and Drug Administration (FDA) and the European Medicines Agency (EMA) [320]. The parameters mainly described in these recommendation articles are spot homogeneity, influence of Hematocrit (HT) and the influence of spot volume [320]. The studies published on DBS sampling in TDM frequently show an adequate analytical validation, but limited clinical validation [321]. Promising developments are Dried Plasma Spot (DPS) using membranes and Hct correction using the potassium concentration [321]. The recent applications were essentially driven by improved sensitivity of triple quadrupole mass spectrometers and an overall view of all instrumental and methodological developments for DBS analysis with mass spectrometric detection, with and without separation techniques was reviewed [322].

HT issue is one of the most discussed challenges in DBS analysis, and there have been numerous studies for various strategies to cope with this [323]. These strategies focus on overcoming the issue of differential spreading of blood on filter paper, as this is generally considered to be the major contributor to HT-induced bias. The influence of HT on recovery and the matrix effect may be minimized by optimizing the extraction or chromatographic conditions. A study demonstrated that the use of "whole spot" prepared by 15 l blood was effective in eliminating the HT effect [324]. There had been work reported a new DBS card, based on hydrophilic-coated woven polyester fibers, showed spot sizes were independent of the HT value of blood [325]. However, an investigation of the performance of 5 different cellulose-based DBS cards showed difference in performance, especially under extreme concentration and HT values, and it would be useful to study DBS card performance before method validation [326]. To overcome the HT issue in quantitative analysis of DBS, numerous articles discussed the use of microsampling technique, which used whole spot prepared by a fixed volume of blood, usually about 10 l [327, 328, 329, 330]. There was also investigation of different approaches to incorporate internal standard in BDS quantitative analysis to nullify HT effect [331].

One of an advantage of DBS is the ease of sampling with small amount of blood, usually obtained from fingerpricks. Several works also studied the validity of DBS sampling, usually capillary blood, as an alternative for venous blood collection in interpreting drug levels or TDM [332, 333, 334]. Cocaine and benzoylecgonine concentrations were qualitatively similar, but DBS had much greater variability (21.4-105.9 %CV) and were lower than in blood [332] while serum, whole blood and DBS concentrations of antipsychotics were highly identical (sensitivity 91.6-97.6%) [333]. Also, since equivalent concentrations were observed in capillary DBS and venous DBS, blood obtained by fingerprick can be considered a valid alternative for venous blood for GHB determination [334]. All these work has provided foundation for the application of DBS analysis in forensic applications. DBS had been reported to be used for postmortem analysis of drugs of abuse [335], new psychoactive substances [336], benzodiazepines [337], antipsychotics [338], antidepressants [339], doping control analysis [340, 341, 342], and also other therapeutic drugs analysis [343,344,345,346,347,348, 349,350].

3.2.2 Automations & Online Extractions

Automated online extraction can minimize the tedious manual extraction, while improve the throughput and quality. For DBS analysis, numerous work has been done on the automated on-line extraction [350, 351, 352, 353, 354], which usually include an automatic elution of DBS cards couple to LC-MS systems via online SPE. Abu-Rabie *et al.* also extended their work to include an automated IS addition system for automated quantitative DBS analysis using IS spray technique [355].

In addition to the automated system for DBS, the use of automated online extraction will certainly increase in the near future. A review of online and automated sample extraction discussed the recent development and future trends in online sample extraction methodologies including online SPE, turbulent flow chromatography, online DBS extraction, online immunoaffinity extraction. The offline automated sample extraction platform, including custom robot scripts for the automation of individual assay, and the platform for integrated multiple sample extract were also included in this review [356]. Another article also discussed the current role of online extraction approaches in clinical and forensic toxicology [357]. Numerous works has been done on method development and validation using online SPE such as toxicology analysis using online SPE coupled with LC-MS/MS for determination of tetrandrine (model drug) in human blood samples [358], LC-DAD for the quantification of acidic/neutral drugs [359] and basic drugs and metabolites [360] in human plasma, GC-MS/MS for quantification of morphine, 6-MAM, cyamemazine, meprobamate and caffeine in 11 fluids and tissues [361].

Another great potential for automation is the use of microextraction [362, 363, 364], which can be readily coupled to analytical instruments and hence facilitates the design of automated on-line systems.

3.2.3 Microextraction

Microextraction, which has advantages such as reduced sample volume, simple analytical protocol and minimized the reagent consumption, attracted research interest and focus in the development, investigation and application of different types of microextraction techniques. A recent review has discussed and given an overview of the development of microextraction techniques, both well-established and recently appeared microextraction for the clinical analysis of biological sample [365]. A review provided a comprehensive survey of past and present microextraction methods for determination of various analytes in urine samples followed by GC analysis [366]. Another review also summarized the current achievements and application of microextraction techniques and is dedicated to the description of microextraction techniques and their application in biomedical analysis [367].

A topical collection of Analytical and Bioanalytical Chemistry in 2014 has been devoted to new approaches of microextraction techniques. This collection has included critical reviews on microextraction with high-throughput potential for medical diagnostic purposes [364], dispersive liquid–liquid microextraction (DLLME) [368] and stir-bar sorptive extraction (SBSE) [369], discussing the state of art and future trend, as well as their various applications.

3.2.3.1 Solid phase microextraction (SPME)

SPME was proposed by Authur and Pawliszyn in 1990, the rapid growth of this application was proved by the numerous reviews focused on the use of SPME, including its recent trends and applications to medical diagnostic, particularly the in vivo and near real time applications [364]; its features and applicability for clinical bioanalysis as a low invasive sampling and sample preparation tool for urine, blood, limited availability samples (tissues), and alternative matrices such as hair, saliva, sweat or breath were discussed. [370]; the use of SPME in analysis of drugs in biological samples [371]; its recent applications and advances in the bioanalytical and clinical applications [372]; selective capillary coating materials for in-tube SPME coupled the LC for determination of drugs and biomarkers in biological samples [373]. There were numerous publications on modifications and improvement of the technique, and its applications. A publication presented the direct coupling of biocompatible SPME fibres to MS via nanoelectrospray ionization (nano-ESI) for fast quantitative analysis of target analytes in biofluids [374]. The sample preparation time of the method was less than 2 minutes and sample volume used could be down to 10 L, suggesting it a powerful tools in bioanalytical applications, such as toxicology and doping control. Another research modified the SPME devices to potential single-use samplers by preparing the thin-film SPME devices on cheap and chemical resistant plastic support, made of polybutylene terephthalate. The devices had been evaluated using plasma and whole blood without observable deterioration of performance or detachment of extraction phase from the plastic support. This work opened up the possibility of using a cost effective materials as support for SPME device [375]. New materials for SPME extract phase were also developed. A new organic-inorganic hydrid SPME material with high efficiency in enrichment of polar analytes was introduced and it was applied to determinate ephedrine and methylephedrine in human urine by SPME coupled with GC-Flame ionization Detector (GC-FID) [376]. The fabrication of this new SPME material was based on the sol-gel procedure with mobilization of -cyclodextrin and polyethylene glycol on the anodized titanium wire surface.

3.2.3.2 Thin-film microextraction (TFME)

TFME is another sample SPME device that has been developed to improve the sensitivity and applicability to large volume sample. The TFME is usually prepared by formation of thin film of extraction phase, typically polydimethylsiloxane (PDMS) on a solid support face with procedures such as physical deposition, in-situ polymerization, sol-gel formation, etc. Extraction of analyte was simply done by submerging the thin-film into the sample solution, and then analytes can be thermally desorbed or re-dissolved into an appropriate solvent for subsequent instrumental analysis. This technique has found several applications in toxicology or clinical analysis, including quantification of prohibited substance in plasma for doping control [377], in vivo sampling of saliva for on-site monitoring of prohibited substances [378], analysis of prepaglinide for its in vitro metabolism study [379], and aldehydes in human exhaled breath condensates [380].

3.2.3.3 Microextraction by packed sorbent (MEPS)

MEPS is a modification of SPE cartridges down to microlitre level, can also be considered as a form of in-needle SPME. The MEPS can be readily connected to the analytical instrument, with great potential of automation. A critical review on microextraction with high-throughput

potential [364] has also extensively discussed MEPS. Another review focusing on MEPS technique gave an overview of the role of online/offline formats of MEPS in sample bioanalysis and discussed about the sorbents, factors affecting its performance, advantages and limitation as compared with other techniques [381]. An overview of the MEPS technique, including fields of application and common formats was discussed [382]. The key aspect of MEPS is that the solvent volume used for the elution of the analytes is of a suitable order of magnitude to be injected directly into GC or LC systems. MEPS has found its application in analysis of various biological samples, such as determination of metoprolol enantiomers in human plasma and saliva [383], β -blockers from human plasma [384], and selected drugs (milrinone, enalapril, carvedilol, spironolactone, acenocumarol, ticlopidine, cilazapril) and metabolites in human urine [385].

3.2.3.4 Stir bar sorptive extraction (SBSE) & Related Techniques

SBSE developed in 1999, is derived from SPME with similar extraction mechanism. Compared with SPME, DBSE uses about 50-250 times larger extraction phase, giving a higher recovery and sample capacity. A review on SBSE focused on its development in the past decade, in terms of coating preparation, automated systems, novel extraction modes, its use with various instruments, and applications in biological samples were published [369]. Another review summarized its applications published in the previous 3 years and most recent developments concerning its effectiveness, discussed the theory behind, and described its advantages, limitations, future trends and novel extraction sorbents and supports [386]. With continuous improvement of sorbents, the SBSE has been applied in analysis of biological fluids [387], losartan and valsartan in urine and plasma [388], diazepam and nordiazepam in human plasma [389], thyroxine in urine samples [390].

An advanced sorption based extraction technique, bar adsorptive microextraction (BAE) was introduced recently. This technique was also proved to be useful in bioanalytical applications, such as non-steroidal anti-inflammatory drugs (salicylic acid, mefenamic acid, diclofenac and naproxen as model compounds) in urine [391], mitragynine in urine [392], steroid sex hormones (estriol, 17 β -estradiol, 17 α -estradiol, 19-nortestosterone, 17 α -ethynylestradiol, estrone, D-(-)-norgestrel, progesterone and mestranol) in urine [393], anti-doping control screening of anabolic steroids (testosterone and epitestosterone) in urine [394].

3.2.3.5 Liquid phase microextraction (LPME)

LPME represents another family of microextraction techniques based on the miniaturization of the traditional liquid-liquid extraction principle by a great reduction in acceptor-to-donor ratio. Reviews on the automation of non-dispersive LPME comprehensively surveyed the developments of automation of non-dispersive LPME methodologies, discussed the automation strategies, techniques, advantages, limitations and their potentials [362, 363]. The variants of LPME has also been used in bioanalysis, such as cocaine and related compounds in human breast milk [395], phenobarbital in hair [396], amphetamines in urine with measurement uncertainty worked out [397], trihalomethanes and halo ketones in biological samples with microwaved assisted headspace LPME [398], hydrochlorothiazide and triamterene in urine by hollow fiber based LPME (HF-LPME) [399], ketamine and its metabolites in urine by HF-LPME [400], benzodiazepines and their metabolites in urine by HF-LPME [401], and antidepressants (amitriptyline, nortriptyline, imipramine and desipramine) in vitreous humor of human and bovine [402]. A comparison for the determination of drugs of abuse in urine and blood samples using HF-LPME and ultrasound-assisted dispersive liquid-liquid microextraction (UA-LDS-DLLME) was reported. Compared with HF-LPME, the UA-LDS-DLLME technique had the advantages of less extraction time, suitability for batches of sample pretreatment simultaneously, and higher extraction efficiency, while HF-LPME has excellent sample clean-up effect, and is a robust and suitable

technique for various sample matrices with better repeatability. Both methods were successfully applied to the analysis of drugs of abuse in real human blood sample [403]. More discussion on the dispersive liquid-liquid microextraction (DLLME) and related techniques will be included in the following paragraphs.

3.2.3.6 Dispersive liquid-liquid microextraction (DLLME)

DLLME was introduced in 2006 as another family of microextraction techniques. The extraction is a mode of liquid-liquid extraction with the extraction solvent scaled down to microliter range and dispersed. With the introduction of a disperser solvent and a water-immiscible extraction solvent into an aqueous sample, the contact area between the aqueous phase and the extraction phase increased drastically, and hence enhanced the extraction efficiency. The principle, recent development and applications in DLLME were summarized in a review [368]. There was another review on DLLME about some of its interesting development [404]. This review discussed the use of low-density solvent (LDS) and high-density solvent (HDS) and described many novel special devices for LDS, as well as various dispersion techniques with LPME. Editorial highlighted the review was in attempts to chronicle the developments in DLLME in recent years, after the first paper (published in 2006) established a very fertile area of research in sample preparation and to signify the extension of the development of DLLME, beyond its basic implementation. To enable routine application of the procedure to real world problems, more operational convenience and some form of automation would be needed [405]. Later in 2015, an article has presented an overview of DLLME applications in the analysis of biological samples (e.g. plasma and urine) [406]. DLLME has been adopted in analysis of biological specimens, such as analysis of drugs of abuse (morphine, 6-MAM, cocaine, benzoylecgonine and methadone in human plasma) by HPLC-PDA [407], melatonin in plasma by HPLC-UV [408], basic tricyclic antidepressant drugs (nortriptyline, imipramine, and amitriptyline) in by tandem DLLME (TDLLME) with two DLLME combined in human plasma by TDLLME-HPLC-UV [409], fentanyl in urine with GC-MS [410], antipyrine in saliva [411], immunosuppressive drugs (cyclosporine, everolimus, and sirolimus) in human urine and serum by surface-assisted laser desorption/ionization MS [412], new psychoactive substances in whole blood by UPLC-MS/MS [138].

3.2.3.7 Ultrasound-assisted emulsification microextraction (USAEME)

USAEME is a modified DMLLE in which dispersion of solvents were aided by ultrasound instead of the dispersive agent. Recent applications of this procedure for analysis of biological samples include the determination of drugs of abuse in biological samples by GC-MS [403] and GC-MS/MS [413], fluoroquinones in human body fluids (plasma and urine) by HPLC-FLD (fluorescence detector) [414], nitrazepam and midazolam in human serum [415], ibuprofen and metabolites by HPLC-MS/MS [416] and benzodiazepines by UPLC-UV in human urine [417]. These methods achieve detection limits down to ng/mL or lower order in general.

3.2.3.8 Ionic liquid dispersive liquid-liquid microextraction (IL-DLLME)

IL-DLLME is another version of DLLME using the ionic liquids as extracting solvent instead of using organic solvents. The minimum used of volatile organic solvents addressed the safety and environmental concerns. IL-DLLME was applied to analysis of biological samples such as the determination of balofloxacin in rat serum [418]. In addition, ultrasound radiation had been also used to accelerate the emulsification in IL-DLLME and this modified technique was also used for bioanalysis including, determination of lornoxicam in human serum [419], antidepressant and antipsychotic drugs (doxepin and perphenazine) [420] and bioactive compounds (anethole, estragole and para-anisaldehyde) [421] in urine. In general, the above methods attained a detection limit down to ng/mL level with recoveries of about 90% or higher.

3.3 Drugs Screening for Toxicology

Unknown drug screening is the first and essential step in toxicology analysis. To achieve the systematic toxicology analysis, various techniques includes, HPLC-DAD, GC-MS, LC-MS and *etc.* would be adopted.

3.3.1 LC-DAD & GC-MS in Drugs Screening

Both HPLC-DAD and GC-MS had been the reference methods for many years, and were still widely used for drug screening, especially with a large database of spectral libraries established over decades. Mut *et al.* [359, 360] had recently developed and validated automated multi-analytes screening methods for the identification and quantification of 92 basic drugs and metabolites, and 50 acidic/neutral drugs based on on-line SPE-LC-DAD. In addition, a database with 870 entries including UV-spectra, relative/retention times and response factors of toxicologically relevant compounds was created. Plasma samples (0.2 mL for basic drugs and 0.1 mL for acidic drugs) were treated with appropriate solvents for protein precipitation prior to on-line SPE extraction and subsequent LC-DAD analysis. A computer-assisted database generated for toxicologically relevant analytes allowed automated peak detection, compound identification and quantification as well as automated reporting of results. The methods have the potential of extending to other analytes owing to the large number of entries in the generic library.

There had also been efforts in the improvement in GC-MS for general screening for toxicology. Ramoo *et al.* [422] described an efficient and rapid GC-MS method for comprehensive drug screening in urine which utilized a liquid-liquid extraction, sample concentration, and analysis by GC-MS. The method is validated for detecting more than 150 drugs within a multitude of drug classes with the use of commercial libraries, and the use of pre-established in-house relative retention time of corresponding drug reference standards for verification of drugs identification. Grapp *et al.* [423] evaluated the use of automated mass spectral deconvolution and identification system (AMDIS) for GC-MS based toxicology serum screening for analysis of 150 serum samples after neutral and basic liquid-liquid extraction. As compared with manual data evaluation, the number of false positive hits using AMDIS-based data evaluation could substantially be reduced without increasing the risk of overlooking relevant compounds.

Yuan *et al.* [424] had published a review on drug confirmation by MS coupled with chromatography, summarizing assay parameters required for drug confirmation based on recent scientific publications, various established guidelines, practical experience. Factors affecting the result quality and correct results interpretation are also critically reviewed. Emerging technologies, such as HRMS, and their potential applications were also discussed briefly.

3.3.2 Use of LC-QqQ & LC-Ion Trap in Multi-drugs Analysis

LC-MS has become a focus for development in screening of biological specimen in both clinical and forensic toxicology. Amongst the LC-MS techniques, ion trap MS instrument generating MSⁿ spectrums was a less discussed option, as compared to the LC-triple quadrupole instrument (QqQ). Kempf *et al.* [425] had developed a screening method for the detection of psychotropic drugs based on the open library concept of a recently developed LC-MSⁿ (ion trap) screening approach, and evaluated the effectiveness of a heated ESI-source and an ionBooster™ (ESI/IB). In their project, an individual spectral library was set up by transferring all available data of psychotropics from the Toxtyper™ library to a new library format. Precursor masses and retention time information of 105 psychotropic

substances and metabolites of the library were used to trigger data dependent acquisition of MSⁿ-spectra. Method evaluation was performed using pooled serum samples fortified with 12 different mixtures containing a total of 99 compounds at low therapeutic concentrations. The customized ESI/IB ionisation led to a higher rate of identifications (92%) - especially at low concentration levels - as the comprehensive screening approach (87%). The generated screening method with Toxtyper open library concept is a fast and robust tool for the detection and identification of 105 psychotropics in human serum at and can easily be extended by adding new compounds. Low therapeutic levels for the majority of compounds made the screening applicable for clinical and forensic samples (intoxication and post mortem cases).

In the last decade, LC-MS/MS screening was mainly using QqQ instrument with multiple reaction monitoring (MRM). There had been lots of efforts in method development of multi-analytes screening in various matrices by LC-MS/MS, using the MRM mode, for different applications, such as general forensic toxicology, screening for drug-facilitated crime cases and doping control [136, 193, 307, 426, 427, 428, 429, 430]. Di Rago *et al.* [426] developed an LC-MS/MS based screening technique that covered a broad range of acidic and neutral drugs and poisons using protein precipitation of 100 µL of whole blood with simple automated data processing. In this method, 132 common acidic and neutral drugs and poisons were detected using an LC-MS/MS system by monitoring two MRM transitions per analyte with ESI of both positive and negative mode. Quantification data obtained using one-point calibration compared favorably to that using multiple calibrants.

Dixon *et al.* [427] also evaluated the use of LC-MS/MS using MRM mode by comparing the results of 31 urine specimens collected from patients taking benzodiazepines against the results by immunoassay. The true analyte concentrations of the 31 urine specimens were determined (after hydrolyzing glucuronide metabolites using beta-glucuronidase) using LC-MS/MS. Those urine specimens were reanalyzed using EMIT benzodiazepine assay and Vista analyzer. The LC-MS/MS values were above the 200 ng/mL cutoff concentration, but EMIT benzodiazepine assay showed false negative results (35.5%) from 11 out of the 31 specimens analyzed, indicating that despite hydrolysis of the specimen to liberate parent drug (glucuronide metabolite often has poor cross-reactivity). Their work concluded that patient compliance with benzodiazepine therapy must be monitored using LC-MS/MS.

Montenarh *et al.* [428] had developed and validated an LC-MS/MS multi-analyte approach based on a simple liquid-liquid extraction for target screening and quantification of benzodiazepines and Z-drugs in case of driving ability and crime responsibility in whole blood, plasma, and serum. The biosamples (500 µL each) were extracted twice at pH 7.4 and at pH 10 with ether/ethyl acetate (1:1). Separation, detection and quantification were performed using LC-MS/MS by ESI in positive mode. Full calibration was performed with ranges from sub-therapeutic to toxic concentrations. The approach was selective, sensitive, accurate, and precise for 16 of the 19 tested drugs in whole blood, 18 in plasma, and 17 in serum. Only semi-quantitative results could be obtained for zopiclone because of its instability in all tested biosamples.

A minimum list of 80 analytes to be monitored in drug-facilitated crime cases had been proposed by the Society of Forensic Toxicologists (SOFT) including the recommended minimum performance limits (RMPL). Remane *et al.* [136] had developed and validated two LC-MS/MS based screening procedures, one in positive (91 basic in method I) and one in negative (nine acidic in method II) ESI mode for detection 100 analytes. Gradient elution was performed on a ZORBAX Eclipse XDB-C18 column after protein precipitation of the urine samples. Detection was carried out in the scheduled MRM mode monitoring two transitions per compound. No interferences were observed in 30 tested blank urine

samples. The RMPLs were achieved for all analytes and ranged from 1 ng/mL for fentanyl to 10 µg/mL for γ-hydroxybutyrate (GHB). Results for urine specimens from nine authentic DFC cases were always negative with exception of drugs prescribed to the victims. Reanalysis with the developed procedure of 24 urine samples, with a positive screening result during routine clinical toxicology analysis, confirmed the routine findings. In an excretion study after a single oral doxylamine dose (30 mg), the parent drug and its nor-metabolite could be detected in urine specimens from a young female volunteer for 10 days. The developed procedure allowed a selective and sensitive screening of urine samples for almost all recommended analytes relevant in DFC cases.

There were other publications in drugs/steroids determination in urine by LC-MS/MS by MRM modes. Cao *et al.* [307] had developed a simultaneous quantification method for the 78 drugs and metabolites in urine with dilute-and-shoot approach. 72 analytes were detected with positive electrospray ionization mode and the remaining 6 analytes with negative mode. A suboptimal recovery rate (60.0-156.8%) was observed for six analytes, potentially due to the lack of available deuterated ISs, requiring comparison to a chemically different IS. Another screening method for 18 frequently measured exogenous anabolic steroids and testosterone/epitestosterone (T/E) ratio in forensic cases has been developed and validated by Andersen *et al.* [429]. The method involves a fully automated sample preparation including enzyme treatment, addition of internal standards and SPE followed by analysis by LC-MS/MS using ESI with adduct formation for two compounds. The LODs ranged from 2 to 40 ng/mL. In the 580 urine samples analyzed from routine forensic cases, 17 (2.9%) were found positive for one or more anabolic steroids. Only seven different steroids including testosterone were found in the material, suggesting that only a small number of common steroids are likely to occur in a forensic context. The steroids were often in high concentrations (>100 ng/mL), and a combination of steroids and/or other drugs of abuse were seen in the majority of cases.

There had been efforts in screening for NPS in biological specimens by LC-MS/MS with MRM mode. Tang *et al.* [430] had studied and established a chromatography/MS-based analytical system for the simultaneous detection of conventional drugs of abuse and NPS in urine. Sample preparation entailed enzyme digestion and SPE; analytes were then detected by LC-MS/MS with MRM. Forty-seven conventional drugs (28 parent drugs, 19 metabolites) and 46 NPS analytes (44 parent drugs, two metabolites) were covered by the established method, which had been validated according to international guidelines. The method was then applied to 964 urine samples collected from drug abusers and the results revealed the presence of two NPS - TFMP and methcathinone - as well as conventional drugs of abuse. The method had been successfully applied to authentic specimens revealing the presence of conventional as well as novel drugs of abuse in their local population. Poklis *et al.* [193] also presented a LC-MS/MS method for the identification and quantification of nine serotonin 5-HT_{2A} receptor agonist hallucinogenic substances from a new class of NBOMe designer drugs in human urine: 25H-NBOMe, 2CC-NBOMe, 25I-NBF, 25D-NBOMe, 25B-NBOMe, 2CT-NBOMe, 25I-NBMD, 25G-NBOMe and 25L-NBOMe in response to an outbreak of N-benzyl-phenethylamine derivative abuse and overdose cases in Virginia. The NBOMe derivatives were rapidly extracted from the urine specimens by use of FAST™ SPE columns. Linearity was verified to be from 1 to 100 ng/mL for each of the nine analytes. The bias determined for the NBOMe derivatives was 86-116% with a <14% coefficient of variation over the linear range of the assay. Four different NBOMe derivatives were detected using the presented method in patient urine specimens.

Stone [431] had reported the use of Liquid Chromatography-Hybrid Triple-Quadrupole Linear Ion Trap Mass Spectrometer (LC-QqLIT) for a broad-spectrum drug screening in urine. Urine is processed with a simple C18 SPE and reconstituted in mobile phase. An initial 125

compounds SRM survey scan was processed by the SRM-information-dependent acquisition (IDA) algorithm. The IDA algorithm selected SRM signals from the survey scan with a peak height above the threshold to define precursor ions for subsequent dependent scanning. Enhanced (meaning acquired in LIT mode) Product Ion (EPI) spectra were automatically searched against a 125 drug library of reference EPI spectra for identification.

Thoren *et al.* [432] developed a broad-spectrum drug screening on a LC-HRMS (QqTOF) that collected data in an untargeted manner and compared its performance to a nominal mass instrument QqLIT that collected data in a targeted manner. With the evaluation of 100 routine clinical urine samples, it suggested that QqTOF performed similarly to QqLIT in screening of drugs in urine samples and could serve as an alternative to SRM-based methods in broad-spectrum drug screening.

3.3.3 Use of LC-HRMS in Broad Spectrum Screening

Recently, as discussed in the “Instrumentation Advances” section, the HRMS has gained lots of attention in toxicology screening area with their potential of setting up an untargeted general unknown screening. In the work of Beck *et al.* [302], they developed methods for urine drug testing using one-step dilution and direct injection in combination with LC-MS/MS and LC-HRMS, and concluded that the non-discriminating nature of LC-HRMS made it an even more attractive option for multicomponent target and general unknown analysis applications.

The application of HRMS, including the Orbitrap and TOF MS, for untargeted drug screening was also discussed by Wu and Colby [433]. With this technique, presumptive identification of unknowns could be conducted without the need to match MS library spectra or comparison against known standards so as to greatly expand on the number of drugs and metabolites that could be detected and reported on a presumptive basis. Definitive assignments of the compound's identity could be retrospectively determined with acquisition of the appropriate reference standard.

Chindarkar *et al.* [434] had investigated using elevated collision energies (MS(E)) for broad-spectrum drug screening by LC-TOF-MS analysis and concluded that MS(E) provides a unique way to incorporate fragment ion information without the need of precursor ion selection. A primary limitation of requiring a fragment ion for positive identification was that certain drug classes required high-energy collisions, such as norphine, which formed many fragment ions of low abundance that were not readily detected.

Teng *et al.* [292] developed a novel method for the screening of 151 drugs of abuse and toxic compounds in human whole blood by online SPE with LC-TOF. Analytes were extracted using a fully automated online SPE and separated by LC system with run time of 26 min. TOF-MS screening of 151 drugs of abuse and toxic compounds was performed in a full-scan (m/z 50-800) mode using an MS^E acquisition of molecular ions and fragment ions data. The compounds were identified based on retention times and exact mass of molecular ions and fragment ions. The LODs ranged from 1 to 100 ng/mL and the recovery of the method ranged from 6.3 to 163.5%.

In the study of Lung *et al.* [435], the utility of non-targeted comprehensive drug screening by LC-TOF in the agitated patients in an emergency department setting was assessed. Upon analysis, it was found that seven different NPS (JWH-073, JWH-081, JWH-200, methylenedioxybenzylpiperazine, mephedrone, methoxetamine and herkinorin) were detected from six patient samples out of 23 patients with severe psychomotor agitation. This study demonstrated that the non-targeted NPS screening in a selected emergency department patient population is feasible and effective in identifying NPS.

Concheiro *et al.* [186] developed a sensitive and specific confirmation method for the 40 NPS stimulants and 4 metabolites in urine by LC-hybrid quadrupole-Orbitrap MS (Q-Exactive). With the use of 100 μ L urine, LOQs of 2.5-5 μ g/L were achieved. Data were acquired in full scan and data-dependent acquisition (DDA) MS² mode. This full scan DDA-MS² approach also offers flexibility to include additional NPS as a broad spectrum screening method with minimal method validation steps saving time and resources.

Helfer *et al.* [296] had tested Orbitrap technology for developing a general metabolite-based LC-HRMS/MS screening approach for urinalysis of drugs necessary in clinical and forensic toxicology. After simple urine precipitation, the drugs and their metabolites were separated within 10 min and detected by a Q-Exactive mass spectrometer in full scan with positive/negative switching, and subsequent Data Dependent Acquisition mode. Identification criteria were the presence of accurate precursor ions, isotopic patterns, five most intense fragment ions, and comparison with full HRMS/MS library spectra. Their library contained over 1900 parent drugs and 1200 metabolites. The applicability was tested and shown to be useful firstly for cardiovascular drugs, which should be screened for in poisoning cases and for medication adherence of hypertension patients.

3.3.4 Acquisition Modes for Systematic Toxicological Analysis (STA)

Sensitive and selective LC-MS analysis was a powerful and essential tool for metabolite identification in drug discovery and development. It appeared that data analysis method was also an essential part in the detection. It was more often that IDA approaches were employed in LC-MS/MS techniques in forensic and clinical toxicological screening procedures. The complexity of a sample and the IDA settings might prevent important compounds from being triggered. Several works suggested sequential windowed acquisition of all theoretical fragment-ion spectra (SWATH) could be a preferred mode for sSTAbY LC-HRMS [436,437,438]. An MS² (or tandem, MS/MS) mass spectrum was acquired from the fragmentation of a precursor ion by multiple methods including information-dependent acquisition (IDA), SWATH, and MS(All) (also called MS^E) techniques. Zhu *et al.* [436] compared the three techniques in their capabilities to produce comprehensive MS² data by assessing both metabolite MS² acquisition hit rate and the quality of MS² spectra. Rat liver microsomal incubations from eight test compounds were analyzed with four methods (IDA, MMDF (multiple mass defect filters)-IDA, SWATH, or MS^E) using an UHPLC-Q-TOF MSplatform. Overall, IDA-based methods acquired qualitatively better MS² spectra but with a lower MS² acquisition hit rate than the other two methods. SWATH outperformed the MS(All) method given its better quality of MS² spectra with an identical MS² acquisition hit rate.

Roemmelt *et al.* [437] employed SWATH method, which used Q1 windows of 20-35 Da for data-independent fragmentation, was systematically investigated for its suitability for STA. Quality of SWATH-generated mass spectra was evaluated with regard to mass error, relative abundance of the fragments, and library hits. Screening results of 382 authentic forensic cases revealed that SWATH's detection rate was superior to IDA, which failed to trigger ~10% of the analytes. Arnhard *et al.* [438] also studied the use of SWATH for systematic toxicology analysis with LC-HRMS. Their experiments revealed that SWATH was a sensitive and specific identification technique, capable of identifying more compounds at lower concentration levels than IDA approach.

3.3.5 General Unknown Screening (GUS) in alternative specimens

Besides blood and urine, LC-MS methods had been developed for GUS in other matrices. Maublanc *et al.* [142] developed a multianalytes method allowing simultaneous identification and quantification of 35 psychoactive drugs by LC-MS/MS. After incubation of 50 mg of hair

in a phosphate buffer pH 5 for one night at room temperature, followed by extraction with a dichloromethane/ether mixture (70:30, v/v). The data acquisition was performed in scheduled MRM mode. Intra- and inter-day precisions, estimated using the coefficient of variation and relative bias, were lower than 20 % for all concentration levels, except for two compounds. The LODs and LOQs ranged from 0.5 to 10 pg/mg. The method has been successfully used in several forensic cases, three of which were reported.

Montenarh *et al.* [439] had developed an LC-MS/MS multi-analyte approach with ESI in positive mode using one single work-up approach in whole blood, plasma, serum, post-mortem blood, liver tissue, gastric content, hair, and urine for 130 analytes. For identification, a scheduled MRM (sMRM) method with 390 transitions was developed covering benzodiazepines, Z-drugs, antidepressants, neuroleptics, opioids, new synthetic drugs, and phosphodiesterase type 5 inhibitors. The simple work-up procedure was suitable for all biosamples with exception of urine in respect to low concentrated analytes, which showed median recovery values of 59%. The method was selective for 130 analytes in all 8 biosamples. For 106 analytes, the LODs in whole blood, plasma, and serum was lower than the lowest therapeutic concentration listed in blood level lists.

With the use of LC-Q-TOF, Krumbiegel *et al.* [440] studied the usefulness of nail samples instead of hair for a GUS for drugs as an alternative matrix for long term detection. Only 10% of the cases showed a disagreement of results in hair and nail analysis where hair samples were tested positive and corresponding nail samples were tested negative in a general unknown screening for drugs. The incorporation of a large number of substances into the nail matrix was proven by the detection of 89 different analytes (e.g. antidepressants, drugs of abuse or antihypertensives) in the tests. They concluded that in cases where the amount of hair available was not sufficient for a general unknown screening for drugs, nails appeared to be a useful comparable matrix for the detection of long-term drug consumption.

Stephanson *et al.* [441] developed a LC-MS/MS method for drugs of abuse testing in exhaled breath employing a sampling device collecting aerosol particles and applied in routine use. Analytes covered were amphetamine, MA, 6-MAM, morphine, cocaine, benzoylecgonine, diazepam, oxazepam and THC. The method involved eluting drugs from the collection filter with methanol, quantification using deuterated analogs as internal standards. The measuring range was 6.0-1000 pg/filter. The LOQ was 6.0 pg/filter with intra-assay CV <5% and accuracy within 99-102% for all analytes. Among the 1096 analyzed samples analytical findings were made in breath in 39 cases (3.6%). Most frequently found substances were amphetamine (25 cases), MA (10 cases), THC (8 cases), cocaine (4 cases), benzoylecgonine (2 cases) and diazepam (2 cases).

3.4 Analysis of Specific Drugs

3.4.1 Volatile Compounds

Carbon monoxide (CO) poisoning is common and often seen in accident or suicide cases. CO could be produced from incomplete combustion of charcoal or carbon fuels, motor vehicles and household fires. Among the suicide by gases in England and Wales between 2001 and 2011, CO was the most frequently used gases in the suicidal cases [442]. CO poisoning cases in Wuhan, China between 2009 and 2014 [443], in Denmark between 2009 and 2012 [444] and in Portugal between 2000 and 2010 [445] were reported. The studies showed that charcoal burning is mainly found as the major cause in suicidal death while fire accident in accidental death.

Blässer *et al.* reported a case of suicidal carbon monoxide poisoning in a car using a gas-powered generator. The carboxyhaemoglobin (COHb) content in the deceased blood was

found to be 68%. A simulation was conducted using the decedent's car. CO concentration already reached the lower explosion limit of 500 ppm after 30 second and the engine started tuttering with about 14 vol. % of oxygen in the air and the experiment was stopped after 25 minutes. The author concluded that it is to be assumed that the victim lost unconsciousness quickly due to rapid increase in CO concentration in the interior of the vehicle [446].

A suicidal poisoning case by inhalation of CO produced by combining formic acid and sulfuric acid was reported by Lin *et al.* [447]. Suicide with formic acid and sulfuric acid is rare in the US with only three prior cases being reported. Greater awareness of this method among death investigators is warranted because of the special risks of accidental intoxication by toxic gas.

Ferrari and Giannuzzi quantified the COHb, total haemoglobin (tHb), methaemoglobin (MetHb) and hydrogen cyanide (HCN) content in 32 out of 33 blood sample from forensic autopsy cases in a disastrous polyurethane mattress fire, which caused 33 inmates died at a prison in Argentina in 2006. A relationship between COHb and MetHb in blood was established, however, no correlation was found between HCN and MetHb in the blood of the victims. In addition, the HCN level in the blood of the majority of the fire victims was larger than 1 mg/L, which could be lethal, while in the majority of fire victim, the level of COHb found in the blood was below the level considered to be lethal [448].

The benzene concentration in blood was previously found to be positively correlated with COHb in fire-related deaths. Oshima *et al.* recently reported 3 cases in which benzene and COHb concentration were not correlated. A high COHb concentration without a hydrocarbon component, such as benzene, indicates that the deceased inhaled CO that was not related to the smoke from the fire [449].

The oxyhaemoglobin (%O₂Hb) and %COHb ratios in right and left cardiac blood in fatal hypothermia and death by fire in 690 forensic autopsy cases were investigated by Kanto-Nishimaki *et al.* The %O₂Hb in the left cardiac blood was found to be significantly higher than that in the right cardiac blood in fatal hypothermia case. In addition, %O₂Hb in the left cardiac blood increases with O₂ inhalation but that in the right cardiac blood increases in parallel in resuscitated group. Furthermore, postmortem cooling appeared not influencing %O₂Hb in cardiac blood. Besides, the author proposed that %COHb of greater than 10% is a reliable maker of antemortem CO inhalation regardless of smoking history in case of death by fire [450].

Butane, which is commonly used as lighter refills and gas cartridge for portable gas stoves, may be misused by inhalation of the gas by some people. Balkhi *et al.* reported a case in France concerned a 15-year-old boy found dead after sniffing a cigarette lighter refill. Toxicological investigation revealed the presence of butane in the heart and femoral blood (1280 and 1170 µg/L, respectively) with propane also detected at concentrations approximately tenfolds lower. From the review of the literature, all the reported cases had the butane postmortem blood concentration of larger than 100 µg/L [451].

Sasao *et al.* reported three cases of butane sniffing death. Butanes, including n-butane and isobutane, and two n-butane metabolites, 2-butanol and 2-butanone, were quantified in the cases. n-Butane and isobutane were detected in all three cases. n-Butane concentrations in heart blood of the three cases were found to be in the range of 25.5-54.3 µg/mL which were considered fatal according to the previous reports. The two metabolites were detected in two of the cases of which the decedents had chronic butane sniffing history while the metabolites were not detected in a case where the decedent had no history of chronic butane use. The author concluded that the detection of n-butane metabolites could be a

useful method for understanding the decedents' pattern of butane sniffing before death [452].

Okuda *et al.* reported a case in which the concentration of n-butane found in the specimen was in the range of 0.48-70.5 $\mu\text{L/g}$ which was considered as toxic or lethal level, but the decedent has no trances of butane use before the fatal traffic accident. According to police investigation, an anticontagious plugging spray, which contains butane, was used in postmortem treatment at the emergency hospital prior to forensic autopsy [453].

Toluene is another volatile substance often chosen for illicit use for abusers. A death case analysis revealed 0.73 promille alcohol and 8 $\mu\text{g/mL}$ toluene in the blood and the cause of death was considered as toxicity due to acute combined intake of alcohol and toluene was reported by Güress *et al.* [454].

A fatal case of acute tetrachloroethylene intoxication in a chronic abuser was also reported by Amadasi *et al.* [455]. Toxicological and histological investigations demonstrated the presence of an overlap between chronic intake of the substance and acute intoxication as final cause of death, with a concentration of 158 mg/L in cardiac blood and 4915 mg/kg in the adipose tissue.

The presence of the volatile substance, dimethylether, was qualitatively identified in brain tissue of a 38-year-old man with history of anxiety, depression and post-traumatic stress disorder. The body was in a moderate state of decomposition surrounded with a number of aerosol cans [456].

Giuliani *et al.* reported a validated analytical method for the determination of nitrous oxide (N_2O), also called 'laughing gas', and the method was applied to a N_2O lethal intoxication case. The concentrations N_2O in the brain, peripheral blood and lungs were found to be 47 g/g, 27 g/g and 370 g/g, respectively [457].

Matrix effect in the analysis of volatile organic compounds (VOCs) in whole blood with SPME was evaluated by Aloneso *et al.* It was found that dilution of 1:2 (blood/water) was enough to allow quantitative recoveries of those compounds with boiling points less than 100°C , while dilution of 1:5 (blood/water) is required for compounds with boiling points between 100 and 150°C . And for compounds with boiling points greater than 150°C , the blood matrix was too strong that dilution would not be adequate [458].

3.4.2 Herbal Medicine

Aconitum alkaloids are highly toxic cardiotoxins and neurotoxins [459]. In Hong Kong, the incidence of aconite poisoning was relatively low in January 2000-June 2004 (0.03 per 100 000 population). However, aconite poisoning increased in April 2004-July 2009 and 2008-2010 (0.15 and 0.28 per 100 000 population). Overdoses and use of inadequately processed aconite roots were important causes [495]. The tubers and roots of *Aconitum* are of great importance in traditional Chinese medicine (TCM) for treatment of various diseases, such as collapse, syncope, rheumatic fever and painful joints [460]. Aconite roots are also eaten as root vegetables and use to prepare herbal soups and meals in Asian communities. *Aconitum* alkaloid poisoning related to culinary use of aconite roots was reviewed by Chan [461]. In some cases, *Aconitum* alkaloid poisoning because of contamination of herbs by aconite roots could also occur [462].

Simultaneous determination and pharmacokinetic study of six *Aconitum* alkaloids, aconitine, mesaconitine, hyaconitine, benzoylaconine, benzoylmesaconine and benzoylhyacoinine in rat plasma using ultra-fast LC-MS/MS with LOQ of 0.025, 0.025, 0.050, 0.025, 0.025, and

0.100 ng/mL, respectively as reported by Liu *et al.* [463]. Bi *et al.* reported the studies on metabolites and metabolic pathways of bulleyaconitine A (BLA) [464] and mesaconitine [465] in rat liver microsomes. CYP3A and 2C were found to be the principal CYP isoforms contributing to the metabolism of BLA [500] while mesaconitine was mainly metabolized by CYP3A [501]. In studies of *in vivo* and *in vitro* metabolites for the main diester diterpenoid alkaloids (DDAs), such as aconitine, mesaconitine and hyaenacitine; and monoester diterpenoid alkaloids, the ester hydrolysis product of DDAs in *Aconitum* species were reported by Zhang *et al.* [466]. In conclusion, cytochrome P450 enzymes, carboxylesterases, and enzymes produced by intestinal bacteria are mainly involved in DDA metabolism in both the gastrointestinal tract and liver after oral administration

Besides *Aconitum*, black henbane or *Hyoscyamus niger*, which contain some alkaloids such as hyoscyamine, atropine, tropane and scopolamine could also produce toxic effect. Alizadeh *et al.* has reviewed black henbane and its toxicity [467].

LC-MS/MS methods have been developed by various groups for the determination and application in pharmacokinetic studies of various TCM. Miltirone is an active tanshinone compound isolated from *Salvia miltiorrhiza* Bunge (Danshen), a well-known TCM herb. Guo *et al.* reported a simple and sensitive LC-MS/MS method with LOQ of 0.5 ng/mL for the determination of miltirone in rat plasma and has applied to a pharmacokinetic study and found that it was poorly absorbed with an absolute bioavailability of approximately 3.4% [468]. The quantification of hydroxysafflor yellow A (HYSYA) (an active component of Honghua, which widely used in TCM to treat coronary heart disease, hypertension, and cerebrovascular disease) by LC-MS/MS in human urine was reported by Li *et al.* [469]. The results suggested that urine was the main excretion way of HSYA in healthy volunteers.

Scopoletin (an extract in *Dinggongteng* (*Caulis Erycibes*)) is used to treat rheumatoid arthritis, hemiplegia, swelling and pain). A method using LC-MS/MS was developed by Zeng *et al.* for determination of scopoletin and applied to the pharmacokinetic research of scopoletin in rats [470]. The result showed that oral bioavailability with a dose of 5-20 mg/kg was in the range of 5.65-6.62%.

The pharmacokinetic study of astragaloside III (a compound isolated from *Radix Astragali*, which has been used in the prevention and treatment of various diseases such as nephritis, diabetes, cancer) in rat plasma as reported by Zhai *et al.* indicated that the oral absolute bioavailability of astragaloside III was calculated to be $4.15 \pm 0.67\%$ with an elimination half-life value of 2.13 ± 0.11 hours [471]. Similar studies on salvianolic acid C in Danshen by Zhao *et al.* gave absolute oral bioavailability of salvianolic acid C was $0.29 \pm 0.05\%$ [472].

Other studies on cucurbitacin B (one of the most abundant forms of cucurbitacins isolated from various plant families) by Zhao *et al.* [473] and parishin (an active component in *Gastrodia elata blume* used for treatment of convulsion, vertigo, paralysis, epilepsy, tetanus, asthma and immune dysfunctions) by Tang *et al.* [474], were also reported.

Gai *et al.* reported the identification of eighteen prototype compounds and four metabolites in rat urine after oral administration of YiGan San, a traditional formula for the treatment of insomnia and irritability in children, using LC-Q-TOF-MS. Pharmacokinetics study of four prototype compounds identified, namely senkyunolide I, ajmalicine, rhynchophylline and isocorynoxine, using LC-MS/MS was also performed [475].

Zhi-Zi-Da-Huang decoction is a TCM formula to treat or alleviate the symptoms of alcoholic jaundice, alcoholic liver disease, and acute hepatitis [476]. Zhu *et al.* used UPLC-Q-TOF-

MS to identify 21 prototype compounds and 22 metabolites in rat plasma after oral administration of Zhi-Zi-Da-Huang decoction [477].

Huan-Nao-Yi-Cong-Fang is a herbal medicine that found to have potential in treating Alzheimer's disease. Wang *et al.* has studied the metabolism pathway of the bioactive components in the Huan-Nao-Yi-Cong-Fang formula in rat plasma after oral administration. A total of five parent active compounds and 10 metabolites were identified from the rat plasma sample using LC-MS-IT-TOF [478].

Fangji Huangqi Tang is a TCM that pharmacological studies indicated to have clinical effects on rheumatic and kidney diseases, insufficiency of the spleen and could reinforce the immune system. Metabolites in rat serum after oral administration of Fangji Huangqi Tang were identified by Liu *et al.* [479].

3.4.3 Chemical Warfare Agents

Sulfur mustard, also known as "King of toxic agents", is a chemical warfare agent belonging to the class of vesicants that cause severe damage to the skin, eyes, and respiratory system and can be genotoxic. A simplified method for the quantification of sulfur mustard adducts to blood proteins, a bio-marker of sulfur mustard exposure in human serum, plasma, or whole blood using isotope dilution UHPLC-MS/MS was reported by Pantazides *et al.* [480].

Methods for quantitation of sulfur mustard-albumin adducts using LC-MS/MS by Andacht *et al.* [481]; using hybriide quadrupole TOF-MS after direct plasma proteolysis by John *et al.* [482]; using UHPLC-MS/MS and with 2-chloroethyl ethylsulfide as internal standard (IS) by Liu *et al.* [483]; using LC-ESI-MS/MS to detect sulfur mustard-albumin adduct and investigated its *cis/trans* isomerism by Gandor *et al.* were also reported [484]. A quantitative analysis of -lyase metabolite of sulfur mustard adducts with glutathione in urine using GC-MS/MS was reported by Lin *et al.* The metabolites 1,1-sulfonylbis[2-(methylthio)ethane] (SBMTE) was determined in domestic rabbits exposed to neat liquid sulfur mustard at three dosage level. The SBMTE was detected on day 21 with mean concentration of 0.07 ng/mL and 0.02 ng/mL for middle (5 mg/kg, 0.05 LD₅₀) and low (2 mg/kg, 0.02 LD₅₀) dosage group, and on day 28 with mean concentration of 0.32 ng/mL for high dosage group (15 mg/kg, 0.15 LD₅₀) [485].

Monitoring of urinary metabolites, thiodiglycol (TDG) and thiodiglycol sulfoxide (TDGO), using isotope-dilution negative-ion chemical ionization (NICI) GC-MS in domestic rabbit after exposure to sulfur mustard was reported by Nie *et al.* [486]. The results revealed that the concentrations of TDG and TDG + TDGO in the urine increased quickly and then decreased rapidly in the first two days after sulfur mustard exposure. It is also concluded that TDG and TDGO in urine existed mainly in free form which can be used as a diagnostic sulfur mustard exposure indicator. Rodlin *et al.* reported a "dilute-and-shoot" LC-MS/MS method for the detection of metabolite of sulfur mustard, sarin, soman, VX and Russian VX in urine fortified with the metabolite standards. The LOD of the method was found to be 0.5 ng/mL for each analyte [487].

2-chlorovinylchloroarsine (Lewisite) is an organo-arsenical vesicant. Naseri *et al.* developed a method for the determination of Lewisite main metabolite biomarker, 2-chlorovinylarsonous acid, in urine by using dispersive derivatization liquid-liquid microextraction (DDLLME) method followed by GC-EI-MS [488].

Organophosphorous nerve agents (OPNAs) such as VX and GB (sarin), can form adduct with acetylcholinesterase, butyrylcholinesterase (BChE), human serum albumin (HSA) or other proteins [489,490,491,492]. Crow *et al.* reported a method for the simultaneous

measurement of seven OPNAs, including tabun (GA), GB, soman (GD), cyclosarin (GF), VR (Russian VX), VX, and VM adducts to tyrosine (Tyr) in serum, plasma and lysed whole blood samples by the use of isotope dilution reversed-phase UHPLC-MS/MS with gradient elution in less than 2 minutes with the use of 50 L of blood products is and the limit of detection was in the range of 0.018-0.097 ng/mL [489]. Pantazides *et al.* had developed a method to determine G- and V- series OPNA adducts to BChE using immunomagnetic separation (IMS) coupled with LC-MS/MS. GB-BChE adduct and VX-BChE adduct were chosen to demonstrate the method. The LODs of unadducted BChE, GB-BChE, and VX-BChE were 1.42, 0.79 and 0.43 ng/mL, respectively. Specimen stability experiments showed that plasma, serum, and blood samples stored at 37°C or colder for 2 weeks are stable, but whole blood samples need to be filtered if frozen [490]. The stability of GB and VX in dried blood, serum and plasma spots were checked for BChE activity using a modified Ellman assay and confirmed the results by quantification of the BChE adducts using LC-MS/MS as described above [490]. BChE activity at centre, halfway and edge of the spots were measured and the activity at the edge of plasma and serum spots was found to be significantly higher. The spotted samples were up to 10 times more resistant to degradation compared to unspotted control samples when measuring BChE inhibition by the nerve agents GB and VX [491].

Determination of OPNA metabolites could also demonstrate exposure of OPNAs. Hamelin *et al.* has reported a quantitative method for metabolites of five OPNAs, including VX, VR, GB, GD and GF, in serum by SPE following isotopic dilution hydrophilic interaction LC-MS/MS where the LODs were in the range of 0.3–0.5 ng/mL [492]. Lin *et al.* also developed and validated a method for the determination of ethyl methylphosphonic acid (EMPA, metabolite of VX), isopropyl methylphosphonic acid (IMPA, metabolite of GB), isobutyl methylphosphonic acid (iBuMPA, metabolite of RVX), and pinacolyl methylphosphonic acid (PMPA, metabolite of GD) in human urine by isotope-dilution GC-MS/MS after pentafluorobenzyl bromide solid phase supported derivatization. The LODs were 0.02 ng/mL for all four OPNA metabolites [493].

Ricin is a high toxic toxin that can be easily extracted from seeds of *Ricinus communis* plants. It is classified as a category B agent by the Centers for Disease Control and Prevention (CDC) in the USA and a Schedule 1 agent of the Chemical Weapons Convention by the Organisation for the Prohibition of Chemical Weapons (OPCW). Chen *et al.* has developed and validated an ELISA kit for detection of ricin toxins in human whole blood and human faeces, and concluded that faeces was the most suitable clinical specimen for diagnosis of ricin poisoning via the oral route [494]. A quantification method for which ricin was extracted and enriched from serum by immunocapture using anti-ricin monoclonal antibody 3D74 linked to magnetic beads, then digested by trypsin, and analyzed by LC-ESI-MS/MS. Combined with immunocapture enrichment, the method provided a LOD and LOQ of about 2.5 ng/mL and 5 ng/mL of ricin in serum, respectively. The method was used to determine the ricin in rat serum after administration of ricin at dosage level of 50 g/kg, and the residual ricin the rat's serum can be detected until 12 hours later at a concentration of about 10 ng/mL [495].

3.4.4 Doping Control

The current anti-doping control analysis becomes more and more challenging, as more and more compounds become available, in particular, the next generations of doping agents, those that are recombinant forms of endogenous compounds, will require new tools, such as the detection of gene transfer vectors, a lateral flow assay for erythropoietin, and a DNA-based approach to identifying blood transfusions [496]. A review by Nicoli *et al.* highlighted the analytical advances in doping control analysis achieved in the past few years, from small molecules to peptide and protein analyses, mostly based on GC-MS and LC-MS techniques

[497]. In addition to chromatographic-based techniques, the advancement in MS instrumentation and MS-based methodologies offered enormous opportunities for detection and confirmation of peptides and proteins, equipped laboratories with very powerful and robust tools to catch doped athletes. The opportunities of MS for peptide and protein analysis, including their amino acid sequence characteristics and current MS-based detection strategies, in veterinary control and sports-doping control with a particular focus on detection of illicit growth promotion were reviewed by Broek *et al.* [498].

Determination of human insulin and its analogues in human blood using liquid chromatography coupled to ion mobility mass spectrometry (LC-IM-MS) with LOD of 0.2 ng/mL and LOQ of 0.5 ng/mL for human insulin was reported by Thomas *et al.* [499]. The author also reported a simplified method for the screening for peptides <2 kDa by direct urine injection followed by LC-IM-MS analysis with LODs between 50 and 500 pg/mL. The method was evaluated by applying to the elimination study of urine sample from having administered GHRP-6, GHRP-2, or LHRH [500]. Determination of growth hormone releasing peptide metabolites in human urine after nasal administration of GHRP-1, GHRP-2, GHRP-6, Hexarelin, and Ipamorelin was reported by Semenistaya *et al.* The analysis was processed by SPE on weak cation exchange cartridges and analyzed by means of nano-LC-HRMS [501].

Blood doping is to increase the red blood cell volume. It could be done by blood transfusion (homologous blood doping), or the use of recombinant human erythropoietin (rhEPO). Pottgiesser *et al.* has reviewed the current strategies of blood doping detection [502]. Direct detection such as isoelectric focusing can identify erythropoietic stimulants, homologous blood transfusion is identified through mismatches in minor blood group antigens by flow cytometry. Indirect methods such as the ABP are the only means to detect autologous transfusion and may also be used for the detection of erythropoietic stimulants or homologous transfusion. New techniques to unmask blood doping include the use of high-throughput 'omics' technologies (proteomics/metabolomics) and the combination of different biomarkers with the help of mathematical approaches. Manokhina *et al.* proposed the detection of homologous blood doping by high-resolution qPCR-based genotyping and demonstrated that assays could be developed that would detect second populations of cells even if the "donor" blood was depleted of 99 % of the DNA-containing leukocytes [503]. Leuenberger *et al.* also reviewed and described the potential and challenges of circulating microRNAs (miRNAs) as a new class of biomarker for detection of doping substances [504]. Longitudinal measurement of circulating miRNAs in the context of the ABP should be a suitable way of using these new biomarkers in anti-doping. The author was of the view that circulating miRNA research is still in its early stages, and further work must be done to characterise the potential confounding factors affecting these biomarkers.

Dietary supplement is popular among athletes, a sport dietary supplement "CRAZE" was found to contain a banned designer doping agent, 2-ethylamino-1-phenylbutane (EAPB) which is a MA analog. During 3 months period from May to July 2013, EAPB and 2-amino-1-phenylbutane (APB) were identified in 42 urine samples from various areas in the US [505]. Wójtowicz *et al.* performed the determination of EAPB in dietary supplement, CRAZE and DETONATE, by GC-MS. Determination of the two designer agents were also performed in urine samples of one male and two female volunteers after administration of three supplements and APB found to be a metabolite of EAPB [506].

β -methylphenethylamine (BMPEA), a positional isomer of amphetamine which is classified as a doping agent by World Anti-Doping Agency (WADA), is another novel additive that could be found in nutritional supplements. Chołbiński *et al.* reported the determination method of BMPEA in urine by UPLC-MS/MS after a two-stop LLE with LOD of 10 ng/mL. Amphetamine

and BMPEA are positional isomers with comparable CID spectra, indicating that insufficient chromatographic separation may result in misidentification. The method was applied to eight doping samples. Investigation of BMPEA metabolism was also performed [507].

Xenon became a banned substance categorized as hypoxia-inducible factor (HIF) activator on the Prohibited List of WADA on September 2014. Thevis *et al.* reported the determination of xenon in human plasma and blood by GC-HRMS and GC-MS/MS [508] and in urine by GC-MS/MS [509] with LOD down to approximately 0.5 nmol/mL. Authentic plasma and blood samples collected pre-, intra-, and post-operative (4, 8, and 24 hours) were positively analyzed after storage for up to 30 hours while patients' urine specimens returned 'xenon-positive' test results up to 40 hours post xenon-based anesthesia.

Triamcinolone acetonide (TA), a potent glucocorticosteroid as anti-inflammatory drug, is included in the list of prohibited substance by WADA, but is allowed for therapeutic purpose using administration routes other than oral, intravenous, intramuscular or rectal routes. An arbitrary reporting level of 30 ng/mL for glucocorticosteroids and their metabolites was defined by WADA. Matabosch *et al.* had evaluated the detection of TA and its metabolites from urine samples of healthy volunteers administered TA by different routes. Results indicated that the current reporting level of 30 ng/mL of TA is not suitable to detect forbidden intramuscular (IM) use of TA and suggested that the best criterion to detect IM administration is the use of a reporting level of 5 ng/mL for TA [510].

Determination of eight anabolic steroid esters (nandrolone phenylpropionate, trenbolone enanthate, testosterone acetate, testosterone cypionate, testosterone isocaproate, testosterone phenylpropionate, testosterone decanoate and testosterone undecanoate) and nandrolone in DBS was reported by Tretzel *et al.* [342]. The detection of the intact esters allows an unequivocal proof of the administration of conjugates of exogenous testosterone and its derivatives. The author also reported the determination of synthetic human adrenocorticotrophic hormone tetracosactide hexaacetate (Synacthen®) in DBS, the target compound was found to be stable for at least 10 days [511]. Studies on the *in vivo* metabolism and excretion of NAD⁺ depending enzyme SIRT1 in rat urine, DBS and plasma were also reported by S. Höppner [512].

3.5 Alternative specimens

Postmortem specimens that are subjected to toxicological examinations range from bodily fluids to tissues, generally focusing on blood and urine. However, there are limitations in using blood and urine for postmortem toxicology analysis. Findings in urine may only reflect recent exposure with limited postmortem interpretation value. Whereas there are other issues in blood analysis in many postmortem analyses, such as limited blood available in cases of traumatic injury, contamination from ruptured stomach contents, or post-mortem redistribution comprising their interpretation value. Therefore, there had been continuous efforts in exploring the use of alternative specimens in postmortem toxicology, usually by evaluating postmortem drugs distribution and correlation of drug levels in various biological specimens [513, 514]. In parallel, development in analytical methods for drugs in different matrices also supported the use of the alternative specimens in toxicology [361, 439, 515, 516, 517].

3.5.1 Brain

The protected and secluded location of the brain rendered it less prone to postmortem redistribution and hence a beneficial substitute of blood. The potential use of drug levels in brain for postmortem interpretation was studied by comparing the concentrations of 30 drugs and metabolites in brain and those in blood [518]. It is found that a high volume of distribution (V_d), high postmortem redistribution is expected. This would result in a low heart

blood to brain concentration ratio (HB/Br) such as chlorpheniramine (Vd 3–6 L/kg, HB/Br 0.11), dextromethorphan (Vd 3.0 L/kg, HB/Br 0.23), meperidine (Vd 3.7–4.2 L/kg, HB/Br 0.25) [518]. The work gave a positive correlation of blood to brain drug concentrations, suggesting brain being a useful postmortem toxicological specimen [518]. A study of postmortem quetiapine reference concentration in brain and blood of 36 case with cause of death unrelated to quetiapine and 5 cases with quetiapine as a contributing factor to death also showed a positive correlation between blood and brain concentrations, on average the drug brain concentrations were about four times the drug blood concentrations. It also summarized the quetiapine brain levels in 23 cases of death with non-toxic quetiapine blood level, which can serve as a reference for evaluating postmortem cases [519].

The understanding of drug distribution over the total brain is important for interpretation of drug level in brain. A study of distribution of venlafaxine (VEN) and its metabolite concluded that level of VEN and its metabolite are differentially distributed in the brain, suggesting that the metabolism may be different across the brain regions. In this study, there is also a significant and direct correlation of VEN levels between femoral blood and cerebellum, but no relationship with temporal cortex, occipital cortex VEN levels [520].

The analytical procedure for quantification of drugs in brain could be challenging, such as tedious extraction and the lack of certified reference material for validation. An automated SPE followed by GC-MS analysis screening procedure was validated using pig brain fortified with target drugs (benzoylecgonine (deuterated), cocaine, codeine, morphine, diazepam, doxepin, methadone, phenobarbitone and ibuprofen) and quantification was done separately by standard addition or by adding isotope-labeled standards [521]. Quantitative analysis of lorcaserin in brain tissues using UPLC-MS/MS was validated and application of the method in a pharmacokinetic study showed that the average brain tissue concentration after oral administration (2 mg/kg) of lorcaserin at 1 and 2 hours were 2,427.69 and 1,671.94 ng/g, respectively [522]. The detection of JWH-250, CP-47,497 and CP-47,497-C8, in mouse whole brain tissue was achievable by liquid-liquid extraction followed by LC-MS/MS analysis with LOQs of 20 ng/g [523]. Validation experiments revealed degradation of CP-47, 497 and CP-47,497-C8 at different temperatures, and significant ion suppression was produced in brain for all compounds tested. The study demonstrated that synthetic cannabinoids are present in the brain, bolstering support that cannabimimetic effects result from a CNS-mediated mechanism of action.

3.5.2 Other Organs – Lung, Kidney & Liver Tissues

Currently, street cocaine or heroin preparations are adulterated with pharmacologically active substances for instances, stimulant, analgesics, local anaesthetics and calcium channel blocker etc. In cases of fatal intoxication, the cause of death is often attributed to the drug itself, but the presence of adulterants or cutting agents of drugs of abuse could potentially lead to additional toxic side effects. The adulterants levels in the lung tissue and blood of 11 cases for typically used adulterants in cocaine preparations were investigated. It was observed that concentrations of lidocaine, hydroxyzine and levamisole, which are common adulterants of cocaine, are higher in the lung tissue than in heart blood and femoral vein blood, suggesting potential post-mortem redistribution from the lung to heart blood and consideration should be made to analyse lung tissue for cause of death determination [524].

Forensic toxicological drug analyses are usually not performed immediately after autopsy. Other than preservation at low temperature, cases requiring analysis of drugs from tissues was preserved by formalin fixation. Uekusa *et al.* investigated the influence of formalin fixation to drug concentrations over time (1, 3, 6 and 13 months) in human tissues, liver and kidney, that were collected from drug-positive autopsy cases using GC-MS assay [525]. The levels of chlorpromazine, levomepromazine, promethazine bromazepam and milnacipran in

human liver and kidney tissues decreased following fixation. The proportions of the drugs that remained within the tissues differed between liver and kidney; in kidney, all assayed drugs exhibited reduced stability during preservation compared to levels in liver. These results suggested that analyses in formalin-fixed tissues needed to include at multiple time points for the duration of preservation with the detection of chemical degradation/denaturation products, such as S-oxides of chlorpromazine and levomepromazine included.

A review on toxicological analyses of medications and chemicals in formalin-fixed tissues and formalin solutions was reported [526]. It reviewed both experimental studies and practical cases involving embalmed bodies in two aspects: reactions of formaldehyde with substances in tissues fixed with formalin (aqueous formaldehyde solution) and the stability of substances in formalin-treated tissues and formalin solutions. It is known that formaldehyde reacts with amines to form Schiff bases, which proceeds at room temperature and the reaction increases with pH and the concentration of formaldehyde.

3.5.3 Meconium

Besides hair, urine and umbilical cord, meconium has traditionally been the 'gold standard' for new born drug testing that can detect maternal drug use for the last two trimesters of a full-term birth. There had been few publications on newborn drug testing in multiple gestations. In a study where meconium was analyzed for amphetamine, MA, barbiturates, benzodiazepines, cannabinoid metabolites, cocaine metabolites, methadone, opiates oxycodone, phencyclidine and propoxyphene by traditional two-step process including a broad immunoassay-based drug screen (ELISA), followed by confirmatory testing (GC-MS or LC-MS/MS) of specimens that screened positive, mismatched results occurred in 13% sets of twins and 10% sets of triplets; among the tested drugs, barbiturates (33%), opiates (30%) and benzodiazepines (28%) were the most common mismatches [527]. Mismatched results of meconium drug testing in multiples are uncommon, with iatrogenic medication administered (e.g. lorazepam and morphine) to one infant but not the other being a frequent cause of such mismatches.

Quantifications of THC-COOH in meconium were reported with sample clean up by liquid-liquid extraction [528] or SPE [529] followed by GC-MS analysis with labeled internal standard after trimethylsilyl derivatization. The SPE method also analysed 11-OH-THC with LODs for both analytes being 5 ng/g [529]. The mean (median) THC-COOH (n = 123) concentrations detected were 55.0 ng/g (33.75 ng/g), while the mean (median) for 11-OH-THC (n = 4) concentrations were 8.25 ng/g (6.5 ng/g) [568]. In other studies, LC-MS or LC-MS/MS was also adopted for examination of meconium for drugs such as buprenorphine [530], cocaine [531, 532, 533] and other drugs of abuse [534].

Understanding of the relation between gestational consumption of drugs and foetal exposure could give insight to the interpretation of meconium drug analysis. In an assessment of foetal exposure to gestational consumption of drugs of abuse, the findings of drugs in meconium were compared with maternal hair samples of 80 mothers. Only 40% of the samples of mother–infant dyads gave positive findings for drug detection in one or more hair shafts. The results revealed that mothers who consumed drugs throughout the whole pregnancy showed drugs of abuse in meconium samples, while sporadic and/or discontinuous consumption in pregnancy gave negative findings [534]. To provide further information for interpretation of meconium drug testing results, a study has analyzed meconium drug testing patterns in a de-identified dataset (N=76631) to investigate the patterns of drugs and their metabolites observed in meconium [535]. The positivity rate was the highest for the cannabis metabolite THC-COOH (25.2%), followed by opiates/oxycodone (23.2%), amphetamine/MA (6.7%), cocaine metabolites (5.5%), methadone (5.3%), benzodiazepines (3.4%), barbiturates (1.1%), propoxyphene (1.0%, and phencyclidine

(0.1%). Patterns observed in meconium exhibited many similarities to those patterns commonly reported with urine drug testing.

3.5.4 Skeletal Remains

Skeletal tissues can be useful for forensic toxicological investigation when no other biological specimens are available due to severe decomposition or degradation of the corpse. An experiment investigated tramadol and its metabolite levels in decomposed bone and plasma sample from rats exposed to different doses. Decomposed bone was ground and incubated in methanol prior to SPE followed by GC-MS analysis of tramadol and O-desmethyltramadol [536]. This work has demonstrated that tramadol and its metabolite may be detected in decomposed skeletal tissues. The data showed that while measurements of the levels of tramadol and O-desmethyltramadol in bone were not significantly different between exposure patterns examined (while those in plasma were), the ratio of tramadol and O-desmethyltramadol levels provided improvement in the discrimination between exposure patterns. However, the bone drug level correlate poorly with blood drug levels, rendering prediction of toxicity based on bone drug level impossible.

The influence of body position and microclimate on ketamine and metabolite distribution in decomposed bone tissue was examined by Watterson and co-workers [537]. Microclimate and body position significantly influenced observed drug levels: higher ketamine levels were observed in carcasses decomposing in direct sunlight, where reduced entomological activity led to slowed decomposition.

Bone marrow aspirate and pericardial fluid are also useful materials to be included in the forensic toxicological routine as well as to investigate pharmaco-/toxico-kinetics and postmortem redistribution [538]. Analysis using automated GC-MS following SPE detected 36 drugs in 218 cases. Most of the drugs showed overall similar distributions in right heart blood, pericardial fluid and bone marrow aspirate with some exceptions.

3.5.5 Vitreous Humor

Vitreous humour (VH) is the aqueous gel located in the posterior cavity of the eye between the lens and retina that contains less protein than urine and blood. It is a favourable specimen due to its accessibility, low contamination and high stability despite of its small volume which greatly limits the number of tests. A comprehensive review discussed the distribution of xenobiotics in VH samples and summarized the concentrations of 106 drugs in VH and blood from more than 300 case reports, which may serve as a practical tool for toxicologist in analyzing and interpreting results [539]. Review of various controlled animal or autopsy studies on the implication of a xenobiotic in a victim's death by interpreting only VH concentrations showed that VH and blood concentrations do not correlate for all compounds, and that in others, the scatter of the autopsy data usually precludes extrapolation to blood concentrations without significant error. Simultaneous screening of cocaine, amphetamines, opiates, cannabinoids and their metabolites in VH was achieved using SPE followed by GC-MS analysis. Except for cannabinoids, which binds strongly to plasma protein and thus less readily diffuse into VH, all samples screened positive for the aforementioned drugs in the blood also gave positive results in VH [540]. Another simultaneous identification and quantification of 24 analytes for forensic relevance in vitreous humor, as well as blood and plasma, were also reported using ESI-LC-MS/MS with a mean recovery of drugs in vitreous humor at about 85% [541].

A study correlating the bile and VH concentration with blood drug concentration using 6 drugs (meprobamate, morphine, cyamemazine, caffeine, diazepam, and citalopram) showed a significant correlation between blood and VH concentration in rabbits. However, no such correlations in the autopsy series were found for cyamemazine or diazepam [514].

Molecules with higher molecular mass can also be identified from VH. Acute mast cell activation occurs in a number of pathologic conditions and is commonly observed in patients with allergic reactions; level of β -tryptase measured with a fluoroenzyme immunoassay can reflect such activation and can serve as diagnostic markers [542]. The findings suggested that elevated pericardial fluid β -tryptase levels in the early postmortem period can be the consequence of β -tryptase elevation in systemic circulation and might be used for diagnostic purposes should postmortem serum prove unavailable during autopsy.

3.5.6 Bile

As bile/blood concentration ratios are high for numerous molecules or metabolites, bile is a matrix of choice for screening when blood concentrations are low or non-detectable: *e.g.* cases of weak exposure or long intake-to-death interval. A review covering literatures from 1970 to May 2015 has provided an update on biliary physiology and xenobiotic excretion for forensic purposes. In addition to discussion on sampling, storage and analysis, it summarized the levels of 133 analytes in bile and blood from more than 200 case reports with the cause of death [543]. Quantitative applications have been little investigated, but small molecules (*e.g.* ethanol, meprobamate, cyamemazine and, to a lesser degree, nitrobenzodiazepines) with low bile/blood concentration ratios seem to be good candidates for quantitative bile-based interpretation. In a study correlating the bile and vitreous humor concentration with blood drug concentration of using 6 drugs of forensic interests gave a correlations between bile and blood concentration of meprobamate and caffeine in both rabbits and human, suggesting bile can be interpreted quantitatively for certain molecules [514].

Certain drugs are more readily absorbed into the bile; an autopsy case showed that the highest concentration of 7-aminoflunitrazepam, the metabolite of flunitrazepam, was observed in the bile, among the body fluids and solid tissues examined for the same target analyte by LC-MS/MS. Although a majority of flunitrazepam was converted to 7-aminoflunitrazepam, the flunitrazepam concentration was highest in the pancreas, followed by the spleen, bile, left heart blood, and brain. The study showed that bile maybe a useful specimen for the detection of unchanged benzodiazepines/their metabolites to be collected at autopsy [317].

3.5.7 Oral Fluid (OF)

OF offers non-invasive and directly observable sample collection reducing adulteration potential, ease of multiple sample collection *etc.* By studying the linear correlation of concentrations and calculating the OF to blood concentration ratios (OF/B), the relationship of drug concentrations, namely for amphetamines, cocaine, THC and opioids, between OF and whole blood was established. In a study by Langel *et al.* a series of median OF/B ratios, for amphetamines 19-22, for opioids 1.8-11, for cocaine and metabolites 1.7-17, for THC 14, for benzodiazepines 0.035-0.33, and for other psychoactive medicines 0.24-3.7 were obtained. As a large variation was observed, drug findings in OF should not be used to estimate the corresponding concentrations in whole blood. However, detection of drugs in OF could be a sign of recent drug use [78].

Cannabis is the world's most commonly used drug of abuse, and the detection times in different matrices including OF had been extensively studied. One study investigated the presence of THC in 26 patients admitted to a closed detoxification; cannabis intake is detected in OF as THC, and a positive finding is considered to be a result of recent smoking, *i.e.* up to 8 days after admission [544], whereas THC concentrations was found to be >1000 $\mu\text{g/mL}$ shortly after smoking [117]. Owing to a stronger correlation than urine with blood concentrations, screening based on OF is greatly gaining value in DUID. After centrifugation

and dilution, sensitive chromatographic methods such as LC-MS/MS, LC-ESI-MS/MS, UHPLC-MS/MS or GC-MS/MS [545] are the preferred techniques for such screening. The LOD was found to be 0.01 ng/mL and 0.1 ng/mL for THC and THC-COOH, respectively [310]. Conveniently, a quantitative dilute and shoot LC-MS method has been described to determine codeine, morphine, hydrocodone, hydromorphone, norhydrocodone, oxycodone, noroxycodone and oxycodone whereby samples were diluted 10-fold in methanol/water followed by analysis with LC-MS/MS system. This is ideal for high throughput laboratories as no sample clean-up or concentration is required as well as a fast LC gradient of 2.2 min with the use 100 µL of sample and the calibration ranges are 2.5-1,000 ng/mL [311].

In a study where 39 new amphetamine-designer drugs were evaluated using two different multi-drugs OF screen devices, cross-reactivity of <1 towards most drugs analyzed in assays targeting amphetamine or MA was observed; only two (*p*-methoxyamphetamine and *p*-methoxymethamphetamine) gave positive results [546]. An EMIT® screening method used in urine assay to detect 6-MAM was validated to qualitatively screen for 6-MAM in OF. The LOD was determined to be 1.844 ng/mL and the sensitivity, specificity and overall misclassification rate were found to be 90, 100 and 6%, respectively [547].

A recent study by Ma *et al.* showed that iridium (III) complex exhibited excellent selectivity towards intermolecular G-quadruplex motif, which can act as a label-free switch-on cocaine detection platform with a detection limit as low as 30 nM. The detection is achieved by measuring the linear relationship between luminescence intensity and cocaine concentration established from 30 to 300 nM. Furthermore, this sensing approach could detect cocaine in diluted OF [548].

With the increasing usage of new non-regulated synthetic designer drugs, these have become a new interest for research as these drugs are not usually included in routine toxicological analysis. 10 synthetic cathinones can be quantitatively measured by purifying the sample by SPE prior to UHPLC-MS/MS [549]. In a cross reactivity study with an on-site immunoassay device for detection of synthetic cathinones, a LC-MS/MS method for detection of selected synthetic cathinones and piperazines (1-(3-chlorophenyl)piperazine and 3-trifluoromethylphenylpiperazine) was developed. The LODs and LOQs in OF were in the ranges of 0.025-0.1 ng/mL and 0.2-0.5 ng/mL respectively. As synthetic cathinones are structurally related to amphetamines and derivatives, the study showed that all analytes in OF produced a MA positive result using an on-site immunoassay device at high concentrations (100 or 10 µg/mL) [550]. The first quantitative analytical examination of XLR11 and UR-144 (and their pyrolysis products and metabolites) in human OF samples was established [551]. The developed method that purified the sample with SPE and then quantitatively identifies the drugs with LC-MS/MS has LLOQs of 5.0 ng/mL for all of the analytes. Human subject data showed that OF can be used to detect the parent drug, the metabolite and the pyrolysis product of of UR-144, however, in the case of XLR11, the metabolite was absent in the OF.

With respect to screening of benzodiazepines, for example alprazolam, clonazepam, diazepam and nordiazepam, Verstraete and co-workers showed that the concentration of the 4 studied benzodiazepines in OF can neither be used to accurately estimate the concentrations in blood nor to correctly identify patients with blood drug concentrations below or above recommended therapeutic levels. When using analytical methods with LOQs corresponding to concentrations less than 0.5 ng/mL in undiluted OF, it may be used to confirm a recent intake of benzodiazepines [552].

3.5.8 Hair & Nail

Hair testing is a convenient, temper resistant and non-invasive technique for the analysis of many controlled drugs and drugs of abuse as compared to blood tests and urinalysis. EtG is a direct marker of ethanol consumption, and its assay in hair is an efficient tool for chronic alcoholism diagnosis. To achieve the recommended LOQ of less than 3 pg/mg as proposed by the Society of Hair Testing (SoHT), an improved LC-MS/MS method was developed with concentration range of 3-1000 pg/mg that has been applied a forensic post-mortem case and two cases of suspension of driving licences [553]. GC-MS/MS operated in the negative ion chemical ionisation (NICI) was developed for the determination of EtG in hair with LOD and LOQ of 0.05 and 0.2 pg/mg hair, respectively, which is more sensitive when compared to an LOD of 0.5 pg/mg hair and LOQ of 1.5 pg/mg hair obtained with a previously validated GC-NICI-MS. [554]. While comparison of results of hair samples (n=58) obtained by GC-NICI-MS and GC-NICI-MS/MS showed no significant difference between both methods, the GC-NICI-MS/MS method should preferentially be used, especially when differentiating between teetotalers and moderate drinkers according to the cut-off (*i.e.* 7 pg/mg hair). Hair can be used for monitoring alcohol consumption by examining the level of EtG or fatty acid ethyl ester (FAEE). For the alcohol abstinence assessment, a cutoff level of EtG in hair would be useful. A study has adopted a highly sensitive analytical protocol with detection down to 0.5 pg/mg for determination of cut-off level based on basal EtG level in hair. Head hair samples from 44 certain abstainers and teetotalers allows to tentatively estimate basal EtG concentrations in hair were found to be around 0.8 ± 0.4 pg/mg, and that cut-off value for EtG in the range of 1.0-2.0 pg/mg was proposed to support alcohol abstinence [555]. According to the current SoHT consensus, the cut-offs for FAEEs of 0.2 and 0.4 ng/mg for abstinence assessment, and 0.5 and 1.0 ng/mg for chronic excessive alcohol consumption were used for analysis of proximal 0-3 or 0-6 cm hair segment respectively. Both sets of cut-offs should be congruent (ratio 0.50) for uniform interpretation. To study the effect of the analyzed hair length on FAEE concentrations in hair, Pragst and co-worker determined FAEE in parallel for the 0-3 and the 0-6 cm segment of 157 hair samples and gave concentration ratio between 0-3 and the 0-6 cm segment ranged from 0.3 to 1.5 (mean 0.83, median 0.82) [556]. The study showed that the current cut-offs (ratio 0.50) are not congruent, and that it should be considered in a future consensus to redefine the hair length for marker on 0-3 cm. Bayesian theory was proposed to be used for assessment of alcohol consumption, in which likelihood ratios (LRs) as the strength of value of the evidence can be deduced from of the both concentration of EtG and FAEE in hair. It allows for the quantification of the strength of the evidence and the factors that affect its value, false-positives and false-negatives are expressed in a more scientific way [557].

It has been shown that the use of alcohol-containing hair products or EtG containing herbal lotions could lead to false positive results in a chronic excessive alcohol drinking study [558]. Of the 11 hair tonics from 8 manufacturers tested, EtG ranged between 0.07 and 1.06 mg/L was detected in 7 products from 4 manufacturers. Ethyl sulphate (EtS) was found in 3 out of the 11 hair tonics [559]. In a study to investigate the influence of repeated bleaching and permanent coloring on EtG concentrations upon GC-NICI-MS analysis of hair samples from alcohol-dependent patients, hair bleaching and permanent coloring was found to reduce EtG concentrations by $65 \pm 24\%$ and $82 \pm 11\%$, respectively [560]. The impact of increasing exposure to hydrogen peroxide on the EtG content of hair was investigated. Using attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) to study the formation of cysteic acid, the oxidative product of cysteine, which is indicated by an increase in absorbance at 1040 cm^{-1} [561]. The study showed that ATR-FTIR is capable of detecting an exposure to hydrogen peroxide when still no brightening was visible and already before the EtG content deteriorated significantly. Another study proved that single use of 'special shampoo', which promises to 'detox' hair after single application to easily pass any drug or alcohol test, does not remove EtG from hair [562]. A study by Kintz and coworkers analysed hair of 97 cases for both EtG and FAEE to evaluate both tests in documenting chronic

excessive alcohol consumption, particularly when the results are in contradiction. Of the 97 cases analysed, 27 (27.8%) results were EtG negative and FAEE positive, when applying the SoHT cut-offs, probably due to the use of alcohol-containing hair products. Four cases (4.1%) were EtG positive and FAEE negative that were attributed to the use of herbal lotions containing EtG [558].

Hair is most frequently used in the screening of drugs of toxicological interest such as abused drugs and prescription drugs, as this could facilitate the monitoring of abstinence, DFC or DUID. Shah *et al.* studied an LC-MS/MS screening method for >200 drugs/metabolites in forensics and doping. The LODs of compounds ranged between 0.05-0.5 pg/mg of hair [563]. In a study on the assessment of both the analytical and pre-analytical sampling variations in segmental hair analysis, another UPLC-MS/MS method was developed for the analysis of 31 common drugs in hair. For the most frequently detected analytes, the pre-analytical variation was 3-7 folds larger than the analytical variation (7-13%) and hence the dominant component in the total variation (29-70%) [564]. The effect of colouring or bleaching hair sample on the detection of drugs was investigated by Agius; no significant difference in the positivity rate was observed between treated and non-treated hair when 9488 non-treated and 1026 cosmetically treated hair (dyed or bleached) was screened against a wide range of common drugs of abuse or EtG [123]. Mönch *et al.* introduced a method which involved simultaneous milling and micropulverized extraction for the pre-treatment prior to a validated GC-MS procedure involving derivatization with pentafluoropropionic acid anhydride to render quantitation of EtG in hair [565]. The study showed that micropulverization yielded 28.0 ± 1.70 pg/mg and was seen to be superior to manually cutting (23.0 ± 0.83 pg/mg) and equivalent to dry grinding (27.7 ± 1.71 pg/mg) with regard to completeness of EtG extraction. Using 50 mg hair incubated in a phosphate buffer pH 5 for one night at room temperature, a multianalytes method using LC-MS/MS allowing simultaneous identification and quantification of 35 psychoactive drugs was developed with LODs and LOQs ranged from 0.5 to 10 pg/mg [142]. In another report using 30 mg hair incubated overnight under sonication, another LC-ESI-MS/MS method was used for the detection of cathinones, piperazines, amphetamine-type substances and synthetic cannabinoids with LODs varied from 2 to 20 pg/mg [120]. Short acting antipsychotic drugs, *i.e.* quetiapine and its main metabolite 7-OH quetiapine, were quantified using LC-MS/MS with time-resolved hair analysis results varied from 0.35 to 10.21 ng/mg of hair and 0.02 to 3.19 ng/mg of hair for quetiapine and its metabolite respectively [566]. An LC-MS/MS method for detection of sildenafil as well as microdenafil, tadalafil, udenafil and vardenafil in hair was developed with LODs in the range of 0.05-1 ng/10 mg hair and LOQs of 1-2.5 ng/10 mg hair [567]. The method is useful to provide information on chronic drug use.

In addition to meconium, hair of the new born (neonatal hair) and/or that from the pregnant or nursing mother (maternal hair) can be evaluated for perinatal exposure to drug of abuse, accounting for a sizable time window of active and passive exposure [568]. Data presented in the review indicated that the key points in the analysis of drugs of abuse and alcohol biomarkers are method sensitivity and specificity which has been significantly improved with the advancement of the MS technology.

With the exception of lower amount of biological material in children versus adults, there is no specific analytical problem when processing samples from children. The issue is the interpretation of the findings, with respect to different pharmacological parameters. In some very young children, the interpretation can be complicated by potential in utero exposure [569]. 24 cases with hair segmentation analysis results from children less than one year old whose mothers admitted having used drugs during pregnancy have been reviewed. The concentrations measured in the hair of children were lower than those observed in subjects using therapeutic (or illegal) drugs (methadone, tramadol, diphenhydramine, diazepam,

cannabis, heroin, amitriptyline and bromazepam). The pattern of drug distribution was similar in all these cases, low concentrations in the proximal segments and highest concentration in the distal segment (last segment). It is proposed to consider 100% in utero contribution to the final interpretation when the ratio concentration of the proximal segment to the concentration of the distal segment is lower than 0.5, applying only to child under one year old with the hair shaft length is at least 4 cm for at least 3-4 segments to observe the variation in drug concentrations. Another report describing 3 separate cases of child (14 months to 7 years) administration of prescription drugs, the drug in question (methadone, tramadol and amitriptyline and nortriptyline) was detected in more than one section of hair [570]. The cases demonstrated the added value of hair testing and emphasized the importance of using hair samples to complement conventional analyses. A court case related to a 6-month-old baby girl who was hospitalized with the presence of amitriptyline (AMI) and its metabolite nortriptyline (NOR) in blood and urine was reported [571]. The judge ordered hair tests for both the child and his parents to document drug exposure. All hair segments tested for both parents were negative. For the baby two strands of hair were collected one day after the acute intoxication for the first and 5 weeks later for the second. The high and relatively homogenous concentrations of AMI (6.65-9.69 ng/mg) and NOR (7.12-8.96 ng/mg) in the first strand of hair measured suggested that contamination could have occurred. The analysis of the second strand after decontamination allowed to detect AMI (0.54-1.41 ng/mg) and NOR (1.26-4.00 ng/mg) in all hair segments, supporting the hypothesis of a chronic exposure during several months before hair collection with regular increase.

The literature regarding the detection and quantification of drugs in nails, and the applications of nail analysis was reviewed [125]. Studies showed that most drugs of abuse and pharmaceuticals are detected in nails in pg/mg to ng/mg range. The major drawback of nail analysis is the lack of research, which complicates the understanding and interpretation of results obtained by nail analysis [125]. The ever-expanding understanding of nail physiology and the incorporation mechanisms of drugs, medicaments or poisons, combined with improvements in technology, provide the opportunity for the development of nail analysis in multiple disciplines of forensic and clinical toxicology [572]. Sensitive analytical instrumentation, mainly LC-MS/MS, allows for detection of femtogram (10^{-15} g) quantities of substances in nails. Both Madry *et al.* [573] and Chen *et al.* [574] reported a single oral dose of zolpidem could be detected in the fingernail samples in a long-term follow-up analysis. Both studies demonstrated a drop of zolpidem concentration in nail clippings from initial peak and a hump of zolpidem rebound within week 10 to 15 after administration. This initial peak of the zolpidem concentration could be explained by the sweat-mediated incorporation of zolpidem into nails. The rebound of concentration in nail clippings was originated by germinal matrix area where the drugs incorporated into the keratin. Both studies proposed the detailed profile of zolpidem incorporation into the nails. Another article from Chen *et al.* [575] also indicated that clozapine was detected in both hair and nails of a bloated immersed cadaver. This finding demonstrated an active and useful role of keratinous biological materials in the identification of history of drug use.

Shen *et al.* [576] recently developed a LC-MS/MS method for the determination of five opiates including 6-MAM, morphine (MOR), acetylcodeine, codeine, and heroin in frozen pulverized human fingernails to investigate the correlation between the concentration of opiates in nail and hair samples from subjects whose urine specimens were positive for morphine. A total of 12 of 18 fingernail samples contained detectable 6-MAM (mean=0.43 ng/mg, range=0.10-1.37 ng/mg), MOR (mean=1.74 ng/mg, range=0.58-3.16 ng/mg) and codeine (range from < LOQ to 0.27 ng/mg). The concentrations of drugs and metabolites in hair except morphine were typically higher than those in nails, but the concentration of

morphine in nails was generally higher than that in hair. The ratio of 6-MAM/MOR was below 0.57 in all nail samples.

Nail samples is firstly reported for a general unknown screening of drug use. Krumbiegel *et al.* [440] compared the qualitative drug results of 70 postmortem hair and nail samples conducted by a LC-Q-TOF-MS system. A total of 89 analytes are identified in those samples. They included common drugs of abuse (cocaine, ketamine, and amphetamine), drugs with psychotropic effects (antidepressants, atypical antipsychotics, benzodiazepines, and anticonvulsants), drugs with antihypertensive and antiarrhythmic effects (atenolol, and bisoprolol), and, antifungal drugs and others (sedatives, analgesics, antihistamines, and antibiotics). The results showed that nails are a comparable matrix to hair for general unknown screening and strongly suggested that great variety of drugs can be detected in nails matrix.

3.6 Interpretation of Toxicological Results

Quantitation of drugs concentration in blood and subsequent interpretation of the toxicological data helps establish the cause and manner of death. Drug concentrations, mostly derived from live patients, usually serve as a useful reference. Due to variables such as post-mortem redistribution, time of survival after intoxication, drug stabilities or presence of other drugs or disease would introduce complications in the interpretation. Therapeutic drug concentrations measured in plasma are of limited value as reference intervals for interpretation in post-mortem (PM) toxicology. An extensive study of PM drug concentrations in femoral venous blood from 57,903 Finnish autopsy cases was reported [577]. Sixty-one (47%) of the 129 drugs studied showed a PM blood/plasma relationship of 1. For 22 drugs (17%), the relationship was <1, and for 46 drugs (35%), the relationship was >1. No marked correlation was found between the PM blood/plasma relationship and the volume of distribution. These drug concentration distributions based on a large database provide a helpful reference in interpreting drug concentrations in PM cases. Langford *et al.* developed a Bayesian network framework to assign a likelihood of fatality based on the contribution of drug concentrations as well as the pathological findings [578]. The author suggested that the Bayesian network could be used as complementary approach to assist in the interpretation of PM drug concentrations.

3.6.1 Post-Mortem Redistribution (PMR)

It is well-known that post-mortem redistribution (PMR) changes in drug concentration can produce interpretation difficulties, but much is still unknown. The continuous efforts in enriching our understanding in PMR in the past three years were discussed below.

3.6.1.1 Evaluation of PMR

The phenomenon of PMR remains poorly understood due to all possible biological processes involved in the body after death before samples are obtained. The sought of a concept that can provide a definitive and authoritative ranking to possibly assist numerical interpretation of PMR is highly desirable. Several approaches have been attempted to evaluate the extent of PMR [579, 580, 581, 582].

In addition to central blood to peripheral blood ratio (C/P), a review of previously published works supported the proposed model that drugs with a liver (L) to peripheral blood (P) concentration less than 5 L/kg as being prone to little or no PMR has been proposed, while drugs with a the liver to peripheral blood (L/P) ratio greater than 20–30 L/kg have propensity for substantial PMR [579]. Many antidepressants, including both tricyclic antidepressants and selective serotonin re-uptake inhibitors, were markedly differentiated from drugs previously verified to be free from, or exhibit little, PMR. McIntyre has also introduced a

post-mortem redistribution factor (F) for a drug, which characterises the direct relationship between PM peripheral blood and the corresponding ante-mortem (AM) whole blood concentration [580]. It has been suggested that employing the factor F would provide a more definitive and authoritative drug ranking and possibly numerical interpretation of PMR for forensic toxicologists. The author has extended the concept of the factor F by further introducing a concept of a theoretical PMR factor (F_t) based upon a drug's unique L/P ratio as the only independent variable [581].

An alternative approach is to apply quantitative structure-activity relationship (QSAR) analysis for modelling PMR data of structurally related drugs. Giahinis *et al.* has employed QSAR models on two data sets (the extent of PMR of benzodiazepines and tricyclic antidepressants) as expressed by C/P ratio as a complimentary tool to provide an informative illustration of the contributing molecular, physicochemical and structural properties in PMR process [582]. Nevertheless, the complexity, non-static and time-dependent nature of PMR cast doubt on whether QSAR methodology could predict the degree of redistribution.

3.6.1.2 PMR of Drugs in Blood

Dinis-Oliveria and co-workers have examined PM cardiac and femoral blood samples from 15 cases of fatal tramadol intoxication to assess the PMR of tramadol and its metabolite *O*-desmethyltramadol [583]. Comparing the cardiac-to-femoral blood ratios of 1.40 and 1.28 for tramadol and its metabolite has led to a conclusion that femoral blood should be considered for quantitative interpretations in fatal cases of tramadol intoxication.

Peripheral PMR of morphine in femoral vessel has been explored [584]. Morphine drug concentrations were evaluated in clamped and unclamped femoral vein blood samples at 3 different times before autopsy. No significant difference between the clamped and unclamped blood concentrations at any period was observed and the authors have concluded that unclamped femoral blood samples do not show significant redistribution for morphine from central sites within first 24 hours after death in bodies kept refrigerated at 4°C.

Skoc *et al.* have determined the post-mortem femoral blood concentrations of the antipsychotic drugs (aripiprazole, chlorprothixene and its *N*-desmethylmetabolite, and quetiapine) [585]. The concentration ranges obtained in femoral blood were largely corresponded to therapeutic plasma levels observed *in vivo* suggested that no or only limited PMR for all four analytes of interest, and this is in opposition to the majority of antipsychotic drugs that tend to exhibit increase in post-mortem values.

In a fatal case of flufenoxuron a benzoylurea insecticide for controlling insects and mites by a chitin synthesis inhibitor, intoxication in a 48-year-old-man, the concentration ratio of the heart/peripheral blood of flufenoxuron was 2.0 and the ratio of gastric contents/peripheral blood was 9.4, suggesting possible PMR and there may be a massive amount of flufenoxuron orally ingested [586].

A case report published by McIntyre *et al.* has provided evidence for a lack of significant fentanyl PMR by comparing AM fentanyl concentration (1.4 ng/mL) to PM peripheral (1.6 ng/mL) and central blood fentanyl (2.2 ng/mL) concentrations [587]. An unusual case report of a suicide involving intraperitoneal injection of pentobarbital, an overdose of zolpidem and the intake of diazepam, ethanol and other psychoactive substances by Hangartner *et al.* has revealed that a significantly higher pentobarbital concentration in femoral blood compared to cardiac blood (36 vs. 15 mg/L). On the contrary, zolpidem and diazepam concentrations in cardiac blood (2700 and 590 µg/L) were found to be significantly higher than in femoral

blood (1500 and 230 $\mu\text{g/L}$). The findings pointed to a PMR with a distinct gradient from areas of high drug concentrations in gastrointestinal tract (zolpidem and diazepam) and the injection site (pentobarbital) to peripheral tissue [588]. PMR of synthetic cannabinoids MAM-2201, AM-2201, AM-2232 and their metabolites in PM plasma has also been suggested by Zaitso and co-workers in a study which revealed site differences between heart and femoral PM plasma concentrations of parent cannabinoids and some metabolites [290]. Quantitation results suggested that defluorination is a major metabolic pathway for MAM-2201, and N-dealkylation is a common but minor pathway for the naphthoylindole-type synthetic cannabinoids in human.

The potential for PMR of hydrocodone, a widely used therapeutic opioid analgesic used to manage pain, has also been investigated [589]. Hydrocodone concentrations in 39 peripheral blood, central blood and liver specimens were compared. The fact that the C/P hydrocodone ratio (1.3 ± 0.35) and L/P ratio (3.4 ± 1.7) L/kg being less than 5 L/kg indicated that hydrocodone is unlikely to undergo substantial PMR changes. Methylphenidate, a widely prescribed stimulant used for the treatment for attention-deficient/hyperactivity disorder has been shown not to subject to significant PMR, considering both the C/P ratio (0.89) and L/P ratio (3.3) as reported in a fatal case investigation of a 62-year-old woman [590]. PM femoral blood concentrations of the antipsychotic drug risperidone and the active metabolite 9-hydroxyrisperidone were determined by an LC-MS/MS method in 38 cases where cause of death might or might not be related to risperidone. The findings did not suggest risperidone was subject to major PMR [591].

Liver trazodone concentrations have been compared to peripheral and central blood ratios in 19 cases [592]. These data suggested that PM trazodone peripheral blood concentrations may be considered non-toxic to at least 1.0 mg/L, and with liver concentrations to at least 2.2 mg/kg. The L/P ratio was shown to have a median value of 2.8 L/kg and given that a L/P ratio less than 5 L/kg is consistent with little or no PMR, it was concluded that the drug is unlikely to demonstrate significant redistribution.

3.6.2 Drug Stability

Understanding the stability of drugs in biological specimens is of crucial importance in the interpretation of the toxicological findings in post-mortem forensic toxicology. It is not unusual that re-analysis of samples might be requested for forensic samples in criminal cases some years after the initial analysis and therefore, it is important to know to what extent drug concentrations would change when biological samples are stored over a period of time.

3.6.2.1 Stability of Alcohol in Blood

An examination of the effect of long-term storage on alcohol stability in post-mortem blood samples [593] has been reported, with BAC measured within 1 to 4 days after being sampled was compared with those being re-analysed after storage (ranging 191 to 468 days). It was shown that about 90% of the results (slope average) lie within 95% limits and 10% were outside but significant deviations observed in some blood samples has led to a conclusion that blood samples should be stored in a tube of suitable volume with minimal or no headspace to defer alcohol oxidation.

3.6.2.2 Stability of Benzodiazepines in Blood

The long-term stability of benzodiazepines, opioids, central stimulants and medicinal drugs in authentic PM blood samples has been studied [594]. Samples screened positive for the selection of benzodiazepines were reanalysed after storage at $-20\text{ }^{\circ}\text{C}$ for 16–18 years. Re-analysis of samples containing diazepam, nordiazepam, flunitrazepam, amphetamine,

morphine, codeine and 'acidic' medicinal drugs such as paracetamol and meprobamate showed only small changes during long-term storage, while a decline in clonazepam concentrations was observed. For many drugs, however, single samples could demonstrate marked concentration changes, both increases and decreases during storage. For 'alkaline' medicinal drugs, concentration losses were observed in most cases.

3.6.2.3 Stability of γ -hydroxybutyrate (GHB) in Blood

In a stability study of GHB in stored blood and urine specimens, GHB was added to GHB-free AM blood and urine samples (at the concentration of 5 and 10 mg/L respectively) and PM blood and urine samples (50 and 10 mg/L respectively) and all samples were stored at three different temperatures (-20, 4 and 20 °C) with no preservatives added [595]. The authors suggested that analyses of GHB both in blood and urine specimens performed within 3 days of sampling stored at -20 °C and +4 °C could avoid instability issues.

GHB was determined in whole blood by GC-FID after conversion into γ -butyrolactone and results were compared with LC-MS/MS. [596]. The study verified that GHB concentrations remain stable in authentic blood samples from living subjects when these are stored for up to 6 months at 4°C in evacuated tubes containing 1% w/v sodium fluoride preservative, and that the LC-MS/MS method tended to give higher results than the GC-FID-GBL method.

3.6.2.4 Stability of Mephedrone in Blood

Assessment of the stability of mephedrone (4-methylmethcathinone) in AM and PM blood specimens has been published [597]. The psychoactive drug which produces stimulant and empathogenic effects was added to whole blood collected from alive and dead mephedrone-free users stored at three different temperatures (-20, 4 and 20°C) with (ethylenediaminetetraacetic acid (EDTA) 3%; with sodium fluoride/potassium oxalate (NaF/KOx) 1.67%/0.2%) and without preservatives up to 6 months for the study. It was concluded that the highest mephedrone stability in blood was maintained when specimens were stored at -20 °C adding NaF/KOx as preservative.

3.6.2.5 Stability of Opiate-type Drugs in Blood

The stability of morphine, codeine and 6-MAM in spiked blood samples was studied after different sampling condition stored at two different temperatures (-20 and 4 °C) on the effect of the type of sampling tubes (glass, polypropylene or polystyrene tubes), the addition of preservative (with addition of dipotassium ethylenediamine tetraacetic acid (K₂EDTA) or sodium oxalate (Na₂C₂O₄)) and the type of anticoagulant (with or without addition of sodium fluoride (NaF)) [598]. It was shown that opiate concentrations were decreased in all conditions and the most stable was 6-MAM. The addition of NaF as preservative improved the stability of opiates at all conditions studies, whereas the type of anticoagulant did not affect the stability of opiates. The authors concluded that blood samples should be stored at -20 °C in glass tubes containing oxalate and NaF for maximum stability.

Results of whole blood samples from deceased and living persons containing morphine, codeine and 6-MAM have been compared with those from which had been stored at -20 °C for 4–9 years and reanalysed using the same analytical methods [599]. The study showed that in real life whole blood samples, the concentration of morphine and codeine are relatively stable during long-term storage at -20 °C although statistically significant decreases were observed for morphine and codeine with no significant change in the morphine to codeine concentration ratios. On the other hand, 6-MAM exhibited a considerable decrease in concentration under the same storage conditions.

3.6.2.6 Stability of Valproic Acid in Blood

Valproic acid, a commonly prescribed anticonvulsant agent for the treatment of all types of epilepsy, is a delicate analyte and a stability study has shown that its level in PM blood declined by 85% after storage at room temperature for 28 days [600]. Its instability appeared to be an issue when the blood sample are stored at 4 or 20 °C, or when the corpse is exposed for a prolonged period to room temperature before sample collection.

4. References

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Media Evidence

Audio, 2013-2016

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1. Introduction

This report continues the audio review of the 16th Interpol International Forensic Science Symposium in October 2013 [1] and catalogues the research, advances, and application of scientific methodologies and techniques relating to the forensic examination of audio evidence. This report primarily consists of a literature review of published articles in forensic science journals and the proceedings of various international organizations and forensic conferences between July 2013 and June 2016. It also contains references from different relevant working groups in the field.

2. Authentication (Catalin Grigoras, Jeff M. Smith)

The forensic audio authentication section presents peer-reviewed papers on: structure, format, audio compression, edit detection, electric network frequency analysis, and recorder attribution.

2.1. Structure and format

Structure and format analysis of digital audio data and comparison against exemplar recordings are parts of the framework to authenticate digital audio evidence. Their results can be of great importance to determine whether or not the evidence is consistent with an original, or to detect traces left by different audio editors.

Koenig & Lacey [2] conducted a study of forty WAV PCM uncompressed files produced on nine different digital audio recorders, and then passing them through four digital audio editing programs. The obvious chunk differences between the originals and their re-encoded versions are presented and explained.

Grigoras & Smith [3] presented an extensive investigation on the structure and format for WAV PCM uncompressed files created by 31 commercially available digital audio recorders and 20 digital audio editors. The results fully support the findings in [2] and indicate that most recorders and editors create files with different metadata.

2.2. Compression analysis

Lossy compression history assessment is another important analysis in digital audio authentication, showing whether or not the evidence signal is consistent with an exemplary

recording, or indicating traces of previous lossy recompression and inconsistencies with an original.

Hicsonmez et al. [4] proposes a technique to identify the initial codec of a lossy encoded audio signal without using any metadata or information about the bit stream format of the codec. The technique samples 2-4 kilobytes of data from a lossy encoded audio and analyzes the randomness and chaotic nature of the sampled data to build statistical models that represent encoding process associated with different codecs. 16 audio codecs were analyzed, including high quality, PSTNs, cellular networks, and VoIP networks. Results show that the initial codec can be recognized with an accuracy of more than 95 percent.

Bianchi et al. [5] presented a method to use the modified discrete cosine transform coefficients' statistics to detect traces left by double compression and localization of possible tampering in MP3 files. The proposed algorithm shows to be effective when the bitrate of the second compression is higher than the bit-rate of the first one.

Braun [6] provides an overview of digital audio authentication techniques for civil and criminal proceedings on music samples, including compression artifacts, sampling traces, watermarking, and Electric Network Frequency Analysis.

Adaptive Multi-Rate (AMR) is another common lossy compression algorithm that can be found in real cases. It was adopted as the standard speech codec by 3GPP and it is widely used in GSM and UMTS mobile phone communications. Luo et al. [7] propose sample repetition rate analysis to recognize Adaptive Multi-Rate (AMR) lossy compression artifacts on WAV PCM decompressed audio files, while in [8] Luo et al. describe a technique to discriminate double compressed from single compressed AMR audio signals.

Luo et al. [9] present a method to recognize a uncompressed wave audio that went through one of three popular compression schemes: MP3, WMA (Windows Media Audio), and AAC (Advanced Audio Coding). By analyzing the corresponding frequency coefficients, including Modified Discrete Cosine Transform (MDCT) and Mel-frequency Cepstral Coefficients (MFCCs), of those original audio clips and their decompressed versions with different compression schemes and bit rates, the authors propose several statistics to recognize the compression scheme as well as the corresponding bit rate previously used for a given WAV signal.

Korycki [10] addresses the problem of authenticity examination of lossy compressed digital audio recordings and the evaluation of statistical features computed based on the MDCT coefficients as well as other parameters that may be obtained from compressed audio files. This work is employed to perform detection of multiple compression stages which may cover up tampering activities, and to identify traces of montage in digital audio recordings. The experiments conducted by the author reveal that it is possible to identify initial encoder correctly based on statistical analysis of re-compressed audio file. The effectiveness of authenticity examination methods is tested on a large, predefined music database consisting of nearly one million compressed audio files.

Wang et al. [11] present an approach to detect double compressed MP3 audio by frequency vibration. With the analysis of double compression effect on MDCT coefficients in MP3 audio signals, the authors propose a simple feature called FVV (Frequency Vibration Value) to measure the vibration caused by MP3 double compression. The experimental results also show the possibility to estimate the original bit-rate for a double compressed MP3.

Yang [12] proposes a method to analyze the additive noise as an artifact of tampering by masking the local traces of audio editing. Noise can cover audible evidence of forgery and destroy traces of other tampering operations. The author describes a feature named Sign Change Rate (SCR) for detecting additive noise. Via theoretical analyze and extensive experiments, it shows the proposed feature is effective in additive noise detection. Also the method can be a potential tool for forgery localization of digital audio.

2.3. Edit detection

Grigoras & Smith [13] propose a new method for forensic media authentication based on quantization level analysis (QL) of a digital audio signal. Five methods for calculating QL are provided including: QL histogram, QL length, greatest common divisor, Fast Fourier Transform (FFT), and Cepstrum. Examples of QL analysis are provided to demonstrate its use in the detection a recording's native QL versus the file format bit depth, the detection of previous signal processing, the analysis of consistency with exemplary recordings, detection of local edits and audio montage.

Korycki [14][15] presents the problem of tampering detection in compressed audio files and discusses new methods that can be used for authenticity analysis of digital recordings. Presented approaches consist in statistical analysis of the MDCT coefficients and other parameters that can be extracted from lossy compressed audio files. Calculated feature vectors are used for training selected machine learning algorithms. The detection of multiple compressions covers up tampering activities as well as identification of traces of montage in digital audio recordings. To enhance the methods' robustness an encoder identification algorithm was developed and applied based on analysis of inherent parameters of compression. The effectiveness of tampering detection algorithms is tested on a p large, redefined music database consisting of nearly one million compressed audio files. The influence of compression algorithms' parameters on the classification performance is discussed as well.

Gartner et al. [16] propose a multi-codec tampered dataset. The doctored speech content does not contain audible artifacts or changes of semantic meanings, but tampered regions which can be detected via lossy compression traces, e.g. using framing grid analysis. Possible applications, content and annotations included, and the steps required to generate the dataset are described.

Chen et al. [17] propose an audio forensics scheme to detect and locate speech audio forged operations in time domain (including deletion, insertion, substitution and splicing) by performing discrete wavelet packet decomposition and analyzing singularity points of audio signals. The described algorithm detects singular points due to the changing of the correlation as an artifact of local tampering. Then the singularity analysis based on wavelet packet is performed and five parameters are used to detect and locate audio forgeries in time domain.

Jin & Kim [18] describe a signal fragment identification and extraction system based on: the audio decoding; applying Support Vector Machine on it; fragment recovery; adding a maximum similar frame header in front of the fragment; then a chroma feature is extracted from the decoded signal to achieve the audio identification.

Xiao et al. [19] propose a method to detect duplicated segments in a WAV PCM uncompressed file. The method is based on the similarity calculation between two different segments. Duplicated segments are prone to having similar audio waveform, i.e., a high similarity. The authors use fast convolution algorithm to calculate the similarity between any

two different segments in a digital audio file and use the similarity to assess the duplicated segments.

Milani et al. [20] present a multimodal approach for audio tampering detection that analyzes both the audio component and the video component of a recorded video file. The proposed solution estimates the volumetric characteristics of the environment where the multimedia content has been captured both from the video and audio signals. Then, the approach checks the consistency of the environment characteristics estimated from the audio signal with respect to those estimated from video files. The described solution proposes to detect audio and video edits.

Kurniawan et al. [21] introduce three features to classify the microphone models while take into consideration the issue of identical model. Those features are analyzed and compared in the authors' experiments. The results indicated that Gabor filterbank potential to detect local traces of edits in digital audio recordings.

2.4. Electric Network Frequency analysis

Elmesalawy & Eissa [22] propose a new method for establishing a robust ENF reference database using geographical information system (GIS) and wide area frequency measurements. The method is based on building the ENF reference database from a number of frequency sensors deployed over multipoint on the grid rather than single point. The minimum number of sensors required is determined according to the frequency sensitivity of the ENF extraction algorithm and the estimation accuracy of the frequency sensor. The sensors locations are decided based on partitioning the power grid to a set of geographical frequency coherent areas. A harmony search algorithm using GIS data and wide area frequency measurements is proposed to identify the geographical frequency coherent areas for different disturbance scenarios. Results showed that the proposed method can be used to enhance the accuracy of the ENF database matching process.

Fechner & Kirchner [23] study with multiple mobile recording devices confirms the hypothesis that background noise, generated by mains-powered electronic devices in proximity to the recording device, is a carrier of ENF artifacts. Experiments in an indoor setting suggest a very high robustness and indicate the presence of ENF artifacts even multiple rooms apart from the noise source.

Liu et al. [24] propose a new method to extract the embedded 50/60-Hz frequency component from an analyzed digital audio recording and then compare it with a reference power system frequency database, trying to determine the recording time and judge if the audio has been tampered with or not.

Campos & Ferreira [25] studied the variation of both ENF and Total Harmonic Distortion (THD) in different locations of the electric grid in Portugal. To this purpose, the authors describe the design and test of a complete ENF and THD monitoring system which includes autonomous sensors that are able to extract these parameters, and send them via web to a server. An interactive web interface is also integrated which allows real-time monitoring and access to stored data. ENF and THD data have been collected on different periods and locations of the Portuguese electric grid. Besides confirming the known features of the ENF parameter, results have shown that THD data is strongly affected by local factors and therefore exhibit little geographical consistency. In a forensic perspective, while the ENF is tailored to spot events in time, the THD has the potential to spot events in space.

2.5. Recorder attribution

Halnics & Kinnunen [26] consider recognizing source cell-phone microphones using non-speech segments of recorded speech. Taking an information-theoretic approach, the authors use Gaussian Mixture Model (GMM) trained with maximum mutual information (MMI) to represent device-specific features. Experimental results using Mel-frequency and linear frequency cepstral coefficients (MFCC and LFCC) show that features extracted from the non-speech segments of speech contain higher mutual information and yield higher recognition rates than those from speech portions or the whole utterance. Identification rate improves from 96.42% to 98.39% and equal error rate (EER) reduces from 1.20% to 0.47% when non-speech parts are used to extract features. Recognition results are provided with classical GMM trained both with maximum likelihood (ML) and maximum mutual information (MMI) criteria, as well as support vector machines (SVMs). Identification under additive noise case is also considered and it is shown that identification rates reduce dramatically in case of additive noise.

2.6. Reverberation analysis

According to Moore et al. [27] identification and verification of the location in which a recording was made are important yet understudied topics in audio forensics. The concept of 'roomprints' is defined as a quantifiable description of an acoustic environment which can be measured under controlled conditions and estimated from a monophonic recording made in that space. The various types of information which could be included in a roomprint are reviewed based on their expected reliability and the feasibility of extracting them from a recording. Frequency-dependent reverberation time is identified as a particularly promising feature in both regards. A room identification experiment was conducted using room impulse responses from 22 rooms. Depending on the frequency resolution and lower frequency extent of the roomprint identification rates of up to 97% were reported.

Zhao [28] analyzed the magnitudes of acoustic channel impulse response and ambient noise as the environmental signature and its use to authenticate the integrity of query audio and identify spliced audio segments. The proposed scheme firstly extracts the magnitudes of channel impulse response and ambient noise by applying the spectrum classification technique to each suspected frame. Then, correlation between the magnitudes of query frame and reference frame is calculated. An optimal threshold determined according to the statistical distribution of similarities is used to identify the spliced frames. Furthermore, a refining step using the relationship between adjacent frames is adopted to reduce the false positive rate and false negative rate. Effectiveness of the proposed method is tested on two data sets consisting of speech recordings of human speakers. Performance of the proposed method is evaluated for various experimental settings. The reported results show that the method detects the presence of spliced frames and forged segments.

3. Forensic Speech Science (Andrzej Drygajlo)

Forensic speaker recognition (FSR) is used as a traditional cover term for forensic automatic speaker recognition (FASR), forensic semiautomatic speaker recognition (FSASR) and the auditory-phonetic-and-acoustic-phonetic approach (Neustein & Patil [29], Hansen & Hasan [30]). Other terms such as "forensic speaker comparison" (Foulkes & French [31]) and "forensic voice comparison" (Enzinger & Morrison [32]) are also in use. This chapter is focused mainly on FASR and FSASR methods and related methodology.

3.1 Reviews and guidelines

The most recent, general tutorial on speaker recognition including forensic speaker recognition is published by Hansen & Hasan [30]. An INTERPOL survey on international practices in FSR by law enforcement agencies is presented in [33]. A guide for thinking and practice in investigations (investigative mode) and in court proceedings (evaluative mode) is

presented in Jackson et al. [34]. In 2015 two important ENFSI documents were finalized and published following this guide: 1) ENFSI Guideline for Evaluative Reporting in Forensic Science [35], 2) Methodological Guidelines for Best Practice in Forensic Semiautomatic and Automatic Speaker Recognition including Guidance on the Conduct of Proficiency Testing and Collaborative Exercises [36]. Other reviews on FSR were presented by Drygajlo [37], French and Stevens [38] and Campbell [39].

3.2 Current approaches in forensic speaker recognition

A diversity in approaches used for speaker identification by law enforcement agencies in different parts of the world emerges from the “INTERPOL Survey of the Use of Speaker Identification by Law Enforcement Agencies” [33]. A questionnaire of this survey was sent to law enforcement agencies in the 190 member countries of INTERPOL. A total of 91 responses were received from 69 countries. Finally, only 44 respondents’ agencies had the capability to analyse voice recordings. Half of them are in Europe. The approaches reported were: auditory, spectrographic or auditory-spectrographic, auditory-acoustic-phonetic, acoustic-phonetic, human-supervised automatic and fully automatic. The most popular was the auditory-acoustic-phonetic approach, followed by the spectrographic or auditory-spectrographic and the human-supervised-automatic approaches. The fully automatic approach was the least popular.

Regarding reporting, the respondents had the following choices: identification/exclusion/inconclusive, numeric posterior probability, numeric likelihood ratio, posterior-probability verbal scale, likelihood-ratio verbal scale and UK Position Statement scales. In general, “identification/exclusion/inconclusive” was the most popular choice. In Europe this choice was the most popular too, but verbal likelihood scales were on the second position.

Other reviews and guidelines are more focused on FASR and FSASR methods and their comparison with other methods. For example, in the tutorial review by Hansen & Hasan [30] different aspects of automatic systems, including voice-activity detection (VAD), features, speaker models, standard evaluation data sets, and performance metrics are discussed. In this review, the first part involves forensic speaker-recognition methods, and the second illustrates how a naïve listener performs this task from a neuroscience perspective.

From 2013 to 2015 the European Network of Forensic Science Institutes (ENFSI) project “Methodological guidelines for semiautomatic and automatic speaker recognition for case assessment and interpretation”, chaired by Drygajlo was conducted in the framework of the ENFSI Monopoly Programme 2011 “Improving Forensic Methodologies across Europe (IFMAE)” within the context of the ENFSI Forensic Speech and Audio Analysis Working Group (FSAAWG). Universal methodology has been developed by participants of this project for forensic automatic and semi-automatic speaker recognition (FASR and FSASR, respectively) when used in casework situations. In general, forensic speaker recognition denotes the different ways of discriminating one person from another based on speech, taking into account the limitations of forensic speech material and the specific needs of reporting the results to a court. The role of the forensic expert is to provide an interpretation of the speech material available to a police investigation or to a law court. The project has recently published “Methodological Guidelines for Best Practice in Forensic Semiautomatic and Automatic Speaker Recognition including Guidance on the Conduct of Proficiency Testing and Collaborative Exercises” [36]. These methodological guidelines constitute a pioneering result in the domain of forensic speaker recognition. They are based on developments in the ENFSI forensic speech and audio analysis community for last twenty years [29]. They present a standard approach for FASR and FSASR based on scientifically approved methods for calculation and interpretation of speech evidence in the Bayesian

interpretation framework and in providing detailed guidance documentation for forensic experts in the field.

The rest of the present review follows the structure and annotated biography of these guidelines for the time period 2013-2014.

3.3 Methodology of FASR and FSASR

Forensic automatic speaker recognition (FASR) and forensic semiautomatic speaker recognition (FSASR) are two types of technical speaker recognition, where automatic or partially automatic speaker recognition methods in their central processing (feature extraction, feature modelling, similarity scoring and calculation of likelihood ratio) are adapted to forensic applications [36].

Text providing introduction to and overview of general automatic speaker recognition is written by Hansen & Hasan [30]. Specific considerations to the forensic aspects of automatic speaker recognition are given by Drygajlo in [40], [41] and [42].

3.3.1 Features

Kinnunen & Li [43] provide an exhaustive review, still up-to-date, of short-term spectral envelope features with special emphasis on features typically used in FASR. Studies on Long Term Formant (LTF) analysis, a typical FSASR application, include papers by Gold [44] and Jessen et al. [45]. Local application of cepstral coefficients to specific vowels and consonants (also called segmental cepstra) is addressed in Rose [46]. Studies in which the dynamics of formants have been investigated in a forensic context include Zhang et al. [47] [48].

Although formants are usually approached semiautomatically, there have been studies in which formant frequencies are measured automatically (Jessen et al. [45] and Harrison [49]). Harrison provides general information on formant measurements in FSR and presents experimental results on aspects such as the influence of the formant tracking software and the analysis settings.

Kinoshita & Ishihara [50] demonstrate how mean, variance, skewness, kurtosis and related global fundamental frequency distribution parameters can be used as dimensions in a multidimensional feature vector and modelled with the Multivariate Kernel Density (MVKD) formula.

Discussion of the forensic aspects of Articulation Rate and relevant AR-population statistics are provided by Gold [44]. The percentage of the syllable that is vocalic or the percentage of the syllable that is voiced, as well as other measures of speech rhythm and timing have been investigated with a focus on speaker-discriminatory information by Leemann et al. [51].

A proposal of how auditory features can be treated within the likelihood ratio framework by using N-grams is presented by Aitken & Gold [52].

3.3.2 Speaker modelling and similarity scoring

The distinction between deterministic and statistical models is summarised, along with further literature in Drygajlo [42] and addressed empirically in Drygajlo & Ugnat [53]. Normalisation methods, supervectors and many other aspects of automatic speaker recognition have been summarised in Kinnunen & Li [43].

Some studies in which the GMM-UBM approach has been applied to FSASR are presented in Jessen et al. [45].

The Multivariate Kernel Density (MVKD) formula is presented and its essentials explained by Rose [46]. Some studies in which this formula has been used in FSASR are as follows: on vowel formant centre frequencies (Rhodes [54]), on formant trajectories with curve fitting (Zhang et al. [47][48], Rhodes [54], Hughes [55]), on fundamental frequency distribution parameters (Kinoshita & Ishihara [50]; on segmental cepstra (Rose [46]), and on long-term formants and articulation rate (Gold [44]).

3.3.3 Bayesian interpretation framework and calculation of likelihood ratio (LR)

Literature on some general principles of the interpretation of forensic evidence in a Bayesian interpretation framework includes Jackson et al. [34] and the literature cited therein. Further introductions to the Bayesian framework include Drygajlo [42][36], with special emphasis on FASR and FSASR.

The difference between the scoring method and the direct method, as well as the significance of the relevant population database, the suspected speaker reference database and the suspected speaker control database is explained by Drygajlo [40][41].

3.3.4 Calibration and fusion

Calibration has two different aspects, calibration as a process of converting an uncalibrated to a calibrated likelihood ratio and calibration as a goodness criterion of a test set in a validation. For example, Morrison [57] focuses on the former aspect and Ramos & Gonzalez-Rodriguez [58] on the latter. Further references addressing one or both of these aspects of calibration are Ramos et al. [59] and Brümmer & Swart [60]. A study using calibration in both deterministic and statistical models is presented in Drygajlo & Ugnat [53]. A recent tutorial of logistic regression calibration and fusion is provided by Morrison [57].

Aside from fusion there are other proposals of how to combine evidence from different methods. Whereas fusion can be considered as a “back-end” combination process, there is a “front-end” proposal by Gold [44] that is based on correlation tests of different features. Another proposal of combining results from different methods has been to use Principal Component Analysis (PCA) as a means of providing an alternative to fusion by combining information from different features (Nair et al. [61]).

3.3.5 Mismatched recording conditions

Mismatched recording conditions compensation with special emphasis on FSR and calculation of likelihood ratio (LR) is presented in Alonso Moreno & Drygajlo [62]. Methods of mismatch compensation that are compatible with the i-vector approach are summarised by Hansen & Hasan [30]. Forensic case simulations and LR-testing based on mismatched conditions and their compensation are presented in Enzinger & Morrison [32] and Enzinger et al. [63].

3.4 Method Validation

The basis for the method validation procedure is the ENFSI “Guidelines for the single laboratory validation of instrumental and human based methods in forensic science” [64] and a guideline for the validation of likelihood ratio methods presented in Meuwly et al. [65], as well as Chapter 4 “Method Validation” in [36]. Validation has to be performed with speech that is typical of the speech material the forensic laboratory is confronted with in everyday work. Examples of validation based on forensic data include paper by van der Vloed et al. [66]. “The purpose of validation of a FASR or FSASR method is to measure its performance in specific conditions and to determine whether the performance satisfies a given validation criterion or several criteria” [36].

3.4.1 Performance characteristics and metrics

“Performance characteristics (e.g., Tippett plots I and II as well as Proportions Trade-Off (PTO), Applied Probability Error (APE), Empirical Cross Entropy (ECE) plots) describe support for the correct hypothesis of a LR method” [36].

Tippett plots I (inverse cumulative distribution functions) are more common in the FASR literature (e.g., Drygajlo [41], van Leeuwen & Brümmer [67], Ramos & Gonzalez-Rodriguez [58]), whereas Tippett plots II (only one cumulative distribution function is inverted) are more commonly used in the FSASR literature (e.g., Gold [44], Hughes [55], Nair et al. [61]).

The probabilities of misleading evidence ($PMEH_0$ and $PMEH_1$) are measures of accuracy related to Tippett plots. The proportions trade-off (PTO) curve and equal proportion probability (EPP) represent discriminating power of FASR or FSASR.

3.4.2 Performance characteristics and metrics based upon ranges of prior probabilities

The empirical cross-entropy (ECE) plots are explained and argued for in Ramos & Gonzalez-Rodriguez [58] and Ramos et al. [59].

3.5 Case Assessment

3.5.1 Cognitive

Many publications about cognitive bias as applied to forensic science have been presented by Dror [68]. The speaker identification group at the Netherlands Forensic Institute since about ten years has developed a method called blind grouping with the intention of preventing confirmation bias, which is one type of cognitive bias (Cambier-Langeveld et al. [69], including further relevant references).

3.5.2 Relevant population

Discussion of the concept of relevant population and different ways of defining it is provided by Hughes [55] and Hughes & Foulkes [70].

3.5.3 Quantity and quality profile of the forensic audio material

Technical Quality. An example of a topic that has been addressed in this category is the effect of telephone filtering on the measurement of formant frequencies. The effect of telephone transmission on the measurement and LR-based testing of formant dynamics has been studied in Zhang et al. [48].

3.5.4. Mismatched conditions.

Long-term non-contemporaneous speech is studied by Kelly et al. [71][72], Kelly & Harte [73] and Rhodes [54]. Language mismatch is addressed in Künzel [74] and van der Vloed et al. [66]. Effects of vocal effort including shouted speech and ways to compensate for it are presented by Hanilci et al. [75].

3.6 Case-specific Evaluation and Interpretation

At the present time, Tippett plots provide a standard representation for performance evaluation of LR methods in FASR and FSASR cases (e.g., Gold [44], Hughes [55]). Examples of tests that are adapted to the specific conditions of a case are given by Rose [76], Enzinger & Morrison [32] and Enzinger et al. [63]. Normally, they are completed by performance metrics such as probabilities of misleading evidence and log-likelihood-ratio cost (Cllr) [36]. A version of the proposal to use case-specific probabilistic error as a way of presenting case-specific strength of evidence results is expressed by Solewicz et al. [77].

3.7 Case File and Reporting

In general, the basis for case file and reporting is the “ENFSI Guideline for Evaluative Reporting in Forensic Science” [35] and Chapter 7 in [36] specifically dedicated to FASR and FSASR methods.

4. Miscellaneous (Jeff M. Smith, Catalin Grigoras)

This section presents additional studies related to forensic audio examination including recorded speech enhancement and gunshot analysis. This section also describes the latest developments in best practice guidelines for audio forensics.

4.1. Speech enhancement and intelligibility

Gower and Hawksford [78] achieve a decreased computational load when separating mixed signals in audio samples by examining the underlying sparsity using a proposed SRS (sliding ratio signal) algorithm. This process simultaneously detects the number of sources as well as their mixture matrix wherein a sliding discrete Fourier transform and a sliding Goertzel algorithm are applied in both time and time-frequency domains.

Javed and Naylor [79] calculate the level of reverberation distortion in speech recordings with an extension to the proposed Reverberation Decay Tail (R_{DT}) metric. This work is motivated by the common forensic audio problem where intended speech is captured in uncontrolled environments (noisy and reverberant) with the target speaker’s voice captured by a microphone outside of the desired proximity. The authors achieve better performance on realistic recordings by configuring the R_{DT} metric for wideband speech and optimizing Bark spectrum calculation based on ISO 226 standard equal loudness contours. With this extension, the authors obtain a more reliable estimate of a signal’s reverberant tail at the expense of increased computational load. Therefore, the authors propose limiting Bark spectra resolution to every quarter Bark.

In [80], Stolbov and Aleinik propose an adaptive technique for capturing speech when using a microphone array. The speech alignment procedure operates with low complexity in the frequency domain, where direct estimation of the transfer functions is made, and can therefore be implemented in real-time. This is done by matching microphone signals to the response from a frequency-domain fixed beamformer (FBF). In an experimental test of the method, the authors found a suppression of diffuse noise by about 23dB and suppression of wide band interference up to 17dB.

Stanton et al. [81] derive a continuous and differentiable approximation of the Speech Intelligibility Index (SII) which is suitable for efficient optimization of speech enhancement algorithms. Using the Jacobian maximum and its inverse, min and max functions are approximated for the SII which can then be used to develop its partial derivative. A simplified derivative is then developed by assuming the effective noise level is greater than the self-speech masking level; a finding established in previous research studies. The proposed approximation was applied to near-end listening enhancement optimization and showed large positive gains in SII.

4.2 Gunshot Analysis

In [82], Maher and Shaw examine the acoustic characteristics of gunshots recorded by a consumer-grade digital voice recorder and make several observations and recommendations in this regard. Since digital voice recorders are not optimized for the amplitude of gunshots nor their transient nature, examiners must take care when analyzing these sounds captured in typical forensic recording situations. The authors present findings related to testing variables presented by digital voice recorders including the effect of automatic gain control (AGC) and encoding algorithms such as MP3. Observations from lab

tests as well as case reconstruction tests include imprecise time and amplitude information in the recorded gunshot signal. The authors caution that misinterpretation of evidence is possible to do potential signal clipping, acoustic reflections, and perceptual coding effects and stress that great care must be taken.

Lacey et al. [83] apply a cross-correlation comparison of recorded gunshots to examine the effect of analysis sample length on accuracy of the comparison. In analyzing material from three real cases, they found that most of the time, with increased sample length (a range they tested from 2 and 50 milliseconds) maximum cross-correlation values decreased. In conclusion, the authors found the method useful for providing objective information to assist more subjective, qualitative assessments of similarity when attributing recorded gunshots to firearm sources in blind.

4.3 Best practice guidelines

There has been much progress internationally in the development of standard criteria for audio forensics in practice. The Scientific Working Group on Digital Evidence (SWGDE), a US law enforcement led group, published a “Best Practices for Digital Audio Authentication” [84] which outlines the major considerations for an authentication examination of digital audio evidence. This document outlines necessary pre-examination activity, examination protocols which can be distinguished globally (analyses related to the digital file as a whole) or locally (time relevant analyses), and the criteria necessary in arriving at and documenting a conclusion. This same group’s “Best Practices for Forensic Audio” [85] was updated in 2015 to address advances in technology as well as the inclusion of a decision tree when receiving evidence and examination requests.

The UK Forensic Science Regulator published an appendix to its Codes of Practice and Conduct on “Speech and Audio Forensic Services” [86]. This document focuses primarily on speech enhancement and forensic speaker comparison providing minimum requirements for the handling and processing of audio recorded evidence, test methods and their validation, equipment and lab considerations, and the terminology to be used consistently in documentation. This document is written to adhere to ISO/IEC 17025:2005 requirements and provides considerations for the forensic audio provider accredited under the same.

The terminology used for this review adheres to standards for digital and multimedia evidence terminologies found in [85],[87], and [88] which define digital evidence, bit stream duplicate, authentication, audio enhancement, original recording, archive image, lossy compression, lossless compression, native file format, timestamp, timeline sequence reconstruction, etc.

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Media Evidence

Video and Imaging, 2013-2016

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1. Introduction

In this review, the most important developments are presented for the following general fields of expertise: : (1) Biometric analysis of image material, (2) Detection of image manipulation, (3) Camera source identification, (4) Video image processing and Image search.

This year we have been requested to add Video to the review. Since there is clearly overlap, we have integrated this field in the review.

Working groups and organizations

The development of forensic image analysis has several international working groups:

- SWGIT: an American group that has produced a lot of guidelines and best practice manuals. <http://www.swigit.org> The group has terminated operations, since the new OSACs are formed <http://www.nist.gov/forensics/osac.cfm> . <http://www.swigit.org/>
- ENFSI DIWG: The ENFSI Digital Imaging Working Group that is focused on methods, techniques, education and training. <http://www.enfsi.org>
- LEVA : an American group focused on video processing and training: <http://www.leva.org>
- EESAG: an Australian-New Zealand group that proficiency tests for video and audio processing: <http://www.nifs.com.au/eesag/about.html>
- AGIB, A working group in Germany that is focused on facial image comparison: <http://www.foto-identifikation.de/> .
- FISWG, An American group since 2009 that is focused on facial image comparison: <http://www.fiswg.org>
- OSAC Facial Identification Subcommittee, An American group part of the Organisation of Scientific Advice Committees, with focus on standards and guidelines related to the image-based comparisons of human facial features: <http://www.nist.gov/forensics/osac/sub-face.cfm>
- American Academy of Forensic Science [1] Within the American Academy of Forensic Science the Digital and Multimedia Sciences Section works in this field.

Since 2003 each year a workshop was organized on Forensic Image and Video processing with handouts on the methods for face comparison, video restoration, 3D reconstruction, length measurement, photogrammetry and image processing. Also each year a scientific session was organized on this field. More information is available on: <http://www.aafs.org>

2. ENFSI Forensic IT Working Group

The forensic IT working group of ENFSI [2], [3] deals with digital evidence as such. There exist some overlap with the Digital Imaging working group, and for that reason joint events are organized.

Since most CCTV-systems are digital nowadays, often the question of handling the CCTV system itself is a question of digital evidence. Hard drives and other digital media should be handled in a secure way with proper forensic imaging software. The working group organizes training conferences each year. More information is available from <http://www.enfsi.eu/>.

2.1 Biometric analysis of image material

Biometrics is regularly announced in news items as a panacea against terrorism, security problems, fraud, illegal migration, etcetera. Biometrics, which can be defined as the (automatic) identification or recognition of people based on physiological or behavioral characteristics, is not a single method or technique, but consists of a number of techniques, with each their own advantages and drawbacks. None of the available biometric modalities combines the properties of an ideal biometrics system. We have to acknowledge that biometrics never can be 100% accurate. However, if requirements and applications are carefully considered, biometric systems can provide an important contribution to investigation, authentication and safety.

Within the context of person identification (individualization), different processes can be defined. Within different areas of science, different terminologies are used for the same process, and sometimes the same terminologies are used for different processes. Therefore, a clear definition of the different terms as used in this text is important and made explicit here.

Recognition can be defined as the process of identifying or matching a person, his/her photograph or image with a mental image that one has previously stored in long term memory. Recognition requires observation and retention of a person's features and the process of comparison of the retained information with an external image whether it be the live person, a photograph or composite image. The word recognition is important for investigation as well as witness statements. Recognition is within the forensic community also used for the automated searching of a facial image in a biometric database (one-to-many), typically resulting in a group of facial images ranked by computer-evaluated similarity.

Identification is the most contentious term because this most often used term can mean several things in different context, like the automated searching of a facial image in a biometric database (one-to-many) in biometrics, the examination of two facial images or a live subject and a facial image (one-to-one) for the purpose of determining if they represent the same person in forensics, or the assignment of class or family name in biology and chemistry. Therefore, the authors of this paper prefer not to use the term identification unless the meaning is unambiguous within the context.

Recall is here defined as the process of retrieving descriptive information of a person from long term memory in the absence of the person, his/her photograph or other image. Recall requires observation, retention and reproduction of a person's features. Recall is essential for the production of composite images, as produced by a police artist for investigational purposes. However, these images can only be used as investigative tools, and can never be used as proof of identity.

2.2 Face

On top of the list of preferred, and in most travel documents required, biometric modalities is the face. The face has always been the most important personal feature on travel documents. The most important change the last decade is that the face is now also stored digitally in passport, and is optimized for automatic facial recognition. The automated systems are still very sensitive to ageing of the person depicted [Agrawal 2015,]; The latest test results indicate that higher resolution and well controlled images may result in a 10-fold better performance than using average passport like images.

Facial image comparison is defined as the visual examination, by a human operator, of the differences and similarities between two facial images or a live subject and a facial image (one-to-one) for the purpose of determining if they represent the same person. In biometrics the one-to-one comparison is termed verification. The Facial Imaging Scientific Working (FISWG) group also uses the term Facial Identification for the same process. However, the authors of this review prefer to use the term facial image comparison, because that exactly describes the process, and cannot be confused with the use of the word identification as used in other contexts.

Facial Reconstruction is used in two different meanings:

1. The process of reconstructing three-dimensional facial (computer) models of individuals from their 2D photographic images or video sequences.
2. The process of recreating the face of an individual (whose identity is often not known) from their skeletal remains through an amalgamation of artistry, forensic science, anthropology, osteology, and anatomy.

These two different uses of facial reconstruction may meet when three-dimensional computer models are used to recreate the face of an individual based on skeletal remains. The current review does not consider facial reconstruction as a means to recreate the face of an individual based on skeletal remains.

Facial composite is a graphical representation of an eyewitness's memory of a face, as recorded by a composite artist, also sometimes termed facial sketch. A facial composite may be based on combination of images of facial features (photo composition) as well as actual drawings by a composite artist.

2.3 Facial image recognition

Biometric systems that can search databases with facial images, using automatically extracted facial features, are still being developed further. Face recognition error rates have declined massively in the two decades since initial commercialization of the various technologies [4]

However, one of the complicating issues is that the images of the unknown person often differ from the target images in the database with respect to the orientation of the head, the distance to the camera, the illumination and the image resolution. Recently, increased attention has been given to the existence of quality measures for face recognition [5][6]. New approaches focus on better acquisition techniques in order to get better images, from which as many facial features as possible can be extracted for comparison to images in the database.

2.4 Unconstrained images

Forensic material in general is not acquired under controlled circumstances. This means that a number of confounding factors may influence the effectiveness of face recognition. A number of academic studies try to tackle the different confounding factors, like pose issues,

resolution issues, and occlusions. How different quality features may influence the performance of face recognition systems has been studied by Dutta et al [6].

Several techniques are being developed to use unconditioned images and media collections for unconstrained face recognition [7] and partially occluded faces [8]. Though issues surrounding pose, occlusion, and resolution continue to confound matchers, there have been significant advances made in face recognition technology to assist law enforcement agencies in their investigations [9]

Park et al [10] report improvements in facial image retrieval of partially occluded images (sunglasses and scarfs), resulting in more than 90% retrieval of partially occluded faces.

Peng et al [11] have reported successful face recognition using a novel method for low-resolution face recognition.

2.5 Pose variation

Pose is the “orientation of the face with respect to the camera, consisting of pitch, roll, and yaw”. An optimal frontal pose may be considered as 0° in all directions. Variations to the optimal pose can be due to photographing a physical subject who can move freely during the capture process, or misalignment of the camera. As images are a 2-dimensional representation of the 3-dimensional world, pose of a subject has a major influence on the image captured by a capturing device. As a result of this the appearance and position of facial features can change depending of the pose of the person and the position of the camera at the moment of capture. This is, together with inter and intra observer variability of landmark annotation, one of the main causes of the limited value of landmark measurements on photographs [12]. However, development of pose detection and automatic landmark detection has been reported to result in almost 90% identification accuracy in side view positions [13].

For predicting face recognition performance in a video, it was observed that face detection confidence and face size serve as potentially useful quality measure metrics [14].

2.6 3-dimensional face comparison

The most promising approach to the complicating issues of pose and illumination is the use of 3 dimensional models for pose and illumination correction. Since the previous review, there has been an increase in reports on development of methods that are based on the use of 3-dimensional computer models of faces. A number of 3d-acquisition systems are now available for the acquisition of these models. Most 3d-cameras work with a configuration of 1 or more normal digital photo cameras, a flash and the projection of a pattern on the face. These models can be used in two ways. A 3d-facial model of a suspect can be compared to a 3d-model of an unknown person, or the 3d-model of a suspect is used to compute an image that can be compared to an image of an unknown person. Since there are many sources of images and video in practice, a number of studies are focused on the (partial) reconstruction of 3d-models from 1 or more images or video streams. Van Dam et al [15] developed a model 3-D face reconstruction algorithm based on 2D landmarks. The 3D landmark reconstruction algorithm simultaneously estimates the shape, pose and position of the face, based only on the fact that all images in the sequence are recorded using a single calibrated camera.

2.7 Deep learning

With the further development of computer technology, neural network approaches for facial recognition have gained renewed interest. Alignment and the representation of the face by

employing explicit 3D face modelling have resulted in improved accuracy of face recognition in unconstrained environments [16][17] [18] [19]

2.8 Facial image comparison

The result of facial image recognition is often the selection of 1 or more target facial images that could be matched with the image of the unknown person. In practice, however, this often leads to hit lists with multiple possible matches to the query image, and the correct target not necessarily on top of the hit list. In such cases, the decision has to be made by a forensic anthropologist or forensic image analyst. Since the previous review, more studies and proficiency tests have been reported on the performance of facial image comparison by lay people and experts, showing that there is a reason for concern, and that better methods and technology are needed. A number of institutes have published documents that describe their procedures for performing facial image comparison. These procedures show that measures are being taken to limit the influence of subjective judgments and that there is a need for quantitative statistical data. The FBI has started a working group in 2009 for facial image comparison that is expected to stimulate the development of better methods and technology (FISWG).

Human and computer performance has been systematically compared as part of face recognition competitions, with results being reported for both still and video imagery. Analysis of cross-modal performance shows that for matching frontal faces in still images, algorithms are consistently superior to humans. For video and difficult still face pairs, humans are superior [20]

People doing facial image comparison can be found in four different kinds of professions: forensic photographers, forensic anthropologists, video investigators and imaging scientists. Knowledge of anatomy and physiology of the face is needed to get a good interpretation of differences and similarities in facial features. Similarities or differences in such images can often be explained by differences in the imaging conditions, pointing to the importance of knowledge about optics. Small facial details can be distorted, and artifacts looking like small details introduced due to noise, pixel sampling and compression, requiring knowledge about image processing for the proper interpretation of observations. Changes in image quality, pose and position, lighting and facial expression greatly influence the comparison process. Therefore, it is strongly recommended that one acquire reference images of the suspect and a number of other people with the same video camera in the same situation under similar lighting conditions. While the techniques of facial image comparison are generally accepted within their practitioner communities, they are not tested, and their error rates are unknown. On that basis, the methods of facial image comparison would appear not to meet the anticipated standards [21]

It is well-established that matching images of unfamiliar faces is rather error prone. Experimental studies on face matching underestimate its difficulty in real-world situations. Photographs of *unfamiliar* faces seem to be unreliable proofs of identity, especially if the ID documents do not use very recent images of the holders [22]

Existing scientific knowledge of face matching accuracy is based almost exclusively, on people without formal training. Human performance curtails accuracy of face recognition systems, potentially reducing benchmark estimates by 50% in operational settings. Mere practice does not attenuate these limits [23], and some training methods may be inadequate [24]. However, large individual differences have been reported, suggesting that improvements in performance could be made by emphasizing personnel selection [25] White et al [26] also have shown that forensic facial examiners outperformed untrained participants and computer algorithms on challenging face matching tests, thereby providing

the first evidence that these examiners are experts at this task. Notably, computationally fusing responses of multiple experts produced near perfect performance.

2.9 Eyewitness identification / Facial composites

In most of the criminal investigations of a crime, one of the first steps is to interview eyewitnesses. In these interviews the witnesses are asked to provide a description of the perpetrators. For investigational purposes this description may be made into an image by a (police) sketch artist. The sketch artist can also help the witness to recall the face of the perpetrator by showing multiples examples of facial features. Instead of sketches, it is also possible to create photo compositions using examples from databases with facial images. As not always images of perpetrators are available, matching of composite sketches with facial photographs (e.g. mugshots) is of interest. Matching performance of composite or forensics sketches against photo galleries are promising but still considerably lower than photo matching performance of commercially available systems [27][28]

2.10 Other biometrics

2.10.1 Ear

Even though current ear detection and recognition systems have reached a certain level of maturity, their success is limited to controlled indoor conditions. In addition to variation in illumination, other open research problems include occlusion due to hair, ear symmetry, earprint forensics, ear classification, and ear individuality [29]. The experimental results show that ear recognition may achieve an average rank-one recognition accuracy of more than 95% [30] Current studies are directed towards more robust automated methods for ear detection, landmark localization and ear recognition using 2D and 3D techniques [31], [32] [33].

2.10.2 Body geometry and gait

With the standardisation of photographs, identification primarily occurs from the face. However, results consistently show that less body measurements are needed to find no duplicates when compared to the face. With the combination of eight body measurements, it is possible to achieve results comparable with fingerprint analysis [34]. Thicker garments produce higher inaccuracies in landmark localisation, but errors decrease as placement is repeated. Overall, comparison to truth reveals that on average statures can be predicted with accuracy in excess of 95% [35]

Also lower leg shape, sometimes the only body part consistently depicted in images, has been reported as “an effective biometric trait” [36]. Recent studies have shown that when face identification fails, people rely on the body but are unaware of doing so [37] Bouchrika et al [38] reported a method to extract gait features for different camera viewpoints achieving an identity recognition rate of 73.6 % processed for 2270 video sequences. Furthermore, experimental results confirmed the potential of the proposed method for identity tracking in real surveillance systems to recognize walking individuals across different views with an average recognition rate of 92.5 % for cross-camera matching for two different non-overlapping views.

Yang [39] describes a method for height estimations on eye measurement through a gate cycle.

2.10.3 Soft biometrics

Soft biometric information extracted from a human body (e.g., height, gender, skin color, hair color, and so on) is ancillary information easily distinguished at a distance but it is not fully distinctive by itself in recognition tasks. However, this soft information can be explicitly fused

with biometric recognition systems to improve the overall recognition when confronting high variability conditions. The use of soft biometric traits is able to improve the performance of face recognition based on sparse representation on real and ideal scenarios by adaptive fusion rules [24]. Depending of the acquisition distance, the discriminative power of the facial regions can change. This results in some cases in better performance than achieved for the full face [40]

Soft biometrics introduce a possibility to automatically search databases based on biometric features obtained from verbal descriptions, resulting in more than 95% identification accuracy [41].

2.10.4 Liveness detection

Spoofing is the act of masquerading as a valid user by falsifying data to gain an illegitimate access. Vulnerability of recognition systems to spoofing attacks (presentation attacks) is still an open security issue in biometrics domain and among all biometric traits. Galbally [42] propose a technique using 25 general image quality features extracted from one image (i.e., the same acquired for authentication purposes) to distinguish between legitimate and impostor samples. The experimental results, obtained on publicly available data sets of fingerprint, iris, and 2D face, show that the proposed method is highly competitive compared with other state-of-the-art approaches and that the analysis of the general image quality of real biometric samples reveals highly valuable information that may be very efficiently used to discriminate them from fake traits. Erdogmus et al [43] studied detection problem of more complex 3D attack types using various texture based countermeasures.

3. Detection of image and video manipulation

Image and video files are changed for numerous reasons with and without a criminal intent. Images are scaled, cropped, rotated and compressed to make them fit for a document or a website. Contrast or colors are changed to enhance the visibility of details. This processing is often referred to as manipulation. However, manipulation could also refer to modification of an image with a criminal intent. One type of modification is a change of the visual content by hiding or inserting visual information in the original image. The other modification is non visual addition of information, like a text message in an image that is published on a website as a means of communication between persons. This modification is referred to as steganography.

A number of clues can be used for detection of manipulation by visual inspection, like discrepancies in lighting, brightness levels, color distributions, edges, noise patterns and compression artifacts in the transitions between the tampered and original parts of the questioned image. Visual inspection could be not feasible anymore when it is not known which region in an image is being questioned. A lot of research was focused on automated detection of regions in an image that might have been tampered with . However, most of the methods that have been published do only produce indications of regions in an image that require inspection by an examiner. The SWGIT group has published a document on detection of manipulation <http://www.swgit.org> .

A special type of detection is based on the indication of ‘resampling’ [44]–[47]. When a part of an image is pasted into another image, it is often necessary to apply rotation and resizing to make them visually fit. This resizing causes a special relationship between color values in the resized region that could be detected.

Double compression detection in JPEGs [48]–[53] is also widely researched, as well as using the Photo Response Non Uniformity (PRNU) for detection [54]

Another type of image tampering is referred to as 'copy-paste' forgery [[46]–[54]]. Objects or persons that are visible against a background with a specific texture, like blue air, green grass, trees, etc. are hidden by pasting a copy of a region in the image with the same texture over them. Detection of this type of tampering looks like a simple straightforward process. All regions in an image have to be compared to each other in order to find regions that are copies. However, the challenge is to limit the number of comparisons and to find a computationally efficient method. This is a requirement when a large amount of images has to be checked. A relatively large number of methods have been proposed in the literature for this task. Also a method with support vector machines (SVM) for object based manipulation by the contour in a video file is proposed, with an accuracy from 70 to 95 % claimed by the authors [55]

A related problem is the detection of illegal copies of image and video files. One technique for protection of original image and video files is the use of watermarking. A watermark is in most cases a hidden mark in the image that will get lost in most common copy processes. Although watermarking is already an old technique there is a lot of research going on [[56]–[74]]. Furthermore on authenticity a survey is published for video[75].

The number of papers published on these topics show that the problem of reliably detecting image tampering has not been solved yet. There are now a number of software packages available that offer a number of methods that can be tried on a questioned image, however interpretation of the results by an expert remains important.

Also anti forensics methods are described based on methods that are published, to prevent tampering from being detected. [76]–[83]

4. Camera identification of images and video

In criminal investigations of child porn production and distribution, identification of the source of a digital image has become very important, because a specific camera, (or a cell phone camera, a webcam, or a flatbed scanner) could be linked to a suspect using other types of evidence. Identification of images that might have a common source can also be helpful in these investigations. The developments that have been started in the period of the previous review have not been stopped and have lead to a number of new methods and software packages. The most used method is based on the estimation of a specific type of fixed pattern noise in an image that is caused by PRNU - *Photo Response Non Uniformity*[84]. The method is also useful in other cases such as murder and fraud to find a links between a camera and images that have been taken [85], [86]

For identification of a specific camera as the source of a specific image, the PRNU patterns have to be estimated from reference images from the camera and the noise that can be filtered out from this specific image. These patterns have to be compared and a similarity measure is used as a measure for the strength of the evidence that the camera is the source. Common practice is to compare the PRNU pattern of a specific image with the PRNU patterns from a large number of camera's. The quality of the estimation of the PRNU pattern from an image depends heavily on the image content and this can be taken into account. However, if there are more images available from the same, unknown source, e.g. the frames in a video file, much better estimations of the PRNU pattern can be obtained by averaging techniques. However several methods are presented to improve the calculation speed as well as clustering images if the camera is not available. Also the use of GPUs is discussed within these methods ([86]–[89]).

Other sources of fixed pattern noise [90]–[92] that have been investigated are based on detection of image artifacts from differences in image processing in the camera chips. If the camera does not work also switching the camera module appears to work for finding a link between the camera module and the images taken[93]

In the forensic practice of a case in which a specific camera has to be identified, a collection of similar cameras from the same brand and type are needed for validation of the results. For using PRNU as evidence, the analyst has to interpret the comparison results. The ENFSI working group for Digital Imaging has conducted two proficiency tests to find out what different experts might report to the court about camera identification. In the practice of investigation of large amounts of images, PRNU is also useful to get indications of possibly common sources. A number of studies have been found on the implementation of this application .

5. Video Image processing and Image Search

The ENFSI worked on a proficiency test of image processing, S-Five <https://s-five.eu/> . The project was for Standardization of Forensic Image and Video Enhancement, and resulted in a best practice manual for processing these. Also a test set with a collaborative exercise and the use of different software products and algorithms have shown that different results are obtained in image enhancement depending on the blur function ([94]–[96].

Several approaches have been published [97] , where also attention is given on logging and archiving processes.

A methodology to event reconstruction has been published by Quentin [98]. They proposed a systematic approach to using trace images in the framework of an investigation. The method is cyclic and iterative. In other publications [99], [100], Quentin provided solutions for image classification, by adding systematic description of image content to metadata as well as timeline chronological reconstruction. For 3D reconstruction using image from fixed and mobile cameras, photogrammetry is proposed.

Several software products have been developed for searching in large amounts of images and video and using similarity detection of images. The software appears to be useful in practice for searching large amounts of image databases, and big multimedia analysis [101]–[103]. The development of deep learning methods.

The development and implementation of deep learning methods

The LASIE project of FP7 funded by the European Commission has published an overview of methods for image search in relation to big data http://www.lasie-project.eu/wp-content/uploads/2015/05/LASIE_D6.1.pdf

Forensic video analysis is also assisted by algorithms, which is published for example by Geronimo [104] on retrieving certain actions, such as walking or fighting and the search for specific persons. Also relevance feedback is important for semi supervised search [105]

The recent deployment of very large scale camera networks with fixed and moving surveillance cameras has led to a novel field of object tracking. Hsu [106]proposes vehicle matching and tracking cross camera by affine and scale invariant feature transform.

5.1 Video games forensics

Another field which is approaching is video games forensics[107]. An investigator might not expect video games and their data files are used as a crime or hide data.

5.2 Video carving

Within the field of video carving several approaches have been made. The software defraser which is partly open source as published by the Netherlands Forensic Institute at <https://sourceforge.net/projects/defraser/> . Since video file formats define only a limited number of mandatory features, they leave room for interpretation. Differences in file structures appear to be available [108].

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Media Evidence

Digital Evidence, 2013-2016

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1. Introduction

This review has drawn on information from many sources. The field is characterised as moving very quickly and, in many ways, too quickly for the publication cycle of refereed journals. There are rapid developments in consumer technology that are quickly exploited by those with criminal intent. In addition, criminals continue to show themselves to be adept at adopting and adapting sophisticated technology.

Due to the accessibility of the technology to consumers and enthusiasts and the high degree of specialisation within information technology/computer science, much of the very useful information for this review came from sources such as news bulletins, enthusiast magazines (mostly online) as well as technology magazines, again, mostly online and Government resourced reference materials, in addition to refereed journals, many of them rapid cycle. There has been a substantial amount of material published. It has not been possible to produce an accessible review that covered every issue that arose over the review period. Selective examples have been chosen, some explored in depth, to provide the reader with a broad sense of the challenges of the field and to provide some guidance to the issues. In this paper, the focus has been on new developments and concepts that have taken on prominence over the past three years. While there have been significant updates in devices, operating systems and forensic tools, to have included all of these evolutionary developments would have, perhaps, made this review inaccessible for some readers.

2. 2013 Future Trends Reviewed

2.1 Cloud Computing and Virtualisation

It was predicted that cloud computing, shared working space and virtualisation would continue to grow. This paper reveals the very significant growth in this sector with a wide range of offerings in a competitive environment now in place including storage, platform and services. This has major impacts on investigative method with ever increasing volumes of data and accessibility to the evidence.

2.2 Anonymous Networks

The battle between those engaging in criminal activity wishing to remain anonymous and law enforcement to expose them did continue. There has been some success by law enforcement to expose some very sophisticated anonymous networks, success achieved through complementarity of good police work, the application of technology and international cooperation. Although there has been an increase in the presence of criminal activity, there have also been some significant investigative successes. Furthermore, some vulnerabilities have been found in some anonymous networks.

2.3 Emerging Technologies

This can be taken as a given. The commercial information and communications technology market continues to grow in breadth and depth. It will continue to challenge digital evidence practitioners and law enforcement investigators into the future.

2.4 Hand-held Devices

The trend of movement towards hand held and mobile devices continues at the expense of desktop and notebook devices. The number of apps available for devices is also continuing to grow.

Although it was anticipated that security features would become standard or at least more prevalent, what was not anticipated was the strength of security that would come as standard on mobile devices. Apple has led the way using encryption that it claims it is unable to decrypt.

3. Useful Statistics

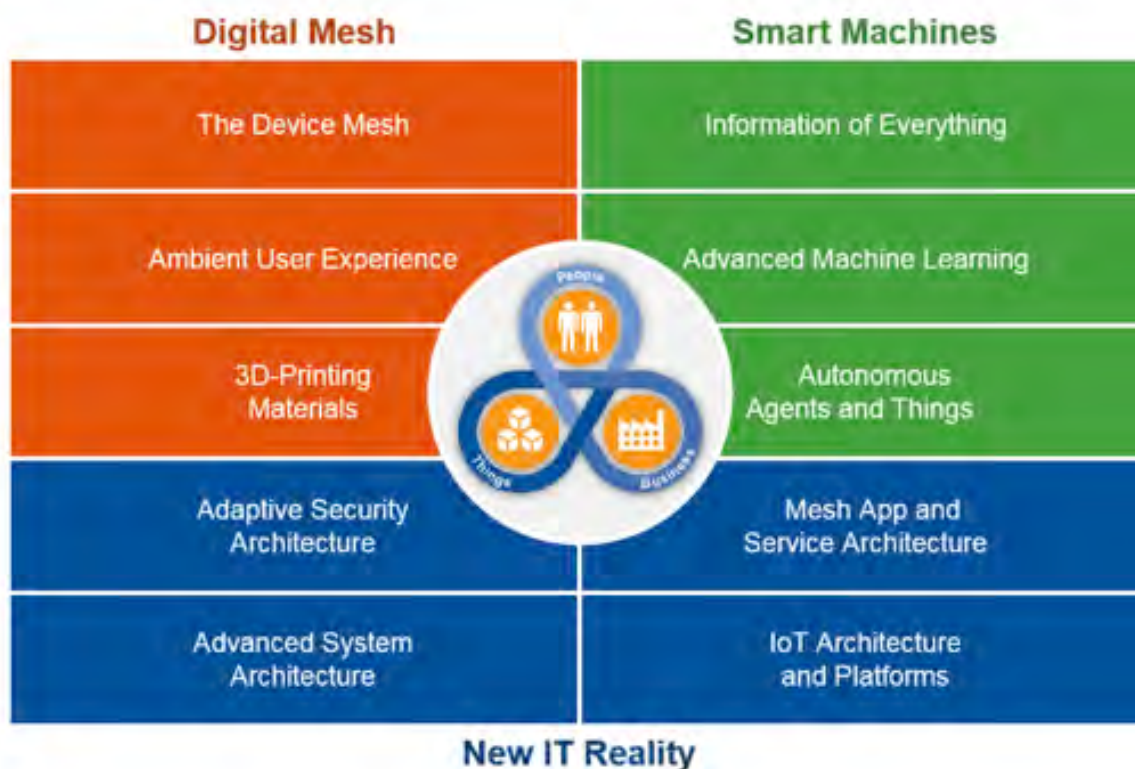
Digital forensics is defined as the computer science and investigative methodology of digital evidence. Digital evidence refers to a repository of electronic data that can be accessed [1]. Proper digital forensics is within bounds of legal guidance, utilizing sound methods of chain of custody, tool validation, repeatable processes, notes, and presentation of evidence [2]. Digital evidence can be on laptops, desktops, mobile devices, networks, virtual and cloud environments. It can also be found within or as images, videos, audio, global positioning systems (GPS), cameras, information and entertainment systems in cars, and social media. The most common areas for storing data are mobile devices but this is changing.

A number of trends and themes are evolving across digital the digital landscape with digital meshing and smart machines becoming more prevalent in the lexicon. A digital mesh is a human-centered theme that refers to the collection of devices (including things), information, apps, services, businesses and other people that exist around the individual. As the mesh evolves, all devices, computer and information resources, businesses, and individuals will be interconnected. The interconnections are dynamic and flexible, changing throughout the day. [3]

The device mesh is an expanding set of endpoints people use to access applications and information, or interact with people, social communities, governments and businesses. The device mesh moves beyond the traditional desktop computer and mobile devices (tablets and smartphones) to encompass the full range of endpoints with which humans might interact. Devices are increasingly connected to back-end systems through various networks, but often operate in isolation from one another. As the device mesh evolves, connection models will expand and increase cooperative interaction between devices.

Smart machines have advanced data analysis technologies and approaches are evolving to create physical and software-based machines that are programmed to learn and adapt, rather than programmed only for a finite set of prescribed actions. As new IT reality themes emerge, new architectures for security, systems, applications and services will be required, as well as platforms to address not only the ongoing mobile computing challenges, but the unique requirements of the Internet of Things (IoT). Unless organizations address these architectural and platform issues, they won't be able to address the opportunities and challenges of the digital mesh and smart machines. With these emerging trends and themes, criminals have at their disposal more sophisticated tools, thus making it more complex for the investigation of digital crimes.

The figure below lists 10 strategic trends include three groupings of complementary trends that are mutually reinforcing with amplified disruptive characteristics:



Worldwide Devices Shipments by Device Type, 2015-2018 (millions of units)

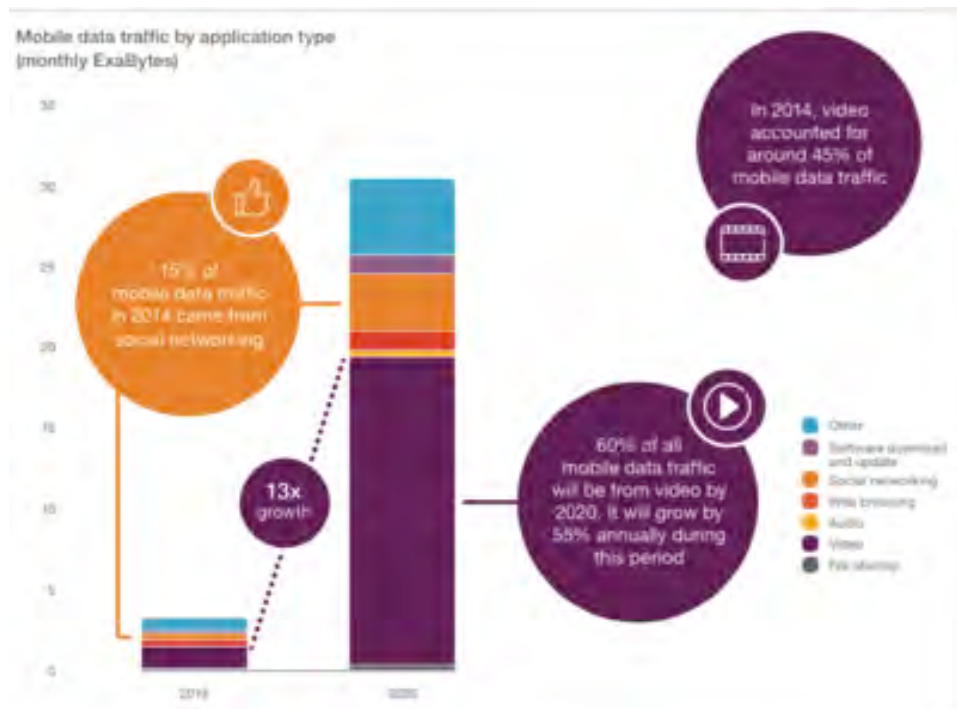
4. Mobile Phones

It is anticipated by the end of 2016, 82 per cent of mobile phones will be smartphones. Worldwide combined shipments of devices (PCs, tablets, ultramobiles and mobile phones) are expected to reach 2.4 billion units in 2016, a 1.9 per cent increase from 2015, according to Gartner, Inc

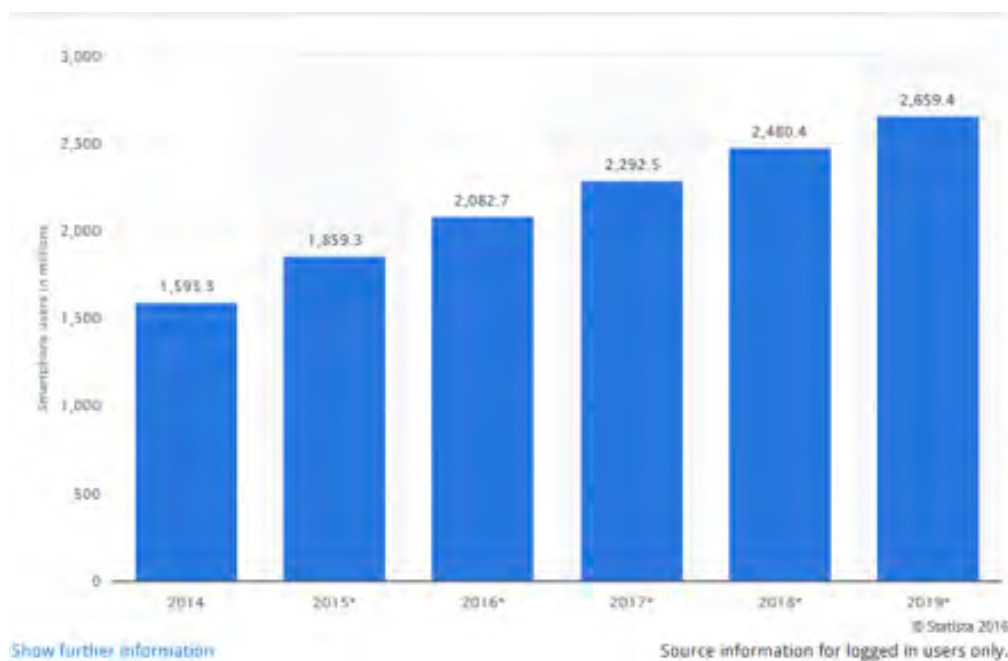
Device Type	2015	2016	2017	2018
PC (Desk-Based and Notebook)	246	232	226	219
Ultramobile	45	55	74	92
PC Market	290	287	299	312
Ultramobiles (basic and Utility)	196	195	196	198
Computing Devices Market	486	482	495	510
Mobile Phone	1,910	1,959	1,983	2,034
Total Devices Market	2,396	2,441	2,478	2,545

Note: The Ultramobile (Premium) category includes devices such as Microsoft's Windows 8 Intel x86 products and Apple's MacBook Air. The Ultramobile (Basic and Utility Tablets) category includes devices such as, iPad, iPad mini, Samsung Galaxy Tab S 10.5, Nexus 7 and Acer Iconia Tab 8.

Ericsson predicts that regions like Asia Pacific, the Middle East and Africa will account for 80 percent of all new subscriptions in the next five years, continuing on with trend that it is already tracking [4].



The statistical chart below illustrates the total number of smartphone users worldwide from 2014 to 2019.



For 2016, the number of smartphone users is forecast to reach 2.08 billion [5].

5. Internet of Things (IoT) Growth

The predominant growing trend in the digital world is the Internet of Things. Although the term was coined decades ago (see later references in this paper) it is now having an impact on the market, the storage, location, movement and usage of data therefore having an increasing impact on investigations involving digital evidence.

Below are identified growth areas of Internet of Things (IoT) between 2013 and 2020 [6]:

- 35.2% Compound Annual Growth Rate (CAGR) will reach an installed base of 25.0 billion units
- In 2020, 8.3 billion 'things' will ship, with more than half of them consumer applications
- The IoT will support total services spending of about \$263 billion in 2020
- By 2018:
 - 20% of all business content will be authored by machines
 - 6 billion connected things will be requesting support
 - 5% of all economic transactions will participate in autonomous software agents outside of human control
 - More than 3 million workers globally will be supervised by a robo boss
 - 20% of smart buildings will have suffered from digital vandalism.
 - 50% of the fastest-growing companies will have fewer employees than instances of smart machines
 - Customer digital assistants will recognize individuals by face and voice across channels and partners
 - 2 million employees will be required to wear health and fitness tracking devices as a condition of employment
- By 2020:
 - 40 % of smart agents will facilitate mobile interactions, and the post-app era will begin to dominate.
 - 95% of cloud security failures will be the customer's fault.

The table below shows shipments of devices by operating system.

Operating System	2014	2015	2016
Android	1,156,111	1,454,760	1,619,030
iOS/Mac OS	262,615	279,425	298,896
Windows	333,017	355,035	393,256
Others	626,358	380,545	261,155
Total	2,378,101	2,469,755	2,572,338

Table1: Worldwide Devices Shipments by Operating Systems (Thousands of Units) [7]

Compared with the corresponding figures from the previous paper three years ago:

Operating System	2012	2013	2014	2017
Android	497,082	860,937	1,069,503	1,468,619
Windows	346,457	354,410	397,533	570,937
iOS/macOS	212,899	293,428	359,483	504,147
RIM	34,722	31,253	27,150	24,121
Others	1,122,213	871,718	702,786	396,959
Total	2,213,373	2,411,796	2,556,455	2,964,783

Table 2: Worldwide Devices Shipments by Operating Systems (Thousands of Units) [8]

Note that Apple's iOS and Mac OS have replaced Windows in second place as the preferred operating system; and that RIM is no longer referred to directly.

6. Today's Consumer and Business Technology

6.1 Cloud Services

The fifth annual report from Cisco [9] states that:

- Annual global data centre IP traffic was 3.4 zettabytes per year in 2014.
- This is forecast to grow to 10.4 zettabytes by the end of 2019 [10].
- 86% of workloads will be processed by cloud data centres and 14% by traditional data centres.
- 30% of cloud workloads were in public data centres that will grow to 56% by 2019. The balance will be in private data centres.
- Annual global cloud traffic in 2014 was 2.1 ZB in 2014 and is expected to grow to 8.6 ZB by the end of 2019. Global cloud IP traffic will more than quadruple from 2014 to 2019.
- Software as a service (SaaS) accounted for 45% of total cloud workloads in 2014 and is expected to grow to 59% by 2019.
- The Internet of Everything accounted for 134.5 ZB of created data in 2014 and is forecast to grow to 507.5 ZB by 2019. The data created by the Internet of Everything devices will be 269 times higher than the amount of data from end-user devices.
- In 2014, 1.1 billion consumer users used cloud internet personal storage which is expected to grow to two billion users by 2019.

Multiple device Connections per User:

Region	2014	2019
North America	7.3	13.6
Western Europe	5.5	9.9
Middle East and Africa	5.4	6.2
Latin America	4.7	5.2
Central and Eastern Europe	4.5	5

Region	2014	2019
Asia Pacific	4.5	5

There is wide spread belief that cloud services are inherently insecure or at least carry security risks and several authors in this review allude to this. There is, however, no evidence presented that supports this assumption. Contrary to this belief, the cloud providers themselves attest to their security and many cloud service providers offer tiered levels of security although this is not the forum for such a discussion. Several Government agencies that work in the defence and intelligence space, agencies that demand the highest levels of security, in the United States and other countries use cloud services. The business model of the cloud service providers is built and dependent on the confidence of their clients who require this security. Logic therefore leads one to assume that the levels of security are sufficient to satisfy their clients who we can assume are well informed.

The security of cloud storage can be considered as a series of layers: 1) system infrastructure, 2) core middleware, user-level middleware, and 4) user applications [11].

1. The system infrastructure comprises the physical infrastructure that may be located in one or more jurisdictions.
2. The core middleware includes virtualisation technology manages the resources as a collection of virtual machines. This will include encryption protocols that protect the data at rest and in transit
3. User-level middleware includes the cloud computing environment and the tools provided by cloud service providers to integrate developer programs. The providers are responsible for providing access control to the data.
4. The user application layer is the responsibility of the cloud service users who are responsible for configuring and controlling administrative roles.

7. The Internet of Things

The Internet of Things (IoT) is a global distributed network connecting physical objects that are capable of sensing or acting on their environment and able to communicate with each other, other machines or computers. These objects can include simple items with embedded sensors such as household appliances, industrial robots, cars, trains and wearable technology [12]. It can also include the management and control of buildings, such as the heating and cooling, lighting and security systems; or for traffic control in cities. The growth of the IoT is driven by business need. Analysis of the collected data produces insights and identifies opportunities for improvements including cost efficiencies and safety. It is estimated that the IoT will comprise 25 billion connected objects by 2020. This rapid growth is assisted by the rapidly falling price of sensor and Radio Frequency Identification (RFID) technology and the greater coverage and availability of wireless and mobile networks. In addition Internet Protocol version 6 supports sufficient IP addresses for 3.4 x 10³⁸ internet-connected devices [13]. This presents a challenge to law enforcement investigations as it can allow for the rapid and frequent change of IP addresses for nefarious activities. Identifying the IP address of a device involved in criminal activity, whether it is as the suspect, victim or witness, is key in the investigation process.

There are a number of security risks to consumers and businesses. These include access to sensitive data through unauthorised access to devices or interception of communications; or, access to servers or cloud services aggregating large amounts of data. Hackers could take control of systems or networks to cause disruption as an act of mischief or for ransom. This might include a power grid, a personal health device or a motor vehicle. A high

proportion of devices themselves will lack the power and physical size to provide the level of security that would normally be expected leaving the data more vulnerable [14].

As the IoT comprises the broad categories of cloud, virtualisation, mobiles devices, fixed computing, sensor and RFID technologies, and artificial intelligence, forensic examination will require forensics in all of these categories. Digital forensic processes that are applied to large data investigations can be expected to be applied to investigations involving the IoT. As the IoT evolves and develops, additional devices including new and unfamiliar special purpose devices capturing, processing and transmitting data will be contribute data to an investigation that has a probative value [15, 16].

Conventional digital forensics recommends that devices should not be turned off if they are running when encountered by first responders in order to preserve modified, created and accessed times of files [17, 18]. As the standards that applied to digital evidence investigations which the emerging cloud computing environment, the same holds true with the greater complexity encountered in IoT investigations. Devices left running at the scene will consume power and may overwrite stored data due to restricted storage capacities. Consideration needs to be given to switching off power or to leave them running [19].

The proprietary formats, protocols and physical interfaces all add further challenge to IoT investigations and complicate the extraction of evidence. Some systems distribute data to adjacent nodes or incorporate cloud services. An appropriate investigation plan might be access information from other nodes, base stations or cloud services [20].

The IoT provides investigators which a richer context of information concerning events that have taken place in the physical world. It is an ambient intelligence in which the environment is reacting to the user's requirements without any conscious interaction by the user. In fact, perpetrators will be oblivious to the incriminating information that can be recorded.

When considering the four phases of the digital forensic investigations process (Identification; Preservation; Analysis; and Presentation) [21], each phase must be considered within the new paradigm.

When performing the search and seizure of the evidence, it is not always immediately apparent where the evidential data is being stored or where it came from. Further, the identification of the user's data can be even more unclear than in 'terrestrial' computing and cloud computing environments, which themselves, were challenging at times.

One potential solution is the integration of IoT device data into Building Information Modelling which is a digital representation of the physical and functional characteristics of a facility. This process can assist in answering the questions of the source, location, format and encoding of the information. It would assist in reducing the data set from which the examiner is trying to identify the probative evidence; and designate the nature of the data. When considering the preservation of evidence, the complexity is greater in IoT than it is in terrestrial forensics or cloud forensics. Improvements to the security of operating systems began when malware writers began reducing the evidence of their presence on the victim's hard drive and started exploiting the Random Access Memory for the storage of information. Extracting process memory became more difficult and forensic examiners began developing methods for recovering process memory [22].

Once the evidence has been identified, there remains a serious challenge with preservation as it is possible or even probable that the data of interest is overwritten and/or compressed if

the devices do not interact with the cloud to store their data and the amount of data generated exceeds the storage capacity of the device itself [23].

The data must be reliable and trustworthy to be of probative value, therefore its origin must be demonstrated beyond doubt. Aggregation of the data and loss due to compression may alter the data to such a degree that it is difficult to demonstrate a linear link between the data and its context. This can cause serious challenges to the presentation of evidence.

The volume of data comprising the IoT domain will double every two years from 2012 until 2020 to reach an estimate of 40,000 exabytes from a base of 130 exabytes in 2005 [24]. This will mean that time and resources spent finding evidence in this greater volume, then putting it into a form that can be understood will be challenging. Digital forensic investigators will need to find efficient and effective ways to collect all relevant evidence from the various devices that has potential travelled through different networks leaving a digital trail concerning a particular incident or investigation [25].

Furthermore, IoT devices can be unreliable as the data can be changed without human input and before it is acquired by investigators. The devices can learn and adjust to the situation based on sensory input [26].

To conduct forensic investigations involving the IoT, established digital forensic knowledge, methods and techniques will need to be built upon recognising that IoT forensics is different to terrestrial forensics and even cloud forensics. Methods and tools will need to be developed and employed that assure the acquisition of all relevant evidence in a time efficient manner. At the same time, the evidence must be forensically sound so that it can be exhibited providing the court with sufficient comfort in its veracity. It must also take into account that the evidence will most likely need to be obtained in parallel with the incident response in all affected networks and devices without modification of the evidence. The authors [27] are proponents of an automated response in which the incident, presumably on the assumption that it is a cybersecurity incident, should be identified as soon as it occurs and trigger an automatic incident response. An automatic forensic collection should be activated and the data pre-analysed. This automated response does not necessarily hold true for investigations that do not involve a cybersecurity incident, but is actually an investigation into a crime of some other nature.

8. Recent Research on Wireless Body Area Networks: A Survey

Major developments in healthcare systems are now occurring in the field of Wireless Body Area Network. It is fundamentally changing the delivery of healthcare systems, for example, through Electrocardiography (ECG), Insulin Pumps, Pacemakers, Implantable Cardioverter Defibrillators (ICD), temperature and pulse sensors. The objective is to gather and analyse physiological and behavioural patient data and deliver to the professional care givers. The sensor nodes can be placed in or on the patient's body or clothing [29].

There are many different protocols designed to facilitate Wireless Body Area Networks as would be expected in a competitive market. Each affliction being monitored lends itself to one or more protocols over others. There are also many variables that can be altered to optimise performance. There are potential issues of significance for digital evidence and investigation, for example, interference and eavesdropping.

8.1 Cloud Apps

Daryabar et al (2016) examined for the presence of artefacts on Android and iOS devices when using four popular cloud storage apps (OneDrive, Box, GoogleDrive and Dropbox).

The researchers were able to demonstrate that various artefacts associated with the use of cloud applications could be recovered from the Android device's internal storage. These included filenames and file data remnants associated with OneDrive and Box apps. On the iOS device, other information associated with the cloud apps, such as product names, device names, application installation locations and last back up time stamps remained on the device. Depending on the app under investigation, differing types and quanta of information were found. For example, log in information could be found for GoogleDrive, whereas with Box, no information concerning cloud activity could be found. For Dropbox, login activity, upload, download, delete and share activities could be found. All apps revealed URLs and IP addresses used, IP addresses, timestamps, server names and certification services providers.

8.2 Banking Apps

The banking apps for Android studied by (Chanajitt et al 2016) [30] are hybrid apps. Therefore security needs to be assured at both the device level and at the external storage level. Inadequate security can allow the exploitation of data and extraction of funds via the hybrid apps. A significant number of mobile banking users use smart phones to transact and the availability of real time financial information influences behaviour regarding purchasing, making payments and other financial decisions.

Several previous studies cited by found artefact and remnant information from several apps on the devices that can be used to reconstruct events. Moreover, other studies cited it was, theoretically, relatively easy to highjack banking transactions to divert funds from where the user intended.

The study by examined seven banking apps in Thailand on two different Android devices. Across the apps, they were able to recover sensitive user data including bank account number, account type, account balance, citizen identification, date of birth, thumbnails of banking transactions, PIN codes and SMS messages that verified prior transactions. In addition, some of the apps do not encrypt user data. Further, some of the apps could be repackaged and reinstalled that could then intercept and obtain sensitive information.

9. Cyber Crime

9.1 Cybercrime Prevalence

The UK National Crime Agency and Strategic Cyber Industry Group recently identified that cybercrime accounts for 53% of all crime in the UK [31, 32]. The UK Office for National Statistics estimated there were 2.46 million cyber incidents and 2.11 million victims of cybercrime in 2015.

The cyber criminal market where expertise and cybercrime tools are developed are available for a price that can be purchased by those less skilled to conduct their criminal activity. Despite these figures, the National crime Authority believes that there is a significant degree of under reporting.

9.2 Cyber-bullying

Cyber-bullying has been recognised since the early 1990's and is describes an intentional act that uses information and communications technologies against a victim. The acts can be perpetrated through SMS, mobile phone, cyber-stalking and cyber-grooming using a range of tools including email, chat, instant messaging, social media [33].

The authors describe a hypothetical case in which a 13 year old victim of cyber bullying eventually succumbed to taunts and suggestions that she kill herself. It is contended that a generic mobile digital forensic readiness solution used to monitor classmates devices could have mitigated the bullying. Forensic readiness is to be in a position to maximise the use of potential digital evidence while minimising the cost of the investigation. It is the collection of evidence in real time before the incidence is identified, which is a process defined by ISO/IEC 27043:2015 before incident identification.

The authors argue that there is some legal precedence for content monitoring on mobile devices as courts have found against organisations that should have been aware of flagrant abuse that was occurring. This is made easier as the suggestion is that this is for parents who want control over what their children are exposed to.

A mobile agent is used to monitor the mobile user's activities and transmit them to a secure storage for analysis. If the analysis reveals nefarious intent, an alert is generated and early intervention can be actioned.

9.3 Drugs

The previous paper [34] included a description of Silk Road, the Internet site on the Darknet that can only be accessed via Tor and will accept Bitcoin transactions for the purchase of goods. It was a site where illicit drugs, firearms, child abuse material and other illegal goods and services could be purchased anonymously. In October, within days of the presentation of the 2013 version of this paper at the Interpol International Forensic Science Managers Symposium, Silk Road was taken down. As anticipated, Silk Road was replaced and there are now approximately 50 cryptomarkets, also known as 'dark net markets' or DNM, and vendor stores selling similar goods and services to those previously offered by Silk Road (Kruithof, K. et al 2016) [35, 36].

Of the eight most popular marketplaces, the vast majority of the listings were for drugs including the majority for cannabis derivatives, stimulants and ecstasy analogues. It was also found that a significant proportion of the listings were for larger amounts implying that many dealers were using the online markets as their supplier for later offline sale. It is estimated that cryptomarkets generate total monthly revenue of \$12m to \$21m for January 2016. Several million dollars more of revenue can be included if prescription drugs, alcohol and tobacco are included. Most vendors are in the United States (890) followed by the United Kingdom and Germany. Intercontinental trade is common with many vendors shipping to Australia and New Zealand.

It is noted that as security is honed, increasing numbers of buyers and sellers are feeling confident in evading detection. It is not just on the Darknet where drugs are available, but also numerous web shops are easily found by search engines where designer drugs, labelled as 'research chemicals' can also be purchased.

Kruithof et al (2016) identified four broad modes of detection and intervention. These were traditional techniques in the drug chain, postal detection and interception, online detection and online disruption.

9.4 Prostitution

Prostitution is thriving through the use of the internet for marketing to new clients and avoiding detection by law enforcement. The anonymity provided by new technologies and websites are easily exploited by the sex trade. Further, targeted enforcement of the virtual world appears to have limited potential. The purveyors of sex use coded language comprising hidden meanings on covert web sites but also on legitimate websites [37]. The

researchers from Michigan State and Loyola University Chicago (Finn, M. and Henion, A. 2016) [38] found that 80% of all sales of sex now occur online. Purveyors have also developed apps that assist in locating a prostitute nearby by the client typing in his address. Notably law enforcement is only able to target the most egregious cases of exploitation with a primary focus on sex trafficking and minors.

9.5 Terrorism

Terrorist groups continue to exploit cyber technology to further its aims. This includes many different platforms.

Following criticism for allegedly not doing enough to reduce extremist speech, Twitter claimed to have removed 235,000 accounts linked to terrorism over the six months to August 2016 and a total of 360,000 since the middle of 2015 (Kravets, D. 2016) [39]. The challenge is the absence of a “magic algorithm” for identifying terrorist content.

Currently under consideration is legislation requiring Internet companies to report suspected terrorist activities to the Government (Kravets, D 2016) [39].

9.6 Corruption

The widely publicised ‘Panama Papers’ arose from a leak to the International Consortium of Investigative Journalists. Together with more than 100 media partners, 11.5 million documents exposed the offshore holdings of world political leaders, links to global scandals and the hidden financial dealings of fraudsters, drug traffickers, and prominent persons in sports and other celebrities. The firm at the centre of the investigation creates shell companies and corporate structures that are used to ownership of assets. The data includes emails, financial spreadsheets and corporate records totalling 2.6 terabytes. Using collaborative platforms, 400 journalists across 80 countries working in 25 languages indexed, organised and analysed the data [40].

9.7 Environmental Fraud

The highly publicised scandal concerning the circumventing of emissions regulations for diesel powered vehicles sold by Volkswagen Group emerged in September 2015. Illegal software was installed on Volkswagen motor vehicles that could detect when they were being tested and change their performance to improve results. Volkswagen has admitted to 11,000,000 cars worldwide having the illegal software installed. The fines to which Volkswagen might be liable could potentially reach \$18 billion (Hotten, R. 2015) [41].

9.8 Copyright Infringement

Copyright infringement continues to be very popular among cybercriminals. The world’s largest BitTorrent distribution site, KickassTorrents, operates by providing .torrent and .magnet links so that users can download unauthorised copies of copyright material. The site does not host the infringing material itself. It is estimated to have distributed over \$1 billion of copyright material. The administrator of the site was arrested in July 2016. To evade law enforcement he moved domains around the world. When eventually caught, he was arrested on charges of criminal copyright infringement, conspiracy to commit money laundering and conspiracy to commit criminal copyright infringement. His advertising revenue is estimated at \$16 million per year (Farivar, C. 2016) [42].

9.9 Revenge Pornography

The newly emerged and legislated crime of involuntary or revenge pornography is becoming better known. Dedicated websites that publish intimate images of, often, spurned lovers have been shut down and the administrators prosecuted. The impact on victims can be severe with careers destroyed, families torn apart and has led to suicide. Legislation that

prohibited the involuntary posting of intimate images has been available to law enforcement in the United Kingdom was strengthened in 2015 [43]. By July 2014, 34 states within the United States have legislation making involuntary pornography a punishable crime. A bill was introduced to Congress to also make it a federal crime. The administrator of UGotPosted.com was sentenced to 18 years imprisonment after being convicted under Californian legislation [44].

10. Criminal Craft and Anti-Forensics

10.1 SQL Injection

The security of dynamic pages of web applications can be vulnerable to SQL injection attacks by exploiting the database layers of the application. Poorly designed web applications are especially vulnerable and most commonly result in privilege escalation and unauthorised access.

10.2 Cryptography and Steganography

In an inherently insecure network environment, cryptography and steganography have long been used to secure data communications. Cryptography is a well established method to encrypt data and there are any methods employed. It is always clear to an intermediary, for example a digital evidence examiner, that encryption has been used. Cryptography is concerned with hiding the contents of the message.

Steganography is used to hide a message in a cover image. The receiver is able to extract the message from the image using a secret key. It is not apparent to the intermediary that a hidden message may be present. Steganography is concerned with hiding the existence of the message. There are many different steganography methods available today. By embedding an encrypted message inside an image, the message is doubly protected. There are over 100 different steganography tools using a variety of algorithms. The simplest and most commonly used one is the hidden message is inserted into the coding for the pixels where it have least bearing on the colour. When coding for colour, each pixel comprises 8 bits that together total between 0 and 255. The colours are coded as follows, the position of each number representing red, green and blue respectively:

- Black (0, 0, 0)
- White (255, 255, 255)
- Red (255, 0, 0)
- Green (0, 255, 0)
- Blue (0, 0, 255)
- Yellow (255, 255, 0)
- Cyan (0, 255, 255)
- Magenta (255, 0, 255)

For a demonstration of colour coding, see RGB Color Code Chart [48].

Several methods have been developed to employ multiple security protocols to hide the message. A method is proposed to encrypt data with a new algorithm and then embed it in the image [49]. The researchers used the Affine algorithm to encrypt the data. The encrypted message is then subjected to the Hash based Least Significant Bit (H-LSB) method which is the most popular steganography method. Using this method, eight bits of encrypted data are embedded in the least significant bit of the red, blue green pixel values which will not noticeably impact on the quality of the image. This is repeated until the entire message is embedded in the image. When the receiver is removing the encrypted message,

the embedded data is sequentially removed in the order that it was embedded. Once complete, the encrypted message is decrypted using the Affine cypher key [50].

Researchers in Ghana have used similar concepts of embedding encrypted messages in video using steganography and then employing lossless compression [51]. Lossless compression provides for a reduction in data size without loss of data. Using video to securely hide data provides for better confidentiality and data recovery. The video stream is a series of images and sounds and any changes by embedding data should be visually undetectable. There are various methods of lossless compression and the researchers used Huffman code compression, first published in 1952 [52]. This method can increase the volume of text that is to be hidden.

There are ten steps to complete the exercise:

1. Encrypt the message using an encryption algorithm
2. Compress the encrypted message using lossless data compression
3. Convert the video into frames
4. Embed the compressed, encrypted file into the video frames
5. Reconstruct the converted frames
6. Transmit the video over (untrusted) communication channels
7. Receiver separates the video into individual frames
8. Extract the secret file from the frames
9. Decompress the secret file
10. Decrypt the secret, encrypted file

The researchers evaluated the performance of the method. Robustness was tested by comparing the signal to noise ratio of the original video to that of the altered video. Capacity was tested by the payload capacity or quantity of data that could be hidden without statistically significant deterioration of video quality. Finally security is a measurement of the ability of a third person to detect hidden information in the video.

Testing a range of videos found that the system had very high security, the bit error rate was minimal and change in pixels between the original and received videos was insignificant [53].

Other researchers submit that many of the steganography techniques caused distortions in the images therefore making them vulnerable to detection. They propose methods that scatter the secret message code into randomised pixels or pixels where the brightness changes sharply such as edge pixels. Most of the focus in other papers has been on English or similar languages. Many languages other than English comprise characters that do not have ASCII (American Standard Code for Information Exchange) values (see ASCII Tables for the 256 characters represented by ASCII code [54]). Characters of other languages are represented by Unicode [55]. Once the font of another language is converted to Unicode format, the Unicode characters are converted to binary bits that are then encrypted using AUS-128 bit key encryption [56]. As in other approaches, the video file is converted into video frames. Each video frame is then subjected Canny Edge Detection to divide the image into edge and no edge pixel bits.

The secret message bits are then embedded in the image. If the pixel is a non-edge pixel, then the image is hidden using the identical match technique, ie finding a pixel that has the same value as that of the secret message, then store that position where it will match the secret message bits. The effectiveness of the technique was evaluated using peak signal to noise ratio, mean square error, bit error rate and histogram metrics. Identical match and

edge detection techniques provides the least distortion in the cover video frames. The combination of LSB approach, edge detection and identical match the payload capacity is very high and the distortion in is not visible to the human eye [57].

11. Cryptocurrency

The previous paper and this current one have made considerable mention of Bitcoin. The status of Bitcoin is still a matter of conjecture, ie is it a currency or not? A 2016 ruling in Florida found that Bitcoin is not money as defined under state law (Farivar, C. 2016) [58].

The Australian Taxation Office has determined that crypto-currency, specifically Bitcoins, is similar to a barter arrangement, it is neither money nor foreign currency. It is not a financial supply for goods and services tax purposes but it is an asset for capital gains tax (Australian Taxation Office, 2014) [59]. The Australian Government recently stated its intention to develop legislation to remove what is effectively double taxation of Bitcoin transactions [60]. The United Kingdom put Bitcoin outside the scope of VAT in 2014 (HM Revenue and Customs 2014) [61].

12. Anonymous Networks

The dark web and the various names by which it is known is still the place where the illicit market places choose to do business. The most famous of the anonymous networks is Tor with its notoriety coming to the fore with that of the Silk Road. The Tor network is only as safe as the volunteers whose computers form the individual elements of the network. In July 2016, researchers found that at least 110 Tor machines were actively snooping on dark web sites.

The snooping allowed the indexing hidden services and attack them. Some tried to attack hidden services using SQL injection, Cross-site scripting, user enumeration and other methods. Several of the snooping machines also served as exit nodes so were able to view unencrypted traffic. More than 70% of the snooping hidden services directories were hosted on cloud services making it hard for most outsiders to identify the operators (Goodin, D. 2016) [62].

13. Investigations

13.1 Policy Frameworks

The recurring theme for this series of papers on Digital Evidence prepared for the past several Interpol International Forensic Science Managers Symposia is the rapid change and development in the field. It seems that each edition of this paper, prepared every three years, notes the rapid cycle strategic response required by law enforcement agencies to maintain a competitive edge in the investigation of criminal activity within their jurisdictions. These adjustments are noted against a background of falling crime rates and generally shrinking law enforcement agency budgets, both phenomena being notable in wealthy western democracies. This scenario presents a challenge for those responsible for allocating resources within law enforcement agencies. Resource allocation considerations must also take into account the high cost of building and maintaining a digital evidence capability in the costs of highly educated people with the necessary technical knowledge and skills; attracting such suitable candidates in a competitive market; maintaining a contemporary knowledge in a rapidly changing technological consumer and business markets through training and education programs; and purchasing suitable tools and equipment.

In 2014, the then Association of Chief Police Officers appointed Chief Constable Stephen Kavanagh to the position of National Policing Lead for digital Investigation and Intelligence (DII) [63]. As referred elsewhere in this paper, cybercrime represents 53% of all crime in the United Kingdom [64, 65]. The College of Policing, the National Crime Authority and the National Police Chief's Council developed a framework that focussed on three areas to address the contemporary challenges of Digital Investigation and Intelligence. The framework centred on three recommendations; 1) accelerating digital investigation and intelligence capabilities; 2) governance for a digital initiative; and 3) driving the digital transformation.

14. Victimology

The underreporting of cyber crime is a noted phenomenon and must improve to assist law enforcement. It was noted in the National Crime Authority's report [66]. Apart from the impacts of a range of activities that should be undertaken by businesses, underreporting is considered to be a serious problem. The causes of underreporting are for a number of reasons, including:

- Lack of awareness by the business
- The IT teams fearing retribution from senior management
- Senior management and boards wishing to quietly settle losses
- Counsel advice that it is not in the company's best interests to report

The flow on impact is that underreporting can lead to under investigation that can affect crime fighting but also corporate risk management. It can impact strategic responses to crime fighting and potentially delay the development, modification and implementation of law enforcement responses.

15. Smart Phone Interception and Spying

The data stored on smart phones comprises three broad types: 1) user generated information including images, audio, video, maps, GPS, stored documents, voice mails and connected computers; 2) internet related information including bank accounts, shopping and subscription accounts, internet access and content and social networking; and 3) apps including alternative communication protocols. In short, the smart phone is the most extensive single source of the thoughts and actions of an individual. Blacklisted mobile devices or SIM cards can be monitored and the information can be obtained from the mobile switching centre without user consent.

Smart phones can be attacked by various means using the communication channels. These can include a stimulus to malware already installed on the phone, spreading malware across previously uninfected devices, transferring data held by the phone among others.

Due to the availability of multitude of smart phone sensors (Audio Sensor, Optical sensor, Magnetometer and accelerometer etc.), sensor based attacks can be carried out to influence a large number of infected devices. These attacks are covert and therefore are difficult to prevent, especially if covert and steganography channels are used. Within this category, the types of attacks can include distributed denial of service, annoyance, embarrassment, safety hazards, interference and distraction attacks. Smart phones can also be attacked using the signalling channels.

The authors propose using the cellular infrastructure to initiate data transfer from the smart phone to law enforcement agencies. This would allow security agencies to remain ahead of

criminals and increase the prosecution rate. It is, however, reliant on collaboration between smart phone manufacturers, telecommunications providers and law enforcement agencies. Most importantly, there would need to be general acceptance by the community that their individual privacy will be lessened [67].

16. Child Exploitation

An investigative method that is being increasingly used is for police to take over a child abuse website. It is believed that FBI was the first law enforcement agency when it identified 135 cases. Queensland police took over a child abuse website called 'The Love Zone', located on the Tor network, following the arrest of the owner. They have identified 30 US IP addresses.

A hyperlink is made available to a potential user. If the user chose to open the file, a video containing images of child exploitation began to play. The video file is configured to open an internet connection outside of Tor so that the IP address becomes visible and can be captured (Farivar, C. 2016) [68].

17. Identity Theft

Identity theft continues to be a significant area of criminal activity. Multiple persons were prosecuted by the Internal Revenue Service [69].

18. Skimmers and Sale of Stolen Data

Point of sale skimmers, ATM skimmers and pocket skimmers have become a significant line of business in Chinese black markets [70]. They are becoming quite sophisticated with some even having an SMS notification function.

19. Forensic Analysis

19.1 Phone Forensics

There are broadly two ways to acquire evidence from a phone, the physical and the logical acquisition. The physical acquisition retrieves data directly from the device's physical storage media such as the memory chip or flash storage by means of a bit-by-bit exact copy. There are three approaches for undertaking a physical acquisition including physical chip extraction, JTAG and bootloader.

The logical approach to acquisition is an image creation process facilitated by a copy of the logical objects such as files and folders from the data storage volumes. The weakness of this approach is that it only acquires logically allocated data, not a raw image and generally does not recover deleted data. Also, this approach requires that the phone is switched on and the operating system loaded. It has the potential to overwrite data stored in flash memory.

The physical acquisition will usually obtain a more complete data set from the phone than the logical one and more likely to include deleted data.

When examining Windows phones, such as the Nokia Lumina 625 and 735 models that are commonly connected to cloud services, the ability to acquire data was limited. One of the tools used

19.2 Data Reduction and Data Mining

There has been much discussion concerning alternative approaches that address the challenge of growing data volumes for the past decade or more. But there has been little published research into a framework or method to apply data mining techniques or other methods to reduce and analyse the increasing volume of data. Quick and Choo [71] published their research into a data reduction method to reduce storage demands and a more efficient forensic data subset collection process.

Facilitated by increasing storage capacities and the falling prices of storage devices, the exponentially growing volume of data is regarded by many commentators as the single greatest challenge to digital evidence. These observations are supported by Moore's Law (the number of transistors on an integrated circuit doubling every 18-24 months) and Kryder's Law (storage capacity doubles every 12 months) which, inferentially, means there is an ever widening gap between the generation of stored data and the ability to process it [72]. There are substantial cost implications to storing all this data although there are benefits to be gained from comparison between cases if the data is stored on a networked solution.

The framework proposed by Quick and Choo does not replace the need for full analysis nor is it consistent with the thorough analysis frameworks recommended by authorities such as the Association of Chief Police Officers [73], the European Union Agency for Network and Information Security [74] and the National Institutes of Justice [75]. The usually forensically sound method of data acquisition is applied. Forensic tools display and select files of interest that are then preserved in their own container. The size of the preserved data can be substantially smaller than when every bit of data is copied. This approach can lead to the production of a report that no longer requires a full forensic image of every item seized.

To compare cases for intelligence purposes or conspiracy investigations, similar data from the various cases under investigation should be collected. A full image of the seized item can be made at a later time if necessary. This also has applicability to investigations where data is stored in the cloud where the data of likely probative value can be identified more quickly than to copy a full image.

The impact on storage is significant. The researchers surmised that applying the savings in storage capacity to the FBI seized data holdings from 2003 to 2012, the total data currently held (20 petabytes) could be reduced to a subset of 4 terabytes. This would result in huge cost savings for storage and searching the data across cases would also be much faster. Further, a reduced subset could be searched while a full forensic image is being completed. They give the example of a subset image taking 79 seconds to complete from a 320 GB hard drive compared with three hours to complete a full forensic image and another three hours to verify the copy.

This principle can be applied in a triage manner. If the subset reveals relevant information on certain items, then the focus of analysis can be on those items. For example, if internet history and registry files reveal items of interest, then further attention can be given to the full disk image. If nothing of interest is identified, then the item can be set aside for later examination following attention being given to items likely to provide higher priority evidence [76].

19.3 Portable Web Browser Artifacts

The common web browsers (Google Chrome, Mozilla Firefox, Microsoft) can be operated in private browsing mode to restrict the browser from storing the web browsing history. This function can be impaired by using third party software to retrieve the history. An alternative to private browsing mode can be to use a portable web browser that is installed on a removable drive such as a USB drive. The user plugs the USB to a computer with Internet

connectivity. The browsing sessions are then stored on the USB and not on the computer. The portable web browsers are a challenge for the examination of the suspect's Internet activities as the stored history is not on the suspect's hard drive.

The evidence can be found in the browser includes the web browser history, cache, cookies, preferences and registry. A user will use private browsing to conceal evidence of their activity. Private browsing mode, when used in Google Chrome, Mozilla Firefox and Microsoft Internet Explorer, will leave behind artefacts of the browsing session in the form of deleted files or pagefile.sys file on the hard drive.

Running a memory leaking program can pull artefacts from private browsing sessions to the memory. Theoretically, when using a portable web browser, the browsing artefacts are stored in the installation folder of the removable media.

Using a combination of live analysis, offline analysis, trace analysis and word searching, the researchers were able to look for evidence of a mobile browsing session that may have left artefacts on the hard drive. It was found that the portable web browser leaves forensic artefacts on the hard drive in live and offline modes.

The researchers did not identify which versions of the three browsers they were using [77]. Microsoft released Internet Explorer 11 in October 2013 [78].

20. VoIP Decoding

Voice over Internet Protocol (VoIP) services and becoming increasingly popular with expectations that there will be one billion VoIP users by 2017, ie three times more than the estimated number of users in 2013 (Pokharel et al 2016) [79]. This is in part driven by the ability to make cheap or free calls over existing telecommunications infrastructure.

RTP (Real-time Transport Protocol) is a network protocol for delivering audio and video over IP networks. It is used in communication and entertainment systems that involve streaming media, such as telephony, video and teleconference applications, television services and web-based push-to-talk features (Wikipedia, Real-time Transport Protocol) [80]. RTP is increasingly being used in VoIP applications such as Skype, Facebook messenger, WhatsApp and Google Hangout. Use of encrypted RTP requires additional memory, processing power and bandwidth. As a work around, the apps encrypt the signalling control packets rather than the RTP data.

The challenge for interception and forensic analysis of VoIP communications requires the efficient identification of RTP streams in order to isolate VoIP communication from what have become big, real-time data streams. Video on demand, streaming of radio and television, video conferencing as well as other communications are all travelling through the same pipes (Matousek et al 2014). RTP detection and classification is necessary for network administrators to understand manage the network load; and for the lawful interception of communications in network forensics.

Traditional interception of telephony was relatively simple as all communication was on a dedicated single line. Interception of VoIP is far more complicated as the communication is broken into RTP packets that are routed over a dynamic network topology.

There are potential vulnerabilities in VoIP communications that have not necessarily been addressed by VoIP providers. Researchers propose an algorithm to identify the codecs

used in intercepted VoIP communications, and develop decoder tools to attempt to decode intercepted traffic. They then tested the tools against 15 VoIP apps.

It was found that of the 15 Android VoIP apps tested, the quality of the transmitted voice was of greater importance than security considerations of the app. As with security considerations in other areas, this is driven by the consumer. Of the 15 apps tested, more than half do not encrypt the voice communications and the original voice conversation could be recovered from intercepted communications. Apps such as Tango and Mo+ encrypt only the communications from app to app and not from app to phone. Communications from app to phone are, therefore, vulnerable to eavesdropping (Pokharel et al 2016).

Matousek et al (2014) [81] developed a fast multi-stage method for on-line detection of RTP streams and codec identification of transmitted voice or video traffic. The RTP streams characteristics that differentiate them from other communication. This involved two types of checks of RTP traffic, the *per-packet checking* and the *per-flow checking*. The *per-packet checking* selects possible RTP packets. The *per-flow checking* selects a codec classification using a specific feature. The researchers constructed a *Codec Mapper Table* based on a set of about 20 known codecs. There were four distinguishing features used in the table comprising payload type, *delta* time, payload size, and *delta* ratio. The table can be easily expanded as additional codecs become known. The simplicity and efficiency is based on integrity checking of the RTP packets which can be accomplished without additional information from VoIP signalisation which might not be available to the analyst or might be corrupted. The codecs could be identified from RTP streams. This method can be used in network forensic analysis in the absence of VoIP signalisation.

21. Forensic Investigations in the Cloud

The volume and sophistication of attacks targeting cloud services have increased significantly over the past few years. This includes the new attack of shadow cloud usage where cloud infrastructure is part of the attack. Rahman et al (2016) present an integrated cloud incident handling and forensic-by-design model and test it popular cloud storage providers Dropbox, Google Drive and OneDrive [82]. Their research is based on the premise that traditional incident management and investigation is not suitable due to the significantly different nature of cloud services compared with traditional terrestrial data storage and services. In addition, cloud service users and providers are likely to have different requirements in a post incident investigation. It is becoming accepted that incident handling strategies should employ forensic practices.

Rahman et al (2016) provide a snap shot of potential evidence that can be available from the various layers of cloud services (described earlier) in support of an investigation:

1. System infrastructure (cloud resources) can encompass hard disks, network logs, packet content, transaction logs, security logs, and admin access logs. Data centre artefacts can include access records, facility logs, activity logs, CCTV, biometric records, visitor records, personnel records, organisation charts and contact information.
2. Core middleware (cloud hosting platforms virtual machine management and deployment) can encompass hypervisor event logs, virtual images and drives, virtual network logs, server logs, host operating system event logs, application programming interface and portal logs, billing records, accounts information, configuration logs, audit logs, registry, antivirus/spyware logs, intrusion detection and protection systems.

3. User-level middleware can encompass source code, performance logs, debugging logs, access logs and accounts information.
4. User applications (cloud applications) can encompass application logs, authentication and authorization logs, accounts information, web content, browser artefacts, host intrusion detection system logs, chat logs, sync and file management metadata, mobile client artefacts including accessed files and database files.

Their forensic-by-design approach has six phases: 1) preparation that is integrated with forensic readiness principles; 2) identification which begins as soon as a suspicious event is detected; 3) assessment that is integrated for forensic collection and analysis; 4) action and monitoring; 5) recovery; and 6) evaluation. The model's efficacy was tested in controlled experiments using Google Drive, Dropbox and OneDrive. It was found that including digital forensic practices will provide an in-depth understanding of an incident, help to identify perpetrators and motives, and identify appropriate response actions. Date of forensic interest could be recovered from the databases of the cloud services' apps to reconstruct the events.

The forensic-by-design approach is useful for both cloud service providers and cloud service users. Cloud service providers as a general principle will maintain security of the cloud but maintain that cloud service users are responsible for security in the cloud (Rahman et al. 2016).

Other approaches to forensic examination of the cloud include examination of the device. Cahyaani et al. (2016) [83] examined the ability of forensic tools to support the forensic examination of the third most popular smartphone operating system with particular emphasis on its use as a cloud access device. The ability to examine Windows smartphones has always been limited and the most common approach is the bootloader approach although it has its limitations (Cahyani et al 2016).

22. Intelligence

Quick and Choo note that researchers rarely publish on potential intelligence to be obtained from digital forensic data holdings [84]. They note the specific identifying information such as names, associates and their contact information, addresses, motor vehicles and correspondence. But a psychological profile can also be built from the information contained in the individual's devices.

23. Tools

This paper is not going to explore and compare the capability of tools at this time, but will be available in subsequent publications. There has, however, been some research and review of tools available in the market.

Indian researchers [85] compared existing computer forensic tools on the basis of several parameters including the capability for accessing digital evidence, the sources that can be examined, metadata parsing ability, and the ability to group artefacts on the basis of the metadata.

Binary abstraction, the process of reading the binary stream of data, is provided by all forensic images and memory dumps. Although some commercial toolkits can support file system images as well as memory dumps many open source forensic tools can primarily handle only file system images in different image formats.

Metadata are very important for identifying the owner of the file, MAC timestamps, privileges etc. but most tools are not able to extract or utilise application metadata. Every forensic tool

uses text indexing and searching on the image by classifying the artefacts present in the image on the basis of the file system metadata. According to the authors, although the tools are capable of supporting multiple forensic images, they cannot associate the different metadata across files. Further, log files are processed like files so they have to be exported for analysis. It is a basic requirement to filter the contents on the basis of dissimilar keywords or classify them based on different attributes during analysis for determining a pattern. The examiner performs this function and is therefore subject to human error that may result in linkages remaining unidentified. The traditional way of combining attributes for categorization involves combining the timestamps and ownership of the forensic images, the username for log files and the IP address required for capturing network packets. This was based on the traditional understanding that hard drives are the main source of digital evidence. It is now recognised that equally important data is also present on volatile memory, log files and network packets. These must now be included in search strategies. Tools or toolkits must now be able to examine consolidated data from various sources.

Alzaabi et al. [86] refer to the current (as of 2013) first generation forensic tools that have limitations. These include a presentation of a tree structure of data that, because of the increasing complexity of the data over time since these tools were conceived, the tree structure has become increasingly complex and therefore overwhelming the investigator. In addition, cross analysis of different types of data source becomes more complicated. The researchers propose a framework for the development of next generation forensic tools. The framework uses semantic web technologies to provide a semantic-rich (the relationship between signifiers such as words, phrases, signs and symbols) environment to facilitate analysis. They tested the framework by modelling some concepts on data retrieved from Android smart phones. They found that the ontology chosen is an interpretation of the developer(s) that may lead to incorrect interpretation of the traces.

A strength of current forensic tools is the simple assessment of evidence – it is there, it is not there, it is there and unseen or unrecognised. There is little interpretation required. Although the researchers' efforts are laudable, this is a key issue that will need to be resolved as the integrity of forensic evidence must be beyond reproach and free from accusations of bias through assumptions of the analyst. With the results of analysis subject to assumptions made by the developer(s) including influence of prior knowledge, the question of bias then arises and is identified as a priority issue in the report 'Strengthening Forensic Science in the United States: A Path Forward'. Next generation tools will need to address this issue if it is to maintain one of the strengths of digital evidence.

24. Legal Frameworks and Considerations

24.1 Court Presentation

The last step in the digital evidence process is to present the results in court. Although experts generally endeavour to use laypersons' terms, the evidence is very technical and digital evidence experts experience similar challenges to those of experts in other fields. The natural language of the expert in his or her own working day is significantly different to that of the layperson and some of the language does not have an automatic simple language translation.

Cahyani et al (2016) [88] examined the impact of multimedia presentations in improving the understanding of technical terminologies and concepts in the presentation of digital evidence in legal proceedings. Members of the judiciary, investigators and linguists in the People's Republic of China, Taiwan and Indonesia were surveyed. Video clips of three technical terms were made comprising cloud computing, botnet and forensic file recovery. Participants were surveyed before and after viewing the videos. The participants' demographic data was also

recorded to determine if there was any demographic differentiation in learning outcomes. The study found that approximately 80% of participants improved their ability to comprehend the technical terms following watching the videos. There is substantial further research to be undertaken to examine the effectiveness of training for officers of the court. These preliminary findings are very promising and becoming increasingly important as scientific and technical evidence use in judicial proceedings becomes increasingly complex.

24.2 Stingrays

Stingrays is a colloquial term for a cell-site simulator. It can be used to determine a mobile phone's location by spoofing a cell tower and in some cases can intercept calls and text messages. The stingray functions by intercepting data from the target phone along with other phones in local area. On occasions, police have claimed the use of a confidential informant when a stingray has actually been used.

The Maryland Court of Special Appeals published an opinion to advise that police must obtain a warrant before deploying a stingray but must also explain what the device is and how it is used. This follows several states, the Department of Homeland Security and the Department of Justice now requiring a warrant before a stingray can be deployed (Farivar, C. 2016) [89]. Additional cases testing the same issue are being heard elsewhere in the United States.

In a further challenge to the use of stingrays, a formal legal complaint has been lodged on the behalf of three advocacy groups that contend the use of stingrays violate Federal Communications Commission rules. The argument is that the police departments do not hold the spectrum licenses to be able to broadcast at the relevant frequencies, frequencies that are exclusively licenced to mobile phone carriers. Further, by deploying stingrays, law enforcement agencies are interfering with the mobile phone network (Farivar, C. 2016) [90].

24.3 Wiretapping

Two recent cases of online wiretapping before Federal courts in the US rules in favour of the person whose data was intercepted. In the first case, a man whose email and instant messaging to a woman were intercepted by the woman's husband and used in divorce proceedings. The marketer of the interception software must face a lawsuit as the intercepted data is stored on its servers. This reversed an earlier decision in favour of the marketer that the spying was done by the customer.

In the second case, a person not using Google's Gmail emailed a person using Gmail was found not to have consented to Gmail's automatic scanning of the email for marketing purposes. Google, therefore could be sued for wiretapping violations (Kravets 2016b) [91].

24.4 Security, Privacy, Human Rights and Civil Rights

An ongoing question that still seems to be far from resolution is the contested space between the tech giants and investigative agencies. The most contested battle ground is in the United States. In short, the tech giants have taken the position of protecting their customers' privacy as a core principle of their business models. This means default encryption on smart phones is becoming standard as is the storage of personal email accounts and other personal data in other jurisdictions. In addition, community leaders such as the Reverend Jesse Jackson are supporting Apple on the basis of civil rights (Kravets, D. 2016) [92].

With the impasse, as of August 2016, the FBI has 650 phones that it is unable to examine due to encryption. Currently the FBI is gathering information with a view to go to Congress in order to seek a legislative solution. The counter argument is that, within the US, 3.1

million cell phones were stolen in 2013. This represents vast amounts of personal information and intellectual property. Without the strong encryption of the phones, that information is available to be misused through financial fraud and theft (banking information), identity theft, business fraud and illegal trading (competition for contracts). Many companies have sustained substantial losses and even bankruptcy through loss of intellectual property through hacking or stolen personal identification information. It should be remembered that law enforcement sought more secure devices due to the large number of stolen phones and the data they held (Mullen, J. 2016) [93].

The long running battle in Microsoft v. United States reached a critical point in July 2016 when the Second Circuit Court of Appeals ruled that the Stored Communications Act does not authorise courts of issue and enforce against U.S.-based service providers warrants for the seizure of customer email content that is stored on foreign servers. This concerned a case from 2013 when a New York judge issued a warrant compelling Microsoft to produce emails of an account hosted on a server in Ireland. Microsoft refused arguing that it should seek access to the emails through Irish authorities using the Mutual Legal Assistance Treaty process (Poplin, C. 2016) [94].

25. Future Trends

25.1 Cyber security mitigation

Perfect security against cyber crime for business is impossible to achieve, but it can be mitigated. The technical and social, including social engineering, challenges are evolving at a rapid rate. The critical thing for businesses is engagement and ownership of the challenge by the board and directors where it is most usually regarded as a technical issue [95]. True engagement by the board will include protocols that include the capture of logs and other evidence of an intrusion or compromise that can be used in a subsequent forensic investigation.

The UK Government designated cyber security and meeting the cyber crime challenge a high priority. This includes an investment of 1.9 billion pounds over five years including enhanced law enforcement capability [96]. The strategies for law enforcement include, but are not limited to, making the UK a hostile country for criminals to host and perpetrate cyber crime; and, identifying, prosecuting and disrupting cyber criminals world wide.

The European Union is assisting in this process by introducing the EU General Data Protection Regulations in 2018. Substantial, far reaching rules (beyond the geographical borders of the EU) concerning what data is kept, where and how, plus data privacy will be governed by enforceable regulation backed by substantial fines [97]. The regulations include notification of data breaches [98].

25.2 Internet of Things

As described in the body of this report, the Internet of Things is going to continue to grow. The volume of data produced and stored over the next two years will exceed all data produced to date. The data will come from many different platforms and the format of the data will continue to diversify. It will continue to present challenges to digital evidence practitioners in both gaining access to the data and understanding its structure and context so that it can be effectively analysed.

25.3 Driverless Motor Vehicles

Several motor vehicle manufacturers have announced the imminent arrival of driverless motor vehicles. In August 2016, Uber announced they will allow customers to summon self driving cars. The motor vehicle manufacturers themselves are investing in ride hailing

companies or looking to supply existing companies with self driving cars. Although accident rates are expected to drop markedly, any incident involving a driverless vehicle will most likely have a digital evidence component to its investigation. This is a development of the existing and growing importance of digital evidence obtained from the computerisation of current motor vehicles.

26. Conclusion

The past three years has seen extraordinary developments in the field of digital evidence. The increased use of native encryption in smart phones has made the analysis of digital evidence even more challenging while at the same time, the smart phone is becoming the most important form of evidence in an investigation.

Cloud computing and the Internet of Things, together with the rapidly growing volumes of data they produce, present new challenges for the digital evidence practitioner. These new challenges will require new and updated skills which has been and will continue to be a challenge for agencies to support.

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28. Glossary

Bit = a binary decision (0 or 1) [99]

Nibble = 4 bits

Byte = 8 bits

Kilobite (kB) = 1000 bytes

Kibibyte (KiB) = 1024 bytes

Megabyte (MB) = 1000² bytes

Mebibyte (MiB) = 1024² bytes

Gigabyte (GB) = 1000³ bytes

Gibibyte (GiB) = 1024³ bytes

Terabyte (TB) = 1000⁴ bytes

Tebibyte (TiB) = 1024⁴ bytes

Petabyte (PB) = 1000⁵ bytes

Pebibyte (PiB) = 1024⁵ bytes

Exabyte (EB) = 1000⁶ bytes

Exbibyte (EiB) = 1024⁶ bytes

Zettabyte (ZB) = 1000^7 bytes [100]
Zebibyte (ZiB) = 1024^7 bytes
Yottabyte (YB) = 1000^8 bytes
Yobibyte (YiB) = 1024^8 bytes

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Identification Sciences

Fingermarks and other impressions, 2013-2016

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<http://www.unil.ch/esc>

1 Introduction

The purpose of this paper is to provide an overview of the papers dealing with fingerprints and other body impressions (exception made of bitemarks) that have been published between July 2013 and July 2016. We tried to offer an extensive coverage of the published sources (mainly in English), but remain conscious that exhaustiveness is not possible. The reader will realise that the area is very active and counts with more than 530 publications reviewed for this report. We cover here both matters in relation to the detection of marks (mainly fingermarks) and matters associated with the forensic identification process. Given the extremely high number of articles (>280) dealing with fingermark characterization (Section 2.2) and detection (Section 2.3), we also made the following choice: all articles were cited in the introductory paragraphs (the *overviews*); however, only a selection of articles was extensively detailed in each section. The selection criteria were mostly driven by the forensic interest, the originality of the published results, or the direct outcomes (application capabilities). We relied also on the review paper by Lennard (1).

Before starting delving into the review, it is nice to remember that Jan Evangelista Purkyně was the first to introduce a classification system for fingerprints in 1823. A short biography has been recently published (2). We also would like to refer to the historical trial of Dennis Gunn in New Zealand in 1920 and the admissibility debate surrounding fingerprint evidence (3). In 2015, the IAI (International Association for Identification) celebrated its 100 years with the publication a special volume (issue 4) of *Journal of Forensic Identification* with significant historical papers. The forms taken by friction ridges on volar surfaces still fascinate and similar shapes will be found in natural species or geological formations (4). But beware of formations that cannot be distinguished. Readers will find beautiful examples of quasi undistinguishable snowflakes in the book by Libbrecht and Wing (5)³.

The field of pattern evidence in general (that includes fingerprints but also other impressions) is still under close public scrutiny and hits the headline on a regular basis,

³ See : <http://www.nytimes.com/2016/01/23/science/who-ever-said-no-two-snowflakes-were-alike.html?>

especially in the USA. The public attention was especially turned to bite marks with a series of articles initiated by the investigation of Radley Balko of the Washington Post⁴. We note also the publication of the book by Sharia Mayfield and her father (6) describing vividly the ordeal suffered by the family following the wrong identification. Two additional cases of wrong identifications involving fingerprint that have shaken public confidence: the case of Lana Canen (7) and the case of Beniah Dandridge⁵ released in 2015 after 20 years in prison following an erroneous identification by the Alabama Bureau of Investigation. When dealing with errors though, it is important to make the difference between practitioner error (the cases reported above), instrument error, statistical error, and method error (for a general discussion refer to (8)).

Forensic science is presented as a discipline in crisis according to *Nature* (9). In March 2016, *Science* had a special report calling for “reversing the legacy of junk science in the courtroom” (10). In July 2016, *National Geographic* (11) features new developments in forensic science, putting an emphasis on methods that can bring systematic and statistical measures to replace what is perceived as dangerous subjective opinions proffered *ipse dixit* by experts. Reports are soon expected from the American Association for the Advancement of Science (AAAS) on the state of affair regarding forensic impression fields and in particular fingerprints⁶. The National Institute of Standard and Technology (NIST) has awarded in 2015 a \$20 million grant over 5 years to set up a centre of excellence made of a consortium of universities with strong statistical research teams tasked to improve the statistical rigor of pattern and digital evidence⁷. The Statistical and Applied Mathematical Sciences Institute (SAMSI) offered from 2015 a program in forensic science⁸. Finally the President's Council of Advisors on Science and Technology (PCAST) of the Office of Science and Technology Policy of the White House is soon to release a report on the state of pattern evidence, including fingerprints⁹. When you combine all these efforts (in the US mainly) with the Organization of Scientific Area Committees for Forensic Science (OSAC)¹⁰ under the auspices of the NIST and the National Institute of Justice (NIJ) and with the work of the US National Commission on Forensic Science (NCFS)¹¹ we observe a complex mesh that does not make progress and coordination easy. Our view is that, at the moment, a lot of non-coordinated efforts is put into the analysis and assessment of the state of affair with tangible outcomes to come.

As we have done in previous reports, we would like to highlight some books, manual and regulatory documents that can be used as key references:

⁴ <https://www.washingtonpost.com/people/radley-balko>

⁵ <https://www.law.umich.edu/special/exoneration/Pages/casedetail.aspx?caseid=4768>

⁶ <https://www.aaas.org/page/forensic-science-assessments-quality-and-gap-analysis>

⁷ <http://forensic.stat.iastate.edu>

⁸ <https://www.samsi.info/programs-and-activities/year-long-research-programs/2015-16-program-on-statistics-and-applied-mathematics-in-forensic-science-forensics/>

⁹ <https://www.whitehouse.gov/administration/eop/ostp/pcast/docsreports>
The report (still in draft from at the time of writing) is already discussed and commented in the media : <http://www.wsj.com/articles/presidential-advisory-council-questions-validity-of-forensics-in-criminal-trials-1472720405>

¹⁰ <http://www.nist.gov/forensics/osac.cfm>

¹¹ <https://www.justice.gov/ncfs>

- The second edition of fingerprints and other ridge skin impression has been published (12). More than 10 years after the first edition, it provides an up-to-date overview of both detection and identification issues in friction ridge skin examination.
- Daluz (13, 14) published two books (one theoretical and one practical) for an entry-level course in fingerprint detection and identification.
- Houck (15) recently edited another textbook with entries published in the 2013 Academic Press / Elsevier Encyclopedia of forensic science (2nd edition).
- Mulawka (16) published a very useful guide for post-mortem fingerprinting.
- The book authored by Craig Adam (17) entitled Forensic Evidence in Court: Evaluation and Scientific Opinion, more specifically its chapter 13 on fingerprints.
- The ENFSI fingerprint working group published in 2015 its best practice manual (18) that will help laboratories harmonize their procedures and increase consistency among European laboratories especially at a time where accreditation will soon be mandatory at EU level.
- The International Fingerprint Research Group (IFRG) published key recommendations for fingerprint research on detection methods (19). It provides “best practice” guidelines for the evaluation of new or modified fingermark detection methods, from initial concept through to final casework implementation.
- The Home Office Centre for Applied Science and Technology (CAST) published its complete manual for fingerprint detection techniques. It covers all detection methods, sequences with formulation, readiness levels and health and safety requirements (20). The manual has been written in order to help laboratories to meet the ISO/SEC 17025 requirements.
- The Home Office Forensic Science Regulator published the section of its code of conduct in relation to fingerprints (21, 22). It sets the terminology and main requirements in the context of ISO/SEC 17025 accreditation.

The 2009 report of the US National Research Council of the National Academy of Sciences (23) triggered additional research that came to completion during our reviewing period. We will review them in the subsequent chapter. We will refer to it as the NRC report. The NRC report is still discussed in the literature. Some forensic practitioners held that it is “hypocritical and unrealistic for The National Academy of Sciences to expect in friction ridge analysis a level of perfection that exists nowhere else” (24).

In our last report (25), we noted the scrutiny both by the courts and by commentators or scholars on the way fingerprint evidence was admitted and presented. During this reviewing period, we report a steady decrease of the number of challenges in court (e.g. *Daubert* or *Frye* hearings). Two cases in Illinois will illustrate the current trend. To our knowledge though these cases haven’t been decided yet. The defence teams submitted two motions to the Circuit Court of Cook County (Illinois) to exclude any testimony to “absolute source identification” and any testimony that “all fingerprints are unique”¹². The motions rely heavily on the NRC report, the NIST human factor report (26) that we highlighted during our previous reviewing period and the recent U.S. Department of the Army (Defense Forensic Science Center) information paper that announce that their experts will not offer categorical opinions regarding fingerprint evidence anymore (27). We expect more challenges as to how forensic identification evidence ought to be presented in court. The report by Jackson *et al.* (28) is helpful here to set the scene across forensic science disciplines.

¹² *Illinois v. Christopher Robertson*. Motion to Exclude Statements Officer XXX Claim of Fingerprint “Identification”, Circuit Court of Cook County, 15 CR 7788, filed on December 21, 2015.

Illinois v. Anthony Guedes. Motion to Exclude Statements of the Prosecution's Fingerprint Examiner Regarding Absolute Source Attribution and Uniqueness, Circuit Court of Cook County, 15 CR 416, filed June 13, 2016.

To maintain a watching brief on the legal and reporting aspects associated with identification evidence, we recommend consulting the blog of Prof. David Kaye, Forensic Science, Statistics & the Law, <http://for-sci-law.blogspot.ch>

Finally we would like to draw the attention of all practitioners (forensic or legal) to the guide to interpreting fingerprint testimony by Edmond and colleagues (29). It gives a full account of the current debate on fingerprint matters and invites all parties to adhere to key principles of expert testimonies: transparency on the underlying basis, on the existence and numbering of error rates and on the need for humble expressions of the weight to be associated with the findings. The guide follows the one proposed for lawyers (30). The authors went further with a model forensic science advocating solutions such as disclosure, transparency, epistemic modesty and impartiality (31).

2 Fingermarks

2.1 Friction ridge skin individualization process

A chapter dealing with the philosophy of forensic identification has been published by Broeders (32). It gives the cross-disciplinary approach (from fingerprints to DNA) that is so required. In previous reports we praised Biedermann and his colleagues for their attempt to articulate the identification process. Their 2016 paper (33) is a useful addition to explain how decision are made in these identification disciplines. Decision theory (34) is the only way to understand and ultimately justify current practices. Swofford presented his personal odyssey (35) that led to major changes for his agency the Defense Forensic Science Center - DFSC (27). Indeed as per December 2015, DFSC has modified the language that is used to express "identification". Their strongest claim towards an association between a mark and a print is now reported as:

The latent print on Exhibit ## and the record finger/palm prints bearing the name XXXX have corresponding ridge detail. The likelihood of observing this amount of correspondence when two impressions are made by different sources is considered extremely low.

As noted by Cole (36), most the previous changes in reporting practice following the NRC report had been semantic and not fully articulated and explained. For Cooper (37), courts should drastically change their way to assess fingerprint evidence and critically weight the claims of, or akin to, individualization. The move taken by the DFSC is a significant shift and has overall been well received by commentators (38).

The policy and research efforts that occurred since the 2009 NRC report have been reviewed by Champod (39) and Desportes (40). We will refer to some of them in the next section.

2.1.1 Fingerprint features

During our previous review, we were able to report quite a wealth of research characterising fingerprint features (from level 1 to level 3). We note a drop of publications reporting statistical data associated with fingerprint features. More efforts have been put into models that consider features jointly or through a score based system without resorting to a piece by piece analysis.

Level 1, 2 and 3 features

Most of the recent work on level 1 features is focused on gender prediction based on the measure of ridge density (41-52). All studies report that females have a slightly higher density of ridges on their fingerprints compared to males. We were surprised by the amount

of efforts put into this area, as we don't observe a lot of operational benefits due to the limited inference allowed by ridge density.

We note some recent publications revisiting the relationship between blood groups (ABO and Rh) and fingerprint patterns (53-56). To our knowledge, the use of such data in casework has never been reported. The same applies to hand dimensions (57). Some researchers have suggested chemical analysis to distinguish between male and female fingermarks (58), but again, operational applications seem quite distant. Reasonable prediction of the hand (right or left) at the source of a mark can be based on the features (notably the distances between cores and delta, sloping of the core, clockwise and anti-clockwise rotation of ridges) of the whorls (59). Similar information is given in Brazelle (60).

Dermatoglyphics studies are rather sparse nowadays. Most of the studies are investigating potential links between pathologies or dental defects and friction ridge skin patterns and are conducted in Iran, India, Sri Lanka, Pakistan or China (61-81). We will concentrate on a few highlights only here. A higher proportion of whorls and a higher mean total ridge count are reported in hypertensive patients (64) or on patients with cardiac diseases (68, 69). The same variables may help to diagnose kidney diseases (65). A decreased number of arches but increased number of ulnar loops have been observed on patients suffering from bronchial asthma (67). Patients affected by multiple sclerosis (MS) tend to show an increased *a-b* ridge count and ridge counts in all fingers (76).

Population studies were published on Limboo, a population of Sikkim (63), a male population from the province of Jujuy in Argentina (82), in Ethiopia (74).

A new index to quantify differences between individuals has been proposed by Buchwald (83). The index is the sum of 45 absolute mutual differences between the numerical values of patterns on the digits of the left hand and the right hand of the individual. It allows measuring the morphological diversity and simultaneously asymmetry of fingerprint patterns.

During the reporting period, we did not come across a lot of papers dealing specifically with individual minutiae (population studies). One key contribution is coming from the biometric field with the PhD thesis of Krishnamoorthy (84). He showed that the use of specific minutiae of rare types in an AFIS matching strategy can significantly improve the accuracy of the matcher (85). Also researchers in biometrics have shown by using a mining technique for combination of minutiae that a 9-point feature is much rarer than a 3-point features. The 9-point feature occurred only once in 1000 fingerprints (86).

Likewise, few systematic research deal with pores or other level 3 features. In 2014, Anthonioz & Champod (87) qualified the limited but reasonable strength that pores may bring to a case balancing their reproducibility against their selectivity. New chemical techniques may help also to study pores and may offer new ways of mapping them (88, 89), but it remains in the early days. De Alcaraz-Fossoul *et al.* (90) showed variations in the reproducibility of the shape of ridges (including mismatching minutiae) that may lead to dissimilarities. More than ever, the "no single minutiae discrepancy" rule should be taken carefully.

2.1.2 Probability models, and measures of quality and distortion

Modern statistical efforts have been reviewed by Abraham *et al.* (91). Kafadar (92) exposed the general statistical issues facing forensic science at the moment. During the reviewing period, we noted the following research efforts:

- Efforts based on a score obtained from a matching system such as an AFIS (93-95). The score-based system of Alberink (94) led to published discussions (96, 97). The thesis of Wang (98) explores how matching algorithms for palmprints can be improved and used to assign a likelihood ratio to the findings. The thesis of Krishnamoorthy (84) also discusses how a score-based system can be used in forensic scenarios provided that appropriate score normalization and calibration. Haraksim in his thesis (99) and with his colleagues (100) offered ways to measure the coherence of computer-assisted likelihood ratio (LR) methods. A score-based system for fingerprint is used as an example. The work has been followed by the proposition of a guideline to be applied for the validation of LR-based methods across forensic disciplines (101).
- Efforts based on the characterisation of fingerprint features without resorting to a score from a matching algorithm, but by modelling the features directly (102). That route offers the main benefit of not having to rely on proprietary matching algorithms that need to be considered as a black box. Neumann and Saunders also indicated the limits of a score-based system from a statistical perspective (96).

One of the main observations made following the NRC report is that the assessment of the quality of marks is entirely left to the fingerprint examiner, without taking advantage of any measurement of quality. Mark quality assessment has received more attention recently (103).

Quantitative research with regards to the effect of force or distortion on fingerprint pattern is in the early stages in forensic science (104, 105). Distortion is also studied in the field of biometrics (106) and its compensation will greatly improve the accuracy of an AFIS system (107). For the fingerprint practitioner, such tools can be of critical importance when it comes to assess whether features claimed to be in correspondence by an expert are truly in line with the expected distortions that we can obtain from marks coming from the same source. The work of Kalka *et al.* (108) or Fagert and Morris (109) is taking the field in that direction. Kellman *et al.* (110) have also shown that metrics characterizing the quality of the mark will help to predict expert performance and assess fingerprint comparison difficulty.

The introduction of probabilistic models in casework is not going without its own difficulties that will need to be overcome (training, communication, culture shift). Langenburg explored some of them (111) as did Lennard (1) who suggested a wise step-by-step change that is not shared by all (112).

2.1.3 ACE-V, bias and expert performance

An ACE-V manual has been published by Brewer (113). A good discussion of the criteria for exclusion was provided by Ray and Dechant (114). Visual clues to detect tonal reversals were presented by Castellon (115). Bourque showed through a survey of examiners how diverse their responses are when it comes to articulate their conclusions (116). Bunter (117) rightly highlighted the deficiencies of some practice of ACE-V, in particular when it comes to the quality of note taking (documentation). He is advocating for a linear application of ACE with a full documentation of the Analysis phase. Following the analysis of a few cases of mis-attributions, Triplett suggested to adopt a complexity scale to describe the comparisons and a range of conclusions that are typically associated with the levels of complexity (118). Variations are not rare in casework and Mustonen and colleagues (119) showed how a forensic laboratory could strive toward clearer criteria for decision making and documentation practices. Mustonen and Hakkarainen (120) published also on apprenticeship in fingerprint identification.

During this reviewing period, studies have explored the reproducibility and variability between fingerprint examiners. Part of these studies are here labelled “white box” studies in

the sense that not only examiners were asked to make determinations at different stage of ACE-V, but they were also asked to document their findings by annotations (markup) or narratives. Substantial variations have been observed between experts' annotations and conclusions whether in analysis or in comparison (121-127). The aim of these studies is to gain a deeper understanding on the factors driving expert conclusions that follows the Analysis stage and the Comparison stage. Overall results show that minutiae count is the best predictor of value judgement. However, substantial variations of both annotations and conclusions among examiners have been observed in all studies. The interexaminer variation is large due to various reasons: absence of standardised training and of clear cognitive link between annotations and decisions. Ulery *et al.* (128) published a complete description of interexaminer minutiae markup data. When focusing only on minutiae, other studies led to the same conclusion that there is quite an important range of variation between experts (129).

Substantial changes in an examiner's markup were reported between what is been annotated during the Analysis phase and what is finally retained in the Comparison phase (126). It highlights the suggestive nature of the known print during the comparison process. It justifies the call made by Bunter (117) for a more transparent documentation of both phases.

Still a number of "black box" studies have been carried out. By "black box" it is meant the measure of the output only (conclusions) when various stimuli are presented to the examiners. Research groups have measured performance of fingerprint experts. Thompson *et al.* have shown that qualified court-practicing fingerprint experts were exceedingly accurate compared to novices (130, 131). The Miami-Dade police department carried out a large-scale test (a "black box" study) with 109 US fingerprint practitioners (132). More studies are deemed necessary to develop a strong research culture in the domain (133). Champod (134) expressed his fear to see future research dominated by bias studies without much effort put into the systematic measurement of fingerprint features. That editorial led to some reactions (135, 136).

These above studies have shown that experts are prone to errors. For example Neumann *et al.* (121), on challenging cases, obtained overall 4.92% of false negatives (wrongful exclusion) and 0.67% of false positives (wrongful association). Pacheco *et al.* (132) reported no false positive following the Verification phase of ACE-V but reported a 7.5% false negative rate. The Collaborative Testing Service (CTS) is publishing all reports¹³ associated with the annual proficiency testing. Every year cases on wrong associations and wrong exclusions are detected. The same applied to the test carried out by the ENFSI fingerprint group. In the 2015 collaborative exercise 5 false positives and 41 false negatives were detected (137).

Haber and Haber (138) challenged the quality of published research (the "black box" studies) regarding their ability to guide as to the accuracy of fingerprint comparisons. It led to quite an animated exchange of letters (139-142). What is clear however is that the profession is moving towards a culture of proficiency testing (143).

The impact of fatigue on the performance of five fingerprint examiners has been shown using eye tracking testing (144). The behavioural performance declined with fatigue, and the eye gaze statistics suggested a smaller working memory capacity with an early termination (giving-up) of the search of a mark against a set of known print.

¹³ https://www.ctsforensics.com/reports/default.aspx?F_CategoryId=21

We move now to the research dealing with cognitive bias. Dror (145) made a review of the research regarding biasability and reliability of expert observations and decisions. He then offered recently a Hierarchy of Expert Performance (HEP) that facilitates greatly the discussion on bias (146).

Osborne and Zajac (147) on a corpus of 319 students (without any forensic background) showed that crime-related context did not play a significant role in participants' judgements about non-complex (unambiguous) comparisons. On complex comparison (ambiguous mark), both the low and high emotion crime-related contexts led to an increase in 'match' decisions. Searston *et al.* (148) conducted controlled experiments with up to 48 undergraduate psychology students. They showed that their conclusions regarding fingerprint comparisons were affected by the case information provided, not always reducing accuracy.

Earwaker (149) showed that the decisions made by an analyst to keep a mark as sufficient for further comparison is influenced by irrelevant contextual information (such as the nature of the crime under investigation).

We note that when practice is analysed from an operational perspective, measuring success rates and efficiency (150), there is no clear evidence that bias (due to the knowledge of contextual element of the case at hand) has a large scale and systematic adverse effect. Kuckuka (151) quickly responded that contextual influences can unwittingly lead forensic examiners to the right decision, but for the wrong reasons and that not only the outcome should be measured but the process whereby these conclusions have been reached.

Edmond *et al.* (152) detailed how contextual information about the case could sway expert decision-making, but here considering the whole chain of the criminal justice system from the initial stages of the interrogations, plea bargains, through trial and appeal. It creates what the authors have called a 'snowball effect' due to the dangers of cross-contamination in all directions. They call for a strict blinding of forensic scientist to 'domain-irrelevant' information. Operational solutions towards reducing risks of cognitive bias have been now proposed, mainly taking advantage of sequential unmasking or blinding (145, 153, 154).

Forensic scientists are invited to pay attention to these issues as recommended by the National Commission on Forensic Science (155) and laboratories are invited to ensure that forensic analysis is based only upon task-relevant information.

2.1.4 Automated fingerprint identification systems (AFIS)

The area of automatic fingerprint recognition is vast and it is not the ambition of this report to review all the activities associated with biometric systems. We can direct readers to the excellent paper by Jain and colleagues on the 50 years of biometric development (156) and on bridging the gap between biometrics and forensic science (157, 158). Here, we propose a very narrow selection of papers that have the potential for a direct impact on forensic practices. We have deliberately avoided the rich literature focused on the technological advances such as the matching algorithms.

Neumann *et al.* (159) showed the possibility of using an AFIS system with less human intervention for marks. AFIS workflow can indeed be streamlined in the sense that marks of high quality can be processed almost automatically (light-out mode) or through a case by case by case basis (160). An overview of the definition, opportunities and challenges regarding light-out mode applied to fingermarks is offered by Meagher *et al.* (161). Changes in the workflow may also mean taking advantage of multiple matching technologies at different steps in the process. Gantz *et al.* (162) present a post-AFIS search ranking using a dedicated algorithm and an overlay method. Hefetz *et al.* (163) showed how systematic

mark-to-mark comparisons in AFIS may help developing new investigative leads, including geographical mapping. Limiting AFIS searches to a limited database of persons of interest and taking advantage of the mark auto-encoding capability of the system will increase detection (164).

Automatic detection of marks on images as shown by Yang *et al.* (165) is also a way to reduce the manual operations required to run large number of cases through an AFIS. Dealing efficiently with overlapped marks (166) or enhancing detected marks by image processing (167) participate to the same objective.

Not only the assessment of mark and print quality has impact on fingerprint comparison as carried out by an expert, but its measure can greatly improve AFIS operations. A recent review by Yao *et al.* (168) and two PhD theses, Yao (169) and Yoon (170), bring additional information for readers interested in this topic. Work on marks is under way in this area and will likely result in improvement of the AFIS part of the discipline, as well as the assistance offered to fingerprint experts in the future (171-173). When linked with AFIS technology, the use of a prior measurement of the expected evidential value can offer the potential to improve performance in the future (174, 175).

Talking about measuring quality would not be complete without mentioning the work on prints. The long-awaited NFIQ 2.0 algorithm (176) is now available with a full documentation through the National Institute of Standards and Technology (NIST): http://www.nist.gov/itl/iad/ig/development_nfiq_2.cfm

The ability to associate fingerprints using AFIS systems despite a number of years between transactions was known from practice but without systematic research. Full longitudinal studies now document this ability (177-179). Variations may also be observed on the fingerprints of elderly people (180).

2.1.5 Fingerprint alteration and pathologies

The Federal Bureau of Investigation (181) recently confirmed that voluntary alterations, either self-inflicted or with surgical assistance, are used to defeat identification efforts. The FBI reported on the discovery of 412 fingerprint records in their AFIS system with clear indications of deliberate alterations. A few groups reported on algorithms allowing the detection of altered fingerprints (182-186). A review of anti-spoofing systems for fingerprints is due to Galbally *et al.* (187) and Marasco et Ross (188). We note also the new type of material used to prepare spoofs (189). Work on the automatic detection of forged marks has been presented by Hildebrandt and co-workers (190, 191).

Lee and coworkers (192) reported that, on average, 41% of patients showing hand dermatitis on their fingerprints failed the biometric verification process. Chemotherapy treatments can also be the cause of a lack of legible fingerprints (193).

2.2 Composition, aging and persistence of fingermarks

Chemical profiling of secretion residue: A new trend emerged recently which aims at establishing the chemical profiling of fingermarks, reflecting the donor's age, sex, drug habits, medical history, or food preference (194). If the forensic interest of such information (or its implementation in an operational routine) is debatable from an investigative point of view, some of the proposed analytical methods provide useful about secretion residue composition and aging – See below.

Composition: Several studies focused on establishing the molecular composition of secretion residue through new extraction/analysis procedures or optimization of existing

ones. In particular: fatty compounds using GC-MS (195-197), LAET-MSI (198), LDI-MS (199), MALDI-ToF-NIMS (200) or SiALDI-MSI (201); amino acids using LC-MS (202); eccrine sweat using SERS (203); various compounds (e.g., amino acids, fatty acids, and other) using LESA-MS (204) or MALDI-MSI (205, 206); wax esters and saturated fatty acids using MALDI-ToF-NIMS (207), artificial secretions using DEFFI-MSI (208). A review about fingerprint composition (and aging) has been published (209), encompassing the contribution of donors, substrates, time, and environmental elements (e.g., light, temperature, humidity).

Aging and age estimation: The evolution of secretion residue with time can be useful in different ways: better understanding of the interactions between secretion residue and the underlying substrate (210) – See details below, evolution of ridge topology/characteristics (90), impact on the detection contrast (dry powder) (211), or determination of the age of fingerprints (i.e., time of deposition). Several studies addressed this last issue by considering different compounds of interest and analytical means: lipid aging using FTIR (212) or GC-MS (213), lipid diffusion using ToF-SIMS (214), protein/lipid oxidation using fluorescence spectroscopy (215), and eccrine/sebaceous compounds aging using UV/VIS spectroscopy (216-218). Observations of the visually perceived modifications over time of friction ridge features on marks (contrast and minutiae counts) have been recently published (219, 220).

If the question of age determination is of a high interest in forensic science, the influence of some (unknown) factors (e.g., donor, substrate, environmental and storage conditions, impact of applied detection techniques) in the estimation of the age currently prevents its application in casework. A review about fingerprint aging has been published (209), encompassing the different existing methods and compounds of interest identified so far. Legal considerations of fingerprint age determination received also a deserved attention (221).

Persistence: The question of persistence of a fingerprint when it is exposed to (detrimental) environmental elements has been addressed (222), including recommendations regarding some common assumptions linking the age of a fingerprint with its easiness of detection. The influence of light on fingerprints left on brass was also briefly explored (223).

Artificial secretion: The use of artificial secretions is sometimes presented as a reproducible way to leave fingerprints presenting similar chemical composition. The efficiency of (commercially-available) artificial mixtures has been evaluated and compared to actual fingerprints in regards with the application of detection methods (224-226) – See details below.

Other topics: Development of a specific substrate to map the distribution of pores (reaction with excreted sweat), using hydrochromic polymers (88, 227) or fluorescein-containing polymers (89). Determination of the gender of an individual from the colorimetric answer with NIN (228); We have some reservations regarding the proposed methodology. Indeed it requires to dissolve/extract the fingerprint of interest and doesn't bring as much value added compared to what touch-DNA could bring.

Used acronyms: **CA** (cyanoacrylate or cyanoacrylate fuming), **DEFFI** (desorption electro-flow focusing ionization), **EDS** (energy-dispersive X-ray spectroscopy), **ESEM** (environmental scanning electron microscopy), **FTIR** (Fourier transform infrared spectroscopy), **GC** (gas chromatography), **GV** (gentian violet), **IND/Zn** (1,2-indanedione combined with zinc chloride), **LAET** (laser activated electron tunnelling), **LC** (liquid chromatography), **LDI** (laser desorption ionization), **LESA** (liquid extraction surface

analysis), **MALDI** (matrix assisted laser desorption ionisation), **MS** (mass spectrometry), **MSI** (MS combined with imaging), **NIMS** (nanostructure imaging mass spectrometry), **NIN** (ninhydrin), **ORO** (oil red O), **PD** (physical developer), **PE** (polyethylene), **PP** (polypropylene), **PVC** (polyvinyl chloride), **R6G** (rhodamine 6G), **SERS** (surface-enhanced Raman spectroscopy), **SiALDI** (Silver-assisted laser desorption/ionization), **SIMS** (secondary ion mass spectrometry), **ToF** (time of flight), **UV** (ultraviolet), **VIS** (visible)

Secretion/substrate interactions – A better understanding of the interactions between secretion residue and an underlying substrate is required to increase the research efficiency in the field of fingerprint detection. In this context, Moret *et al.* (210) conducted a study based on the optical/microscopic observation of different types of fingerprints (*i.e.*, natural, eccrine- and sebum-rich) left on five substrates (*i.e.*, glass, 2 PVC, PE, PP). Numerous observation techniques were compared among which microscopy (*i.e.*, bright field, dark field, phase contrast, cross-polarization) and ESEM (combined with EDS). Phase contrast microscopy was determined to be the best technique for smooth, non-textured material, allowing the observation of lipid droplets in the secretions, as part of the emulsion. ESEM coupled with EDS showed some advantages in terms of minute morphology and composition. Preliminary results also showed interesting and valuable information about the interactions between secretion residue and substrates, such as the apparent penetration of molecular compounds in plastic-based substrates in the days following the deposition. This study should be followed by further developments shortly.

Artificial secretions – Zadnik *et al.* (224) evaluated the possibility to use commercially available artificial secretion pads (*i.e.*, “sebum” and “sweat/eccrine”) as standards for quality control assessments. To reach this goal, the authors compared how artificial-based fingerprints behave when processed with conventional detection techniques (*i.e.*, IND/Zn, NIN, ORO, PD), in regards with actual fingerprints (natural and sebum-rich). Differences of behaviour were observed: (a) sweat/eccrine pads seem to contain more amino acids than in an actual fingerprint, leading to a greater color intensity with IND/Zn or NIN, (b) reaction with NIN led to orange-red marks instead of the awaited purple (due to the Ruhemann’s purple), (c) same remark as 1 for sebum pads and ORO, leading to an overestimation of the reagent efficiency, (d) lack of reaction with PD for artificial sebum. Consequently, the authors concluded that such pads are not currently suited as replacement for actual secretions in the context of quality control assessment. In another study, Sisco *et al.* proposed to mix artificial sweat (containing 19 compounds, including inorganic salts, amino acids, and other molecules) with artificial sebum (containing 23 compounds, including free fatty acids, triglycerides, and other molecules), in the presence of an emulsifying agent (Steareth-20) (225) – [Note: detailed formulations are provided in the article]. Their initial motivation was to propose a standardized emulsion to be used for the cross-comparison of MS and chemical imaging techniques. By comparing the chemical signature of their emulsion with the analysis of actual sebum-rich fingerprints, they showed strong similarities between the two emulsions. They also showed that their complex emulsion reacted quite convincingly with conventional detection techniques (*i.e.*, dry powder, NIN, IND/Zn, CA+R6G, GV), when compared with actual sebum-rich fingerprints. Both these studies show that the consideration of artificial secretion is still of interest in the field of fingerprint detection/analysis. However, if commercially-available products suffered from their simplicity of composition (*i.e.*, oily mixture for sebum, and amino acids mixture for eccrine secretions), a complex emulsion may succeed in mimicking some properties of actual secretions.

2.3 Fingerprint detection and imaging/recording

Preliminary remark – For easiness of reading, all the articles covered in this section were structured according to five main categories: detection techniques (**T**), nature of the substrates (**S**), context (**C**), imaging methods (**I**), and other purposes (**O**).

Research trends – When classifying published articles in their respective sub-categories, it appeared that research in fingerprint detection has undergone a drastic shift towards a technological profile (Figure 1). Indeed, *detection/identification of contaminants* (e.g., drugs, explosives) and *chemical imaging* represent the 2nd and 3rd topics, respectively, in terms of total number of articles in the context of detection. Unfortunately, such technological trend seems to occur at the expense of the conventional/field detection techniques and of the overall quality of research/publication: lack of following studies (“one-shot” publications), over-specialized equipment requiring specific abilities, overlook of forensic considerations, absence of integration in operation procedures, to cite the major issues. More surprisingly, *powder dusting (micro- and nano-sized)* represents the topic presenting the largest number of publications in the context of the detection. Current research interests are consequently doing the splits between low-tech detection techniques (dry-dusting) and high-end technology (chemical imaging/analysis). This trend should be confirmed in the next period (2016-2019), and its impact on the number of studies dealing with conventional detection techniques (closer to field operators) surveyed. In addition to that, an overview of the 2011-2013 research efforts of the International Fingerprint Research Group (IFRG) were summarized in a publication (1).

Publication trends – In addition to their classification in sub-categories, all the articles covered in sections 2.2 and 2.3 have been sorted according to the scientific journals they were published in (Figure 2). All journals were then further sub-categorized according to their main scopes (i.e., “forensic” and “non-forensic”). It can be seen that the majority of articles were published in forensic-oriented journals (61%) compared to non-forensic journals (39%). The three most popular forensic journals are *Forensic Science International* (16% of all publications), *Journal of Forensic Identification* (14%), and *Journal of Forensic Sciences* (12%), representing together 69% of the forensic-oriented journals. The trend is different with the non-forensic journals: *Analytical Chemistry* is the most popular one, but represents only 5% of the overall publications (13% of the chemistry-oriented journals). This is due to a surprising phenomenon: the “Other (≤ 2)” category which encompasses all the articles associated with journals appearing only once or twice for the covered period. With 18% of the overall publications (46% of the chemistry-oriented journals), this trend may reflect either a lack of pertinence in the choice of the journals or a consequence of the rejection rate of such manuscripts in more forensic journals. Finally, it should be noted that most of the non-forensic journals are chemistry-oriented, which reflects the technological trend associated with chemical imaging/analysis applied to fingerprints.

IFRG guidelines – In an attempt to provide guidelines for people interested in performing research in fingerprint detection, the International Fingerprint Research Group (IFRG) members have published guidelines describing the different steps that any technique should go through before being considered for operation use (e.g., proof-of-concept, optimization, validation, pseudo-operational trial) (19). The primary targets of these guidelines are researchers as well as editors of scientific journals who may seek some reviewing guidance. The published recommendations are not mandatory but could greatly help in quickly estimating if a technique is in its developing stage or close to be proposed for operational use. From the ca. 280 articles cited in sections 2.2 and 2.3, 29 articles have cited the IFRG guidelines – which represents 10% of the publications. We can see this figure as twofold: encouraging, as it is awaited that this number will increase in the forthcoming years; or mitigated, as most of the authors who cite these guidelines are already well aware of the issues associated with forensic science and fingerprint detection.

It must be specified that these guidelines emerged from the facts that some results tend to be overstated in numerous publications (especially witnessed for publications associated

with dry-dusting or nanoparticles in solution) or that experimental designs present serious lacks regarding forensic/fingerprint considerations. As a reader, the combination of the following elements should raise concerns: (a) exotic technique barely applied for fingerprint detection, (b) small-scale study including only a couple of donors, sebum-rich and fresh fingerprints, and limited number of substrates, (c) minimalistic or insufficient performance assessment of the new technique regarding conventional and well-accepted methods, and (d) overstating conclusion regarding the applicability of the method.

It is hoped that the spreading of these guidelines among researchers (being forensic scientists or not) will help in focusing the research efforts in accordance with the current forensic needs.

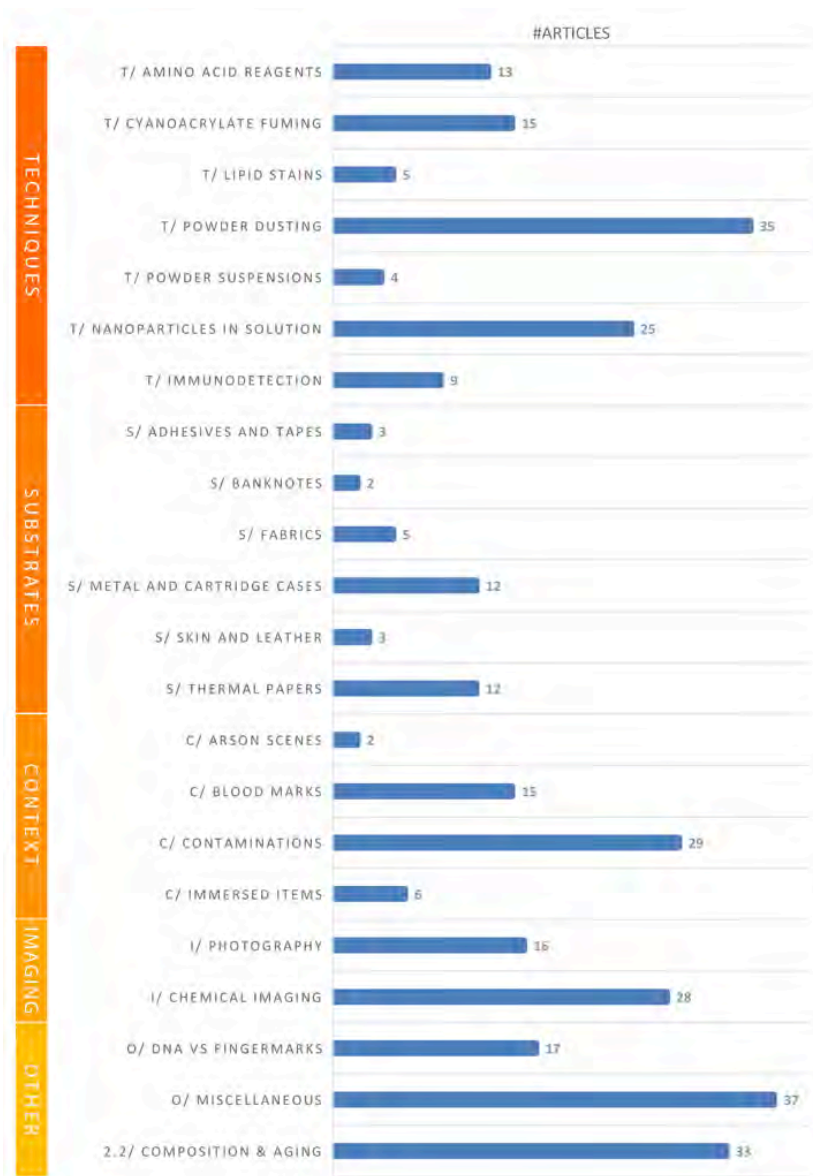


Figure 1 – Number of articles per defined category (please note that some articles can be present in two categories if they present more than one main scope)



Figure 2 – Sunburst representation depicting the number of articles per journal, for all contributions cited in sections 2.2 and 2.3. The sub-category “Other (≤ 2)” contains all the articles having been published in journals in which a maximum of two articles dealing with fingerprints were published in the covered period.

2.3.1 T/ Amino acid reagents

Fundamental studies: ^{13}C -MAS-NMR was successfully applied to study the reaction products between fingerprints and amino acid reagents (i.e., IND/Zn, DFO, NIN) as well as the molecular interactions with the cellulose matrix (229). GC-MS combined with molecular derivatization (230) as well as LC-MS (231) were used to determine the amount of amino acids left on a porous substrate after reaction with IND/Zn, DFO or NIN, applied as stand-alone or in sequence; The obtained results go in favour of the application of these reagents in sequence. The impact of different parameters linked to donors (e.g., age, gender, activity prior deposition – such as food consumption or hand washing) has been studied in regards with IND/Zn performances (232) – See details below. Finally, a computation study describing the structure of genipin in solution may find its interest in any future development considering this molecule for fingerprint detection (233).

Practice-oriented studies: A thorough evaluation of two detection sequences dedicated to porous substrates was conducted, including amino acid reagents, PD and NR (234) – See details below. The addition of molecular sieve pellets to an HFE 7100-based DFO solution may help extending the shelf life and stability of the solution by preventing the formation of aqueous particles (so-called “second phase”) (235). A new pDMAC formulation (236) and a solvent-free pDMAB (237) have been proposed to detect marks on porous substrates; Both formulations were extensively studied and compared to conventional amino acid reagents, leading to the conclusion that they both lack of sensitivity and that further optimization/research are consequently required. Different formulations of IND/Zn were qualitatively and quantitatively compared to DFO in an attempt to find a replacement for this latter (238); The best results were obtained with a formulation based on HFE-7100 and containing 0.08% w/v of IND. A study aiming at determining the effect of NIN on the paper structure showed an increase of the paper thickness after the detection process (239).

Future prospects: Sublimation of NIN under vacuum has been presented as a way to detect marks on porous substrates such as thermal paper or banknotes (240); A vacuum of 50mTorr (0.067mbar), a heating temperature of 80-90°C, and exposition to environmental atmosphere for the reaction to take place were shown to be the best configuration, even if differences in performance were observed among the porous substrates. The concept of “fingerprint developing membrane” has been proposed to detect fingermarks on porous (and non-porous) substrates using encapsulated NIN molecules in a solid matrix (241) – See details below.

Used acronyms: ¹³C-MAS-NMR (solid-state carbon-13 magic angle spinning nuclear magnetic resonance), DFO (1,8-diaza-9-fluorenone), pDMAB (p-dimethylaminobenzaldehyde), pDMAC (p-dimethylaminocinnamaldehyde), GC (gas chromatography), IND (1,2-indanedione), IND/Zn (IND combined with zinc chloride), LC (liquid chromatography), MS (mass spectrometry), NIN (ninhydrin), NR (Nile red), PD (physical developer), RH (relative humidity), RT (room temperature)

Detection sequence – In a thorough study, Marriott *et al.* compared two detection sequences to be applied on porous substrates: [Seq1] IND/Zn → NIN → PD → NR and [Seq2] DFO → NIN → PD → NR (234). The aim was to determine which sequence gives the best results in terms of number of detected fingermarks and ridge detail quality, as well as to assess the impact of the local climate on the performances. For this last parameter, the experiments were conducted in Canberra (dry and continental climate, 50%RH) and in Sydney (temperate and coastal climate, 61%RH). The conclusions were the following: (a) negligible difference between the two sequences when considering controlled experiments, but [Seq1] outperformed [Seq2] during the pseudo-operational trials conducted on 5-year-old examination booklets from local universities (+21% in Canberra and +16% in Sydney), (b) marks detected by IND/Zn are of better quality compared to DFO, (c) the impact of the subsequent application of NIN is greater on DFO than on IND/Zn, (d) PD led to a limited number of additional marks, (e) further developments are required before considering NR in an operational sequence, mainly because the used formulation failed in detecting any fingerprint supposedly due to the solvents used for IND/Zn and NIN, and (f) no significant role of the environmental conditions were observed (however, only a small difference in %RH was monitored between the two cities, for the duration of the study). The following sequence is consequently recommended for the processing of porous substrates (detection protocols between brackets): IND/Zn (160C, 15s) → NIN (RT, 24-48H) → PD.

1,2-Indanedione – In their study Fritz *et al.* (232) assessed the influence of different parameters on the composition in amino acids of fingermarks, and eventually on the performance of IND/Zn. This study included a large set of fingermarks (*i.e.*, 120 donors,

natural marks, left on conventional paper, processed after 24-36H with IND/Zn 160C-10s) which were observed readily after processing and three years after (to assess the effect of time on IND/Zn-processed items). Parameters for which an effect was observed are: the age of the donor (donors under 25-year-old leading to better quality marks), the washing of the hands prior deposition (quite logically), as well as the time after processing (significant degradation of ridge details when the items were observed again three years after the initial observation). No apparent effect of the gender or of food handling/consumption was observed. About this last parameter, it somewhat appeared that the marks left by donors having handled/consumed food before the deposition showed a higher rate of degradation when observed again three years after the application of IND/Zn (unexplained phenomenon).

Membranes – In an attempt to propose a new way to detect fingermarks on porous (and non-porous) substrates, Yang and Lian (241) introduced the concept of “fingerprint developing membranes”. These membranes were synthesized by encapsulating NIN molecules into a water-soluble or lipo-soluble solid matrix which is then applied on the item to be processed. This publication remains a proof-of-concept as the efficiency of the membranes was assessed by using extremely fresh and rich marks left on paper and leather.

2.3.2 T/ Cyanoacrylate fuming

One-step luminescent CA: Several studies aimed at comparing the efficiency of various commercially-available one-step luminescent CA processes: Lumicyano (242-244), PolyCyano UV (244, 245), CN Yellow Crystals (244) and PECA Multiband (244) – See details below. A synthetic study was carried out to try better understanding the mechanisms behind the one-step luminescent CA (246); Results go in favour of a co-vaporisation of CA and fluorescent dyes instead of the covalent binding of fluorophores on CA mono-/oligomers (derivatives).

Practice-oriented studies: Atmospheric and vacuum fuming processes were compared using plastic carrier bags and one-step/two-step CA (247) – See details below. A comparative study aimed at determining the best fluorescent dyes which could be applied subsequently to CA (248); Considering commonly-encountered non-porous items, BY40, MRM-10 and MBD presented the better performances, but no dye could successfully perform on all the considered substrates. The choice for aluminium container in fuming cabinets was briefly studied by considering alternatives (i.e., glass, steel, and ceramic containers) (249); Contrary to the hypothesis saying that aluminium would act as a polymerization retardant, the authors rather retained the fact that aluminium is overall a good thermal conductor. “Rejuvenation” of fingermarks prior to CA has been induced by exposing them to UV, X-ray, or thermal neutrons (250); Exposure to any of these three ionizing radiations could enhance the detection performance by 20-30% (in terms of minutiae count), supposedly by acting on the cross-linked lipid molecules. Finally, CA has been identified as part of an optimized sequence aiming at detecting marks on Canadian polymer banknotes (251) – See section 2.3.9 for details.

Future prospects: Detection of fingermarks on fabrics has been proposed by combining CA with FTIR chemical imaging (252). A NIR two-photon induced fluorescence imaging technique has been proposed to image CA-processed fingermarks on highly-reflective substrates (253).

Used acronyms: **BPS** (black powder suspension), **BY40** (basic yellow 40), **CA** (cyanoacrylate or cyanoacrylate fuming), **FTIR** (Fourier transform infrared spectroscopy), **IND/Zn** (1,2-indanedione combined with zinc chloride), **LCA_{x%}** (Lumicyano solution

containing x% of Lumicyano powder), **MBD** (7-p-methoxybenzylamino-4-nitrobenzene-2-oxa-1,3-diazole), **MRM-10** (mix of MBD, R6G and BY40), **NIN** (ninhydrin), **NIR** (near infrared), **PE** (polyethylene), **R6G** (rhodamine 6G), **RH** (relative humidity), **SB3** (solvent black 3), **UV** (ultraviolet), **WPS** (white powder suspension)

One-step luminescent CA – One-step luminescent CA is definitely the biggest advance in fingerprint detection over these last three years. Different manufacturers/providers have almost simultaneously presented their products, among which: Lumicyano (CST – Crime Science/Scene Technology, F) (242), PolyCyano UV (Foster + Freeman, UK), CN Yellow Crystals (Aneval Inc., US) and PECA Multiband (BVDA, NL). At the exception of Lumicyano (which is liquid and should be heated at 120°C), all the other one-step products are sold as solid polymers which should be heated up to 230°C to vaporize. In this context, co-vaporization of a luminescent dye with CA monomers/oligomers seems to be the most likely technical solution chosen by the different providers (246).

Quite logically, several studies aimed at assessing the absolute and relative efficiency of these products (243-245, 247, 254). The conclusions of these studies are the following (chronologically sorted):

- Farrugia *et al.* (243) compared “LCA_{1%} → BY40” with “CA → BY40” and BPS/WPS on carrier plastic bags – [Please note that at the time of this study, CST sold Lumicyano as a premix solution, which contained 1% of dye; explaining the choice for the following notation: LCA_{1%}]. All three techniques performed similarly when LCA_{1%} was applied alone (without dye staining), with an equivalent number of marks detected. However, when LCA_{1%} was followed by dye-staining, +15% additional marks were detected; making of “LCA_{1%} → BY40” the best detection sequence for this study;
- Chadwick *et al.* (245) conducted a study about the PolyCyano UV, including an optimization of the fuming procedure and a comparison between “PolyCyano UV → R6G” and “CA → R6G” on aluminium, glass and PE bags. Optimized parameters for PolyCyano UV were determined (*i.e.*, 0.5g for an MVC1000 cabinet, 75%RH, 230C and 25min fuming time). When PolyCyano UV is used alone, the luminescence of the detected marks is weaker than the conventional sequence. Dye-staining of PolyCyano-processed marks significantly improved the performance. However, the authors concluded that PolyCyano UV did not represent an advantageous replacement of the conventional sequence for common non-porous substrates, mainly for cost issues [At the time of this study, PolyCyano UV cost 150AUD for 10g compared to 6-7AUD for 20g of conventional CA];
- In a multi-step study, Farrugia *et al.* (254) assessed the performance of the new Lumicyano packaging (composed of two separate bottles: “LCA_{solution}” containing the monomers to be fumed and “LCA_{powder}” containing the luminescent dye which has to be weighted and mixed with LCA_{solution} before fuming). In a first step, they compared “LCA_{4%} → BY40” with “CA → BY40” on carrier plastic bags. Similarly to their first study, they observed that an equivalent number of marks were detected with LCA_{4%} alone compared to the conventional “CA → BY40” sequence. When LCA_{4%} was followed by dye-staining, +20-30% additional marks were detected; making of “LCA_{4%} → BY40” the best detection sequence for this study. In a second step, they considered the use of “LCA_{solution} → BY40” compared to “CA → BY40”, which resulted in +16% of additional marks for the sequence using LCA_{solution}. Finally, they assessed the performance of LCA_{4%} in regards with the processing of several semi-porous substrates (*e.g.*, junk mail, magazines, cardboard packaging), using conventional reagents (*i.e.*, IND/Zn, NIN, BPS, black magnetic powder, and SB3). On glossy magazines and junk mail, amino acid reagents performed better than LCA_{4%}; on food/cosmetic cardboard packaging, LCA_{4%}, NIN, BPS and magnetic powder performed similarly; on fast-food packaging, BPS provided the highest detection rate (+19% and +28% compared to “LCA_{4%} →

BY40” and SB3, respectively). Overall, semi-porous substrates led to a low number of detected marks. The authors also indicated that LCA-processed marks seem to be more easily visualized using a blue-green excitation source with 529nm observation filter, rather than a UV excitation source;

- In their latest study, Farrugia *et al.* (247) concluded that “LCA_{4%} → LCA_{4%} → BY40” was the best sequence so-far to detect marks on plastic carrier bags – See below for details;
- Khuu *et al.* (244) compared four one-step CA products available on the mark: LCA_{4%}, CN Yellow Crystals, PolyCyano UV and PECA Multiband. The “one-step CA → R6G” sequences were compared to the conventional “CA → R6G” (using Cyanobloom from Foster+Freeman) when applied on PE (non-porous), glossy cardboard and polystyrene (semi-porous) substrates. Under white light, the quality of conventional CA decreases as the age of the marks increases (especially true for semi-porous substrates), contrarily to LCA_{4%} which showed increased performance on aged marks for all substrates. Under luminescence, fingermarks with higher intensity were observed with R6G rather than with one-step CA (confirming the observations made in previous studies). Also, no one-step CA outmatches the others, as they all perform varyingly according to the substrates and the ages of the marks. The authors concluded that the conventional sequence (CA → R6G) remains competitive compared to one-step CA, except for polystyrene and older marks. Finally, the authors did not observe a substantial increase of detected marks when considering the “one-step CA → R6G” sequence, contrarily to the observations made by Farrugia *et al.*

To summarize: all the studies agree that one-step luminescent CA present some serious advantages, the biggest being the possibility to obtain luminescent marks on semi-porous substrates, for which dye-staining is prohibited. However, some limitations were also identified (*e.g.*, cost issues and weaker luminescence compared to the conventional sequences). From these studies, it can also be concluded that a subsequent dye-staining step is still required to obtain the best results. From almost all the published studies, “One-step CA → Dye-staining” appears to be the best-so-far sequence to detect marks on non-porous substrates based on cyanoacrylate technology.

Atmospheric vs. vacuum fuming process – In their study, Farrugia *et al.* (247) aimed at assessing the difference in performance between the atmospheric (conventional) and vacuum (5 torr) fuming protocols. Plastic carrier bags from different providers were collected and readily used in successive pseudo-operational trials. The number of detected marks was recorded for each step of the studies. Different sequences were compared, which are not described here for clarity reasons. At the completion of their study, the authors observed that:

- Marks detected using the vacuum protocol (CA^{vac}) are not readily visible through naked eye and should be dye-stained to be observed (CA^{vac} → BY40);
- +50% of marks were obtained when using the atmospheric cabinet (CA^{atm} → BY40) compared to vacuum (CA^{vac} → BY40), mostly due to a stronger background staining with the vacuum process after the application of BY40;
- It is possible to detect marks using the one-step CA under vacuum (LCA_{4%}^{vac}), which has not been reported in the forensic literature yet. However, the LCA luminescence decays much faster for the marks detected under vacuum. Moreover, the subsequent application of LCA_{4%} under atmospheric conditions (LCA_{4%}^{vac} → LCA_{4%}^{atm}) led to a substantial increase of detected marks (+372%). This indicates that vacuum conditions are not optimal for the one-step process;
- The best sequence consisted in performing two successive cycles of LCA_{4%} in an atmospheric chamber followed by dye-staining (LCA_{4%}^{atm} → LCA_{4%}^{atm} → BY40). Quite surprisingly, performing two successive cycles of LCA_{4%}^{atm} instead of one increased the

number of detected marks by +32%, with an additional +12.5% obtained with the ultimate application of BY40. The authors tried to explain this through a morphological study of the polymer.

2.3.3 T/ Lipid stains

Fundamental studies: A study describing the solubilization properties of organic solvents in regards with fingerprint material, as well as their ability to partition dyes into secretions, may provide valuable information for any further development in the use of lipid stains (255).

Practice-oriented studies: The performance of ORO was assessed comparatively with PD and NIN on dry and wet porous substrates (256) – See details below. The performance of curcumin (NY3) to detect marks on naturally-weathered metal and plastic items has been extensively assessed (257) – See details below.

Future prospects: Two different ways of using NR to detect fingerprints were proposed: aqueous solution of NB (258) and oil-in-water microemulsion of NR (259) – See details below. A lipid-selective bodipy dye (LD540) solubilized in a solvent mixture optimized for fingerprint secretions has been compared to NR (255); This interesting study is however counterbalanced by the use of perfluorocarbon-based solvents which have a negative environmental impact.

Used acronyms: **NB** (Nile blue A), **NIN** (ninhydrin), **NR** (Nile red), **NY3** (natural yellow 3), **ORO** (oil red O), **PD** (physical developer), **SB3** (solvent black 3)

Oil Red O – In Honig and Yoak's study (256), NIN led to the best performance on dry substrates (67% of test marks detected), followed by ORO (42%) and PD (25%). The authors also confirmed the observation stating that the performance of ORO decreases with older fingerprints, contrary to PD (whose performance increases with time). On wet substrates, ORO (ca. 90% of test marks detected) outperformed PD (ca. 40-50%). The study showed that the buffer rinsing bath recommended in the original formulation could be replaced by water rinsing. Finally, it should be noted that eccrine and sebaceous pads as well as so-called "natural" marks (rather eccrine and/or sebum-rich) were used in this study, but that sebaceous pads could not be used as positive controls as ORO reacted poorly with such mixture.

Natural Yellow 3 – In their extensive study, Perry and Sears (257) optimized the formulation and application protocol of NY3 and showed that this dye can be effective in detecting marks on metal or plastic items which have been exposed to detrimental weather conditions. They also concluded that NY3 can be used in sequence with SB3 (SB3 → NY3). Further work is still needed, especially regarding the brand of NY3, the storage conditions of the working solution, and the application in sequence with other reagents.

Nile Red – NR has been previously reported as a new lipid stain able to detect fingerprints (25), but required further developments. Two different application protocols have been recently proposed in the literature: aqueous solution of NB, leading to NR by spontaneous hydrolysis (258) and oil-in-water microemulsion of NR (259). In Frick's approach (258), the trace amount of NR generated by the spontaneous hydrolysis seems to be sufficient to stain the fingerprints, which are observed under white light (blue-stained, due to NB) and under luminescence (due to NR). Frick's protocol is simpler and cheaper, but has only been tested on fresh sebum-rich marks left on a limited number of substrates. Nevertheless, promising results were obtained by the authors. In de la Hunty's approach (259), the choice has been made to encapsulate NR in an oil-in-water microemulsion. Three formulations have been compared in this study (*i.e.*, the original/methanol-based, Frick's aqueous NB, and the microemulsion) on sebum-rich and natural (fresh) marks left on paper. Results showed that

NR is more efficient in detecting sebum-rich marks (compared to natural). No consensus has however been reached regarding the formulations (balance between cost, ridge quality, and shelf-life) but the microemulsion formulation seems to outperform the aqueous NB while offering advantages compared to the original methanol-based formulation.

2.3.4 T/ Powder dusting (micro- and nano-sized)

Preliminary remark: Quite surprisingly, dry-dusting of powders (micro- and nano-sized) is the category presenting the highest number of publications along the period covered by this review (35 articles in total). Despite the relative efficiency of already existing commercially powders, studies of varying quality are still conducted – most of the time for economic reasons. We note the important number of publications dealing with the dry-dusting of nanoparticles (ca. 60% of the publications referring to the use of dry powders). Such research philosophy should raise concerns from the scientific/forensic community for this could lead to serious health and safety issues for practitioners, some powders containing heavy metals such as cadmium. One article specifically addresses the issues related with the dry-dusting of nanoparticles (260), but is unfortunately not considered by those most concerned. For this reason, publications referring to the dry-dusting of nanoparticles are only cited (261-281), but not further described in this report.

Fundamental studies: Gürbüz et al. (282) have studied the relation between the particle sizes and the background staining induced by the dusting of porous substrates with magnetic powder – See details below.

Practice-oriented studies: Several kinds of powders were proposed to detect fingermarks on non-porous substrates, with more or less success: cationic pigment-intercalated montmorillonite (283), chilly (284), coal (284), imperata cylindrica (285), pepper (284), Robin® powder blue (commercial whitening agent) (286), and turmeric/curcuma (284). A contactless application protocol based on aerosolized powder (i.e., Powder Puff, from Lynn Peavey Company, US) has been assessed in a small-scale study (287). Weston-Ford et al. (288) conducted a study aiming at optimizing the detection of fingermarks on elephant ivory; The best results were obtained with the “SupraNano” range of powders (ARRO SupraNano Ltd, UK) – [Note: despite the presence of the “Nano” suffix, the powder distribution size is claimed to be in the micron-range]. A study aiming at assessing the risks of drug cross-contamination through the dry-dusting process has been carried out (289) – See details below.

Future prospects: The use of powders optically active in the NIR range has been reported through the use of spirulina platensis (290), cuprorivaite/Egyptian blue (291, 292), and dye-doped porous silicon microparticles (207) – See details below. A proof-of-concept study presented the use of a diacetylene-based magnetic powder to detect marks on non-porous substrates (293); Briefly: UV irradiation of the dusted marks induces the photopolymerization, leading to blue marks, which can further be heated to result in red and luminescent marks.

Used acronyms: **MALDI** (matrix assisted laser desorption ionisation), **MSI** (mass spectrometry combined with imaging), **NIR** (near infrared), **SALDI** (surface-assisted laser desorption ionization), **UV** (ultraviolet).

Particle size and substrate porosity – Using different Fe₃O₄-based magnetic powders, Gürbüz *et al.* investigated the relation between the particle size (from <20µm to 150µm) and the background staining induced by the dusting of substrates presenting different porosities (282). Natural marks of different ages were considered for this study, as well as various substrates chosen for their belonging to general classes (*e.g.*, raw wood, paper, glass slide)

– [Note: the porosity has been characterized from close-up and microscopic observations]. Results indicated that (a) background staining is directly related with the amount of fine particles in the powder, which can be explained by the entrapment of fine particles in the substrate pores; (b) background staining starts to become detrimental when a critical amount of fine particles in the mixture is reached; (c) a powder containing only coarse particles will result in the lowest background staining, but also the lowest detection contrast even with fresh marks; (d) for a same powder, the detection performance varies with the porosity of the substrates and the age of the marks. The three powder mixtures performing the best on most substrates are characterized by an average particle size of 57-67 μ m.

NIR luminescence – The NIR region covers wavelengths ranging from 700 to 1000nm. Observing marks in this area of the spectrum offers many advantages among which the fact that most conventional dyes lose their optical properties, which can be helpful for patterned or challenging substrates such as banknotes. It is possible to distinguish “NIR” reagents (excited in the visible range and observed in NIR) and “NIR-NIR” ones (excited and observed in the NIR range). In both cases, specific material is required: adapted excitation source and IR long-pass observation filters. In the literature, two NIR powders based on *spirulina platensis* (290) and cuprorivaite (Egyptian blue pigment) (292) are reported, as well as a NIR-NIR powder based on cuprorivaite (291). Another NIR powder, based on dye-doped porous silicon microparticles, is also reported but results in poor ridge details (207); This powder has rather for aim to be used for chemical imaging – See section 2.3.19.

Drug cross-contamination – In an attempt to assess the risks of drug cross-contamination during the dusting process, Sundar and Rowell (289) conducted a study using magnetic powders (applied with a magnetic wand) and conventional powders (applied with a squirrel hair brush and a Zephyr). Spiked marks were generated according to two scenarios: (i) aliquots of drug of different concentrations were applied on the fingertips, and left to dry before fingermarks were deposited, (ii) the donor was first asked to touch a crushed drug-containing tablet before leaving fingermarks. Adjacent to the spike marks, non-contaminated marks were left. Different dusting practices were then compared, always beginning by the dusting of the spiked mark. Dusted fingermarks were then imaged by chemical imaging techniques (*i.e.*, MALDI-MSI and SALDI-MSI) to check the presence of drug molecules. The observed cross-contamination cases were mostly caused by the used material (*i.e.*, contaminated hair brushes and/or powder pot), leading to the conclusion that best-practices should be adopted to prevent such cases.

[Note: in the context of cross-contamination caused by dusting, it should be noted that DNA cross-contamination is a more serious problem; drug cross-contamination being rather linked to chemical imaging purposes or if dusted marks are actually analyzed for the presence of drugs – which is quite uncommon in practice. Nevertheless, best-practice recommendations including the regular decontamination of the dusting material are applicable in both cases].

2.3.5 T/ Powder suspensions (micro-sized)

Fundamental studies: The presence of pigments (*i.e.*, TiO₂) within the top 30nm of a polymer-based substrate can influence the unwanted deposition of C-BPS, supposedly due to surface energy variation (294); On the contrary, MoS₂-based SPR and CA seem to be unaffected by the presence of pigments – certainly due to different detection mechanisms.

Practice-oriented studies: SPR-W (BVDA, NL) has been assessed as the best technique to detect blood marks on a dark substrate (*i.e.*, black polypropene sheet) (295) – See section 2.3.15 for details. Fe-BPS has also been identified among the best techniques for the processing of (artificial) leather items (296) – See section 2.3.12 for details. The addition of

crystal violet and basic fuchsin dyes to a ZnCO₃-based SPR led to the obtaining of a violet- and purple-colored SPR, respectively (297, 298).

Used acronyms: **BPS** (black powder suspension), **C-BPS** (carbon-based BPS), **CA** (cyanoacrylate or cyanoacrylate fuming), **Fe-BPS** (iron oxide-based BPS), **SPR** (small particle reagent), **SPR-W** (white-colored SPR)

2.3.6 T/ Nanoparticles in solution

Fundamental studies: The underlying mechanisms leading to the detection of fingerprints by PD were studied (299, 300) – See details below. Similarly, functionalized silica-based NPs were used to try understanding the interaction mechanisms between NPs in aqueous solution and secretion residue (301, 302) – See details below.

Practice-oriented studies: MMD and SMD were assessed in different studies (303, 304) – See details below. For those not accustomed with PD, a brief overview of this technique has been published (305) – [Note: the referred formulations are exactly not those currently recommended by the US Secret Service].

Future prospects: A bi-functional reagent based on IND-functionalized gold NPs was developed as a new way to detect fingerprints (306, 307); The underlying mechanism consists in first making the nanocomposite interact with secretion residue through the IND chemical group, followed by a PD-like enhancement of the gold NPs. Further studies are however required before assessing the performance of such an approach. Several nanocomposites dispersed in solution were proposed to detect fingerprints with more or less success, among which block copolymer-functionalized gold NPs (308), C-dots (309, 310), cadmium-based or ZnS QDs (311-314), conjugated polyelectrolytes (315), Cu₇S₄ nanocomposites (316), lanthanide-based upconversion NPs (317), lanthanide-doped silica NPs (272), and ZnO-SiO₂ NPs (266). Aptamer- and antibody-functionalized NPs were also proposed for the specific detection of secretion residue (318-321) – See section 2.3.7 for details.

Used acronyms: **BY40** (basic yellow 40), **C-dots** (carbon dots), **CA** (cyanoacrylate or cyanoacrylate fuming), **Fe-BPS** (iron oxide-based black powder suspension), **IND** (1,2-indanedione), **IND/Zn** (IND combined with zinc chloride), **MMD** (multi-metal deposition), **NPs** (nanoparticles), **PD** (physical developer), **PE** (polyethylene), **PVC** (polyvinylidene chloride), **QDs** (quantum dots), **SMD** (single-metal deposition), **VMD** (vacuum metal deposition), **VMD_{Ag}** (silver-based monometallic VMD)

Insight into the PD detection mechanism – de la Hunty *et al.* (299, 300) tried to identify the underlying mechanisms involved in the detection of fingerprints by PD. If PD is known for its ability to detect marks on (wetted) porous substrates through silver reduction in solution, the actual detection mechanism is still unknown. In a two-step study, de la Hunty *et al.* considered the hypotheses stating that PD targets the lipid fraction of the secretion residues (299) or the eccrine constituents (300). In their first study, they considered: (a) spot tests of fatty acids, cholesterol and squalene; (b) removal of the lipid fraction through washing with various organic solvents, and (c) close observation of silver deposition along the ridges and pore sites. In their second study, they considered: (i) depletive series of natural marks characterized by no time interval between each deposition and (ii) depletive series [...] with a 10-second-interval between each deposition. The obtained results were then compared with IND/Zn. The combination of both studies goes in favour of a third hypothesis, which is that PD rather interacts with a complex mixture of both eccrine and non-water-soluble components. Their observations can be summarized as follows: significant silver deposition caused by cholesterol; performance of PD more affected when solvents able to dissolve water-soluble components were used; silver deposition varied at pore sites;

consistency between PD and IND/Zn regarding the depletive series of natural marks; poor results of PD compared to IND/Zn when considering eccrine-rich marks.

Interaction between NPs and secretion residue – In their studies, Moret *et al.* (301, 302) explored the possibility to use functionalized (dye-doped) SiO₂ NPs to try understanding physico-chemical interactions between NPs and secretion residue. By grafting various chemical groups, monitoring the zeta potential, and varying the pH of the solution, they showed that the presence of carboxyl groups is mandatory to the successful detection of fingerprints using such NPs. Moreover, instead of a mechanism solely driven by electrostatic interactions, they showed that the detection was most likely chemically-driven (*i.e.*, formation of amide bonds with the amine groups contained in the secretion residue). This study is a first step in a better understanding of the detection mechanisms of physico-chemical techniques, such as MMD/SMD methods, which both involve aqueous suspension of carboxylic acid-functionalized gold NPs.

MMD/SMD – MMD and SMD are two sibling techniques, based on the use of gold nanoparticles in aqueous solution (*i.e.*, colloidal gold) combined with a metal deposition step (enhancement). These two techniques have for main advantages to be able to detect fingerprints on a wide range of substrates (*e.g.*, porous, non-porous, semi-porous, adhesive, wetted). In sequence, MMD/SMD are generally applied after conventional techniques and often opposed to PD. In a recent article, Moret and Bécue (304) present the latest evolution of the technique (“SMD-II”), encompassing a detailed recipe and application protocol. Briefly, SMD-II has been thought to be compatible with operational use (*i.e.*, simplified synthesis, increased volume of colloidal gold per synthesis allowing storage for further use, no more need for temperature and pH monitoring), more efficient (*i.e.*, +50 marks detected compared to SMD-I in an experiment involving 14 substrates and marks aged from one month to two years), and more robust towards some porous substrates. In 2013, Charlton *et al.* evaluated the performance of MMD for detecting marks on a particularly challenging substrates: cling film (303). In their study, the authors considered the original formulation of MMD (“MMD-I”), five different brands of cling films (PE- and PVC-based), on which depletive series of natural marks were left. Some of the cling films were used for comparing MMD with other detection techniques (*i.e.*, CA+BY40, VMD_{Ag}, Fe-BPS) while others were exposed to various operational-like scenario (*i.e.*, exposure to drug contamination, immersion in water, simulation of drug wraps, realistically handling of cling films). Their conclusions were the following: (a) on dry and clean substrates, MMD detects more marks on PE- and PVC-based cling films than the other techniques; (b) MMD succeeded in detecting marks on drug-contaminated substrates with little effect of the contaminant, except for mephedrone, MDMA and cannabis resin, which resulted in unwanted background staining; (c) MMD succeeded in detecting marks on substrates immersed for up to 50 hours; (d) the wrapping of the cling film did not prevent the detection of fingerprints, but the authors observed mirror-imaged ridge patterns due to a transfer of secretion caused by the wrapping process; (e) little benefit is obtained from the sequential application of MMD before or after VMD_{Ag}/Fe-BPS, no additional mark/ridge detail being observed. Moreover, a detrimental effect of CA was observed on the subsequent application of MMD. Consequently, MMD is currently proposed as the best-so-far technique to detect fingerprints on PE- and PVC-based cling films, and should be used as a stand-alone technique rather than in sequence.

2.3.7 T/ Immunodetection

Unbound antibodies: Immunodetection of antigenic targets present in fingerprints can be performed by using unbound antibodies (not attached to the surface of a carrier, such as NPs). In a preliminary study, simultaneous detection of two antigenic targets (*i.e.*, dermcidin and HSA) has been performed by using two different fluorophores (322); This study confirmed the presence of dermcidin at the pore sites. In another study, immunodetection of

dermcidin was performed on natural marks left on various substrates (e.g., metal, plastic, ceramic, wood, paper, thermal paper) (323); At the exception of laminated chipboard and copy paper, successful results were obtained. The implementation of immunodetection subsequently to conventional fingerprint detection techniques has finally been assessed (323, 324) – See details below. In another study, immunodetection of various antigens (i.e., hIgG, EGF, lysozyme, dermcidin) was combined with electrochemiluminescence imaging (325).

Antibodies-NP: In a different approach, immunodetection is performed by antibodies bound to a carrier. In that case, NPs are generally chosen to offer additional properties (such as a magnetic core). In a proof of concept study, antibody-functionalized gold NPs were used to target different antigens present in secretion residue (i.e., hIgG, EGF, lysozyme), before being enhanced through metal reduction in solution (318).

DNA aptamers: DNA aptamers are short DNA strand able to specifically recognize a molecular target (similar to the recognition of antigens by antibodies). A couple of preliminary studies considered the use of lysozyme-binding aptamers attached to UC NPs (320), silver nanocrystals (319), or SERS probes (321) to detect fingerprints on non-porous substrates.

Used acronyms: **BY40** (basic yellow 40), **CA** (cyanoacrylate or cyanoacrylate fuming), **EGF** (epidermal growth factor), **hIgG** (human immunoglobulin G), **HSA** (human serum albumin), **IND/Zn** (1,2-indanedione combined with zinc chloride), **LCA** (Lumicyano, one-step luminescent CA), **NIN** (ninhydrin), **NPs** (nanoparticles), **PD** (physical developer), **SERS** (surface-enhanced Raman spectroscopy), **UC** (upconversion)

Impact of conventional detection techniques – van Dam *et al.* (323, 324) assessed the possibility to implement immunodetection after the application of conventional detection techniques. Natural fingerprints were first left on two substrates (*i.e.*, nitrocellulose and glass) before being processed for detection accordingly (*i.e.*, for nitrocellulose: NIN, IND/Zn, IND/Zn→NIN, PD; for glass: magnetic powder, CA, CA+BY40, LCA, PolyCyano UV). Immunodetection of dermcidin was then carried out. In both studies, the presence of dermcidin was successfully enhanced after almost all detection techniques, proving that antigenic sites are still available for immunodetection. The two exceptions are LCA and PolyCyano UV, for which detrimental effects were too important and which are consequently not recommended if immunodetection is scheduled in the sequence – [*Note: if both these studies showed that immunodetection can still be performed in sequence with conventional techniques, the authors did not investigate the potential loss of antigenic sites caused by the application of the detection techniques, by comparing their results with a direct immunodetection of latent fingerprints*]

2.3.8 S/ Adhesives and tapes

Practice-oriented studies: Olenik briefly described the use of a 0.2% (w/v) formulation of BY40 (water-ethanol 25:75%), applied as a CA staining dye on duct tapes (326).

Future prospects: A new range of fluorescent dyes (based on an indole structure) were applied in aqueous solution to detect fingerprints on the adhesive side of tapes (327); In this preliminary study, promising results were obtained in terms of contrast and sensitivity. A cadmium-based QD suspension (water) has been applied to detect marks on adhesives (312).

Used acronyms: **BY40** (basic yellow 40), **CA** (cyanoacrylate or cyanoacrylate fuming), **QD** (quantum dot)

2.3.9 S/ Banknotes

Practice-oriented studies: A thorough study aiming at providing recommendations to detect fingermarks on (Canadian) polymer banknotes has been carried out, leading to an optimized detection sequence (251) and photographic/imaging conditions (328) – See below for details.

Used acronyms: **BY40** (basic yellow 40), **CA** (cyanoacrylate or cyanoacrylate fuming), **R6G** (rhodamine 6G), **VMD** (vacuum metal deposition), **VMD_{Au/Zn}** (conventional gold/zinc VMD)

(Canadian) Polymer banknote – In their first study, Lam *et al.* (251) considered 50CAD polymer banknotes. After a thorough experimental design, they confirmed that the sequence “CA → BY40/R6G (locally/clear windows) → VMD_{Au/Zn} → BY40/R6G (if insufficient ridge detail so far/whole item)” was the most effective in terms of mark detection compared to any other combinations. If CA gave relatively poor results on its own, it appears to participate to the success of the subsequent techniques (*i.e.*, VMD and – logically – dye-staining). In the final step, dyes were readily applied on the VMD-processed marks and rinsed off by gently running water over the substrate. Both dyes behave similarly (with a preference for R6G, maybe due to the use of a LASER for the observations in luminescence). The authors observed an increase in ridge details when dyes are applied subsequently to VMD (especially true for marks lacking of ridge details after VMD). When processing casework-related banknotes, it appeared that marks were detected at each step of the sequence, confirming the importance of carrying out a sequence to its end, when possible (251). In their second study, Lam (328) proposed photographic/imaging recommendations to optimize the recording of the detected marks after each technique. Finally, it should be noted that both these studies have been performed with sebum-rich marks which is justified by the harsh Canadian climate preventing the presence of natural secretions on the donors’ fingertips.

2.3.10 S/ Fabrics

Practice-oriented studies: Two studies aimed at assessing the performance of VMD for the recovery of grab marks on fabrics (329, 330) – See details below.

Future prospects: The possibility to transfer blood-contaminated fingermarks from fabrics using an alginate gel, followed by chemical enhancement using amido black, has been explored (331); If promising results were obtained on dark-patterned silk, detrimental effects caused by the lifting procedure were observed on the other fabrics, meaning that further optimization studies are still required. In a previous study on the same topic, Munro *et al.* concluded that alginate lifting led to overall poor results, with a lack of transferred ridge details (332). IR thermal imaging was used to enhance the presence of blood marks on dark (acrylic and polyester) fabrics after exposition to steam (333); This technique is based on the diffuse reflection of IR by blood, which is further enhanced by the addition of steam. Electrostatic dust print lifter has been applied on grabbed fabrics as a way to promote the transfer of biological material (334); If no ridge details were observed [Note: it was not the purpose of the experiment], this technique showed some potential in terms of touch-DNA but requires further development.

Used acronyms: **BY40** (basic yellow 40), **CA** (cyanoacrylate or cyanoacrylate fuming), **IR** (infrared), **VMD** (vacuum metal deposition), **VMD_{Ag}** (silver-based monometallic VMD), **VMD_{Au/Zn}** (conventional gold/zinc VMD)

Grab impressions – The use of VMD_{Ag} to detect grab impressions on dark fabrics has been evaluated (329), as well as the comparison between VMD_{Au/Zn} and CA+BY40 (330). Both studies were based on a similar experimental protocol including four different fabrics (*i.e.*, satin, polyester, cotton, and polycotton), 15 donors, and marks aged from 1 day to >1 month.

They differ only by the color of the fabrics (*i.e.*, dark (329) and white (330)) and by the deposition protocols (*i.e.*, “grabbing” and “pushing” (329) and “grabbing” only (330)). In the first study, VMD_{Ag} gave good ridge details on polyester (best), followed by satin, but failed in giving ridge details for cotton and polycotton. Among the other parameters influencing the performances, a strong influence of the donors has been observed, while the age of the marks as well as their deposition protocol had a limited impact (nevertheless: in favour of the press protocol for all fabrics). The main advantage of VMD_{Ag} lies in the resulting contrast (light-colored over dark substrate) as well as in the fact that only one metal is vaporized (compared to VMD_{Au/Zn}). In the second study, VMD_{Au/Zn} and CA+BY40 were compared in their ability to detect grab marks on white-colored fabrics. In overall, VMD_{Au/Zn} gave better results than CA (which would rather be compatible with smoother manmade fabrics). The conclusions regarding the influencing parameters were similar to the first study, with a strong influence from the substrate (*i.e.*, nylon gave the best results, followed by polycotton, polyester, and cotton) and the variability between donors. Unfortunately, these two studies were not jointly discussed to provide general guidelines regarding the choice between VMD_{Ag} and VMD_{Au/Zn}. Finally, it should be noted that even if no ridge details were detected, VMD can provide indications regarding a contact and hence orienting the collection of touch-DNA.

2.3.11 S/ Metal and cartridge cases

Fundamental studies: Wightman et al. tried to offer a better understanding of the detection mechanisms related with detection techniques applied to metallic surfaces (e.g., thermal oxidation, anodizing, oxidation induced by iodine, ammonium sulphide and peroxide, water-/ acid-induced corrosion) (335). Aging of fingermarks left on brass has been studied using silver electroless deposition (223).

Practice-oriented studies: Detection of fingermarks on (fired) brass cartridge cases has been extensively studied, including the determination of the best sequence for fired and unfired cases (336) – See details below, as well as the proposition of new techniques such as cold patination (337) or inorganic aqueous electrolytes (338, 339). Digital reconstruction of fingermarks left on cylindrical objects (such as cartridge cases) was described and optimized, using digital stitching of successive pictures taken while rotating the item (340).

Future prospects: The phenomenon of metal corrosion induced by secretion residues has been studied using electrochemistry and X-ray photoelectron spectroscopy (341), as well as the application of heat to detect marks on metals (342). Deposition of electrochromic copolymer films of pyrrole and EDOT has been proposed as a new technique to detect marks on stainless steel (343, 344); In this approach, secretion residue act as a mask and prevent electrodeposition on the ridges, leading to reverse detection. Similarly, electrochemical reduction of graphene oxide has been proposed (345).

Used acronyms: **BY40** (basic yellow 40), **CA** (cyanoacrylate or cyanoacrylate fuming), **EDOT** (3,4-ethylenedioxythiophene), **GB** (gun blueing), **H₂O_{2ac}** (acidified hydrogen peroxide)

Cartridges cases – The detection of fingermarks on unfired and fired cartridge can be challenging, especially for the latter category. In their study, Girelli *et al.* (336) compared various detection techniques/sequences (*i.e.*, powder dusting, GB, H₂O_{2ac}, and CA followed by BY40, powder, or GB). They first conducted experiments on (heated) brass discs, then on unfired and fired brass cartridge cases. In case of fired cartridges, natural fingermarks were left on the cases which were then immediately fired. The fingermarks (left on metal discs, on unfired cases, and on fired cases) were processed after 1 day, 1 week and 2 weeks. The authors concluded that the sequence “CA → GB → BY40” was the best for fired and unfired cartridge cases. The firing process seems to cause most of the damages (compared to the

mechanical cycling of the cartridge inside the gun), resulting in most of the remaining ridge details being located at the base of the cartridges. This is consistent with previous publications in the field (not cited in this report). Finally, a peculiar behavior has been encountered with brass discs heated up to 200C and processed with GB, with the obtaining of reverse development (*i.e.*, darkened ridges on light background). This phenomenon has not been explained.

2.3.12 S/ Skin and leather

Practice-oriented studies: An extensive study aimed at proposing a detection sequence adapted to (artificial) leather (296) – See details below. The sequence “2% SSA (fixating) → HR (staining) → water (rinsing)” has been proposed for the detection of blood marks on skin (346); HR has been preferred above the methanol-based AB and LCV for toxicity and efficiency reasons, respectively. The use of an electrostatic lifter has been proposed to collect dust-/dirt-contaminated fingerprints from skin (347).

Used acronyms: **AB** (amido black), **BPS** (black powder suspension), **C-BPS** (carbon-based BPS), **CA** (cyanoacrylate or cyanoacrylate fuming), **DFO** (1,8-diaza-9-fluorenone), **Fe-BPS** (iron oxide-based BPS), **HR** (Hungarian red), **LCV** (leuco crystal violet), **MMD** (multi-metal deposition), **NIN** (ninhydrin), **PD** (physical developer), **SSA** (5-sulfosalicylic acid), **Ti-WPS** (titanium dioxide-based WPS), **VMD** (vacuum metal deposition), **WPS** (white powder suspension)

Processing of (artificial) leather – The effectiveness of 14 fingerprint detection techniques was assessed when applied on leather and artificial/faux leather items (296). These two substrates are considered as difficult substrates in the context of fingerprint detection. A preliminary trial allowed the authors to determine which detection techniques are able to detect marks on dark- and light-colored leather-based items (genuine and artificial). For this part of the study, favourable circumstances were considered (*i.e.*, fresh marks) and 14 techniques were compared (*i.e.*, AgNO₃, C-BPS, CA, DFO, Fe-BPS, gel lifting, iodine, MMD, NIN, PD, black magnetic and luminescent powder dusting, Ti-WPS, VMD). In a second part of their study, three techniques which passed the first step (*i.e.*, CA, Fe-BPS and C-BPS) were applied on 2-day-old and 1-week-old fingerprints. On overall, the recovery rates were extremely low on genuine leather, with a lot of background staining upon application of BPS. If all three techniques having passed the first trial led to some positive results for marks up to two days, only Fe-BPS gave positive results on older marks. Regarding artificial leather, the recovery rates were higher than on natural leather, with less background staining due to BPS. All three techniques can be recommended for application onto artificial leather, which is an advantage as it is not always easy to determine if a leather-based item is made of genuine or artificial leather. Finally, please note that dye-staining of CA was not considered (mainly for issues related with background staining) and that this study was conducted before the availability of one-step luminescent CA.

2.3.13 S/ Thermal papers

Practice-oriented studies (observation): A high-intensity UV-A source (in that case: a 250W/m² LED torch emitting at 365nm) can be used to visualize latent marks on the thermal side of papers (348); In case of detection, the ridges appear darker than the substrate.

Photography in the NIR range has been applied to help improving the contrast on chemically-processed thermal papers presenting strong background staining (349) – [Note: formulations of amino acid reagents not adapted to thermal papers were used in this study].

Practice-oriented studies (treatment): An optimized detection sequence has been proposed for the processing of thermal papers (350) – See details below. The development of formulations preventing the darkening of thermal papers upon processing led to the following propositions: addition of PVP in a conventional DFO solution before its application (351),

optimization of IND/Zn, NIN, and DFO formulations (352), and assessment of optimized IND and ThermaNIN formulations for the Illinois State Police Latent Prints Procedures Manual (353); All studies led to good detection performances. The sequence “2% SSA (fixating) → AB (staining) → WEAA (rinsing)” has been proposed for the detection of blood marks on both sides of a thermal paper (354); LCV and HR have been found to be inadequate. The monitored application of heat to detect marks on the thermal side of thermal papers has been assessed by different groups (355-357) and a “control” test proposed (358) – See details below.

Future prospects: Immunolabeling has been considered as a way to detect marks on various substrates, among which thermal papers (359) – See section 2.3.7 for details.

Used acronyms: **AB** (amido black), **DABCO** (1,4-Diazabicyclo[2.2.2]octane), **DFO** (1,8-diaza-9-fluorenone), **HPS** (Hot Print System), **HR** (Hungarian red), **IND/Zn** (1,2-indanedione combined with zinc chloride), **LCV** (leuco crystal violet), **LED** (light-emitting diode), **NIN** (ninhydrin), **NIR** (near infrared), **PVP** (polyvinylpyrrolidone), **SSA** (5-sulfosalicylic acid), **UV** (ultraviolet), **WEAA** (water – ethanol – acetic acid)

Detection sequence – A study aimed at proposing an updated/optimized sequence of detection for the processing of thermal papers (350). After a selection step which encompassed 19 techniques compatible with thermal papers, the proposed detection sequence has been validated through a pseudo-operational test. Mostly based on amino acid reagents (*i.e.*, IND/Zn and NIN), the final sequence offers a choice to the operator: (a) considering formulations specifically designed for thermal papers (no risk of darkening) or (b) applying conventional formulations followed by a “whitening agent” (*i.e.*, DABCO) in case of unwanted darkening. DABCO chemically reverses the darkening of the thermal paper while preserving the detected fingermarks. The first approach gave the best results but was the most expensive, compared to the second one (very good results and cost effective). These two ways of doing (*i.e.*, preventing or getting rid of the darkening) are commonly encountered in the literature related with thermal papers, with no consensus about the best way of doing.

Hot Print System (Consolite Forensics Ltd, UK) – The HPS is a device aiming at detecting marks on the thermal side of thermal paper through the monitored application of heat. Three studies were carried out to assess its performances compared to ThermaNin (355) or dry-contact IND/Zn (357), and through the processing of thermal papers from four countries (*i.e.*, Australia, China, United Kingdom, and United States) (356). In a first study, Bond concluded that the controlled application of heat resulted in more ridge details compared to ThermaNIN and was quicker (less than a minute vs. 12 hours) (355). Moreover, he observed that the use of HPS had no effect on the (subsequent) application of NIN on the non-thermal side. In her study, Goel (357) concluded that the application of dry-contact IND/Zn resulted in better mark quality (more ridge details) than with the HPS, for which the detected marks were of low quality and faded quickly. It should be noted that sebum-rich marks were used in this study, which could be explained by the harsh climate conditions encountered in Canada. No consensus has consequently been reached, but the highest sensitivity of IND/Zn (luminescence + formulation adapted for thermal papers) compared to ThermaNin may play in favour of Goel’s study. In a second study, Bond collected 288 printed paper receipts from four countries, left marks on them, and processed them with a device which can be likened to the HPS (356). Thermal papers from China/US differed from those originating from UK/Australia on three aspects: (a) mode of detection, (b) fading of the detected marks, and (c) optimized detection temperature. About the modes of detection, he observed two main behaviours: “normal” which consists in dark ridges on colourless/white paper, and “reverse” which consists in colourless ridges on a darkened substrate. Both

modes of detection have been encountered with thermal papers originating from US and China, while only the “normal” mode has been observed for UK and Australia. Thermal papers from UK and Australia were resistant to fading, contrarily to most of the substrates from US and China (which faded in one day). Finally, higher temperatures were required to detect marks on thermal papers from US and China (64-71°C and 75-95°C, respectively) compared to UK and Australia (43-50°C). Finally, a calibration test has been proposed as a quality control assessing that the right amount of heat has been applied on a processed item (358). This test is composed of a water/glycerol emulsion mixed with various amounts of butylene glycol, which is known to induce a colour change of thermal papers at specific temperatures.

2.3.14 C/ Arson scenes

Practice-oriented studies: In an attempt to recover and detect fingermarks on items recovered from an arson scene, three different soot removal methods (i.e., tape lifting, NaOH solution, and liquid latex casting) and four detection techniques (i.e., black magnetic and aluminium powders, black powder suspension, and CA+BY40) have been assessed and compared (360); Their results confirmed what has already been published on this topic – See below for details. In the same context, fluorescent dye-doped ZnCO₃ SPR has been successfully applied on items exposed to elevated temperature, soot, then water (361); The authors also noticed that SPR failed in detecting marks above a particular temperature, different for each substrate.

Used acronyms: **BY40** (basic yellow 40), **CA** (cyanoacrylate or cyanoacrylate fuming), **SPR** (small particle reagent)

Soot removal and mark recovery – In their study, Gardner *et al.* (360) carried on a thorough study by considering burned cars as starting scenario, requiring the detection of fingermarks from recovered rear view mirrors. The mirrors were put in a cremation oven to control the temperatures and exposition time (without soot and smoke), as well as in a shipping container in which fire were simulated (including soot and smoke). They assessed the efficiency of three soot removal methods (i.e., tape lifting, NaOH solution, and liquid latex casting) and four different detection techniques (i.e., black magnetic and aluminium powders, black powder suspension, and CA+BY40). About soot removal: no significant statistical difference has been observed between the three tested techniques. About the effect of temperature: a strong influence of the temperature on the recovery success has been observed, with most marks recovered at 300C while no identifiable marks were observed at 600C. About fingermark detection: CA+BY40 and black magnetic powder gave the overall best results, followed by aluminium powder and black powder suspension. However, no significant statistical difference has been observed between these four techniques.

2.3.15 C/ Blood marks

Practice-oriented studies: The addition of R6G in the fixating bath (i.e., SSA+R6G) was assessed/optimized to obtain luminescent marks prior to application of blood reagents (i.e., AB and LCV) (362); Successful results were obtained with no detrimental effect on the performance of the subsequent blood reagents. The performance of numerous blood reagents has been assessed by considering fingermarks and shoemarks left on a variety of household surfaces (i.e., non-porous: painted drywall, laminate wood, linoleum, painted metal, treated cement; porous: non-painted drywall, non-treated cement, carpet) (363); Results were in accordance with previous studies (i.e., for non-porous substrates: AY7 > AB > HR; for porous substrates: NIN > AB > DFO). The performance of four blood reagents (i.e., AB_w, AY7, CBB, LCV, and LCV→AB_m) has been assessed by considering depletive series of blood marks, as well as dilution series of blood stains, on various substrates (e.g., paper,

wood, plastic, glass, metal, ceramic) (364); A recommendation table combining the nature of the substrates with the initial visibility of blood marks is proposed to choose the best reagent, CBB being considered as a good alternative for both porous and non-porous substrates. Genipin and lawsone failed to compete with NIN or DFO to detect blood marks on paper (365). SPR-W has been assessed as the best technique to detect blood marks on a dark substrate (i.e., black PP sheet) (295) – See details below. The problematics of blood marks on fabrics (331, 333), on skin (346) and on thermal papers (354) have been covered in sections 2.3.10, 2.3.12 and 2.3.13, respectively.

Future prospects: In direct continuation of works performed on blood shoemarks, Munro et al. determined that blood fingermark lifting is not recommended for non-porous substrates, given the poor overall performance due to a lack of transferred ridge details (332); Nevertheless, the addition of protein stain in the alginate mixture led to a promising alternative (in situ reaction). MALDI-MS was used to provide information about the composition of AB-processed blood marks (366). The use of HSI to detect and identify(*) blood fingermarks on various substrates has been assessed (367-369), as well as its application in sequence with a conventional blood reagent (370). [*Note: “identify” standing for the determination of the nature of the fluid]

Used acronyms: **AB** (amido black or acid black 1), **AB_m** (methanol-based AB formulation), **AB_w** (water-based AB formulation), **AY7** (acid yellow 7), **CA** (cyanoacrylate or cyanoacrylate fuming), **CBB** (coomassie brilliant blue), **DFO** (1,8-diaza-9-fluorenone), **HR** (Hungarian red), **HSI** (hyperspectral imaging), **LCA** (Lumicyano, one-step luminescent CA), **LCA_{x%}** (Lumicyano solution containing x% of Lumicyano powder), **LCV** (leuco crystal violet), **MALDI** (matrix-assisted laser desorption ionization), **MS** (mass spectrometry), **NIN** (ninhydrin), **R6G** (rhodamine 6G), **PP** (polypropylene), **SPR-W** (white-colored small particle reagent), **SSA** (5-sulfosalicylic acid)

Dark substrates – The processing of blood fingermarks on a dark substrate (i.e., PP plastic sheet) was assessed by considering depletive series of 1-day-old to 1-year-old marks processed by four reagents (i.e., AY7, SPR-W, CA, LCA_{1%}) applied alone or in sequence (295). Quite surprisingly, SPR-W (applied as sole technique) showed the best performances in terms of contrast, while the sequence “CA → AY7” gave more marks of better quality. Among other conclusions: no influence of the age has been observed; full DNA profiles could be obtained from the first mark of depletive series, with no apparent detrimental effect of the applied reagents; the sequence (L)CA → SPR-W is not recommended. [*Note: at the time of this study, CST sold Lumicyano as a premix solution, which contained 1% of dye; explaining the choice for the following notation: LCA_{1%}*]

Hyperspectral imaging – Cadd et al. (367) started from the observation that blood absorbs visible wavelengths between 400 and 500 nm (due to the presence of haemoglobin) to develop an HSI-based method to detect and identify the nature of blood marks on ceramic tiles as well as on various substrates (i.e., light- and dark-colored ceramic tiles, glass, plastics, paper, cardboard, cotton, wood, pig skin) (368). In their last study, the authors assessed the performance of their system in sequence with AB, a protein-stain commonly used to detect blood marks (370). Depletive series of blood marks and dilution series of bloodstains were considered to assess the sensitivity of the method. Deposition of fingermarks contaminated with a whole range of red/brown substances and protein-rich substances (knowing to react with AB) was carried out to assess the selectivity of the method (risks of “false positives”). The presence of a narrow and intense absorption peak at 415nm (+ two weaker bands between 500 and 600nm) was determined as the main identification criteria for blood. Promising results were obtained from these studies, in terms of selectivity and sensitivity. Gain compared to conventional imaging is rather to be found on

dark substrates, for which the optical contrast is difficult to set. Finally, HSI present the additional advantages of being quick, contactless and non-destructive.

2.3.16 C/ Contaminations

Practice-oriented studies: The effect of fingerprint detection techniques on the subsequent recovery/analysis of drug and explosive residues was explored (371-376) – See details below. Also, the presence of various household contaminants on fingertips was considered in a study aiming at assessing the persistence of fingerprints exposed to (detrimental) environmental elements (222).

(Illicit) Drugs (handling): Fast Blue B was proposed as a new reagent to detect THC-rich fingerprints, as it produces a red complex upon reaction with cannabinoids (e.g., THC, CBD, and CBN) (377); Promising results were obtained but this approach should be carefully thought in a forensic context as it only detects THC-containing fingerprints + the link with the activity of handling cannabinoid may not be straightforward. Chemical imaging of contaminated fingerprints using DESI-MSI and ToF-SIMS, jointly with a printed pattern of cocaine, heroin, and methamphetamine spots, was proposed as a way to quantify drugs in secretion residues (378); A strong influence of the substrates was observed, especially for heroin and methamphetamine. An anti-cocaine-based immunoassay was developed to quantitatively assess the presence of cocaine in banknotes and fingerprints (379). Aptamer-functionalized NPs were used to detect cocaine-contaminated fingerprints (320). Other studies involved the analysis of drug-spiked fingerprints: oily marks using DAPNe-NSI-MS (380), sebum-rich fingertips using DART-MS and MALDI-MS (375).

Endogenous metabolites (drug consumption): Fingerprints from people attending a drug treatment service were analysed through DESI, LESA-MS, MALDI-IMS-MS/MS, and SIMS to detect illicit drugs and their metabolites (204, 381); Good correlation were found for DESI, LESA and MALDI in comparison with oral fluids, while the sensitivity of SIMS was found to be insufficient. Various drugs and metabolites were analysed in fingerprints using LC/MS (376, 382); The main outcome of these studies is to evaluate how fingerprints could be used as alternatives for body fluids (drug testing). Finally, chemical imaging of drugs and their metabolites in natural/artificial secretions is covered in section 2.3.19 – briefly: DEFFI-MSI (208), DIOS-MSI (various drugs) (383), MALDI-(ToF-)MSI (various drugs) (372-374, 384), SALDI-MSI (289).

Explosives: Functionalized NPs were proposed as sensors to detect the presence of explosive residues in fingerprints: aptamer-functionalized gold NPs to detect RDX (385), aptamer-functionalized silver nanoclusters (319) and dual-emitting QD nanohybrid (313) to detect the presence of TNT. Explosive-contaminated fingerprints were analysed through different techniques: LESA-MS (RTX and TNT) (204), ECL-based image contrast technology (TNT) (386), laser pointer-based Raman spectroscopy (387), IMS (after having been dry-dusted and lifted) (388), photothermal-imaging (TNT detected through the fluorescence quenching of Cu₇S₄ nanocomposites) (316). Explosive residues (i.e., ammonium nitrate, black powder, smokeless gun powder, dynamite) were detected/mapped in finger-/handmarks by using NIR-HSI (389, 390) and chemically-modified glass surface based on DPA, pDMAC and pDMAB to detect urea nitrate (391). Finally, chemical imaging of explosives in natural/artificial secretions is covered in section 2.3.19 – briefly: DEFFI-MSI (RDX) (208), MALDI-(ToF-)MSI (RDX and TNT) (372, 373, 384).

Used acronyms: **CA** (cyanoacrylate or cyanoacrylate fuming), **CBD** (cannabidiol), **CBN** (cannabinol), **DAPNe** (direct analyte-probed nanoextraction), **DART** (direct analysis in real time), **DEFFI** (desorption electro-flow focusing ionization), **DESI** (desorption electrospray ionization), **DIOS** (desorption ionisation on porous silicon), **pDMAB** (p-

dimethylaminobenzaldehyde), **pDMAC** (p-dimethylaminocinnamaldehyde), **DPA** (9,10-diphenylanthracene), **ECL** (electrochemiluminescence), **HSI** (hyperspectral imaging), **IMS** (ion-mobility spectrometry), **IND/Zn** (1,2-indanedione combined with zinc chloride), **LC** (liquid chromatography), **LESA** (liquid extraction surface analysis), **MALDI** (matrix assisted laser desorption ionization), **MS** (mass spectrometry), **MSI** (MS combined with imaging), **NIN** (ninhydrin), **NIR** (near infrared), **NPs** (nanoparticles), **NSI** (nanospray ionization), **PD** (physical developer), **PETN** (pentaerythritol tetranitrate), **QD** (quantum dot), **R6G** (rhodamine 6G), **RDX** (hexahydro-1,3,5-trinitro-1,3,5-triazinane), **SIMS** (secondary ion mass spectrometry), **THC** (Δ -9-tetrahydrocannabinol), **TNT** (2,4,6-trinitrotoluene), **ToF** (time of flight), **VMD** (vacuum metal deposition)

Sensors vs detection – As it can be seen, several publications dealing with contaminated fingermarks were focused on the development of drug/explosive sensors rather than detection techniques (313, 376, 379, 381-383, 385, 388, 391). They were nevertheless cited, despite the fact that they deviate from the scope of this review.

Fingermark detection vs. contamination residues – When considering the recovery and analysis of contaminants in secretion residue (caused by the handling of illicit drugs or explosives, for example), it appears necessary to assess the impact of conventional fingermark detection techniques on these contaminants. Indeed, most of the fingermarks are initially latent and would consequently require to be detected beforehand. In that context, King *et al.* (371) conducted a research aiming at first estimating the quantity of explosive residue left in fingermarks subsequently to the handling of bulk material, followed by the quantities remaining after conventional detection techniques were applied. To reach this goal, they considered four substrates (*i.e.*, paper, glass, plastic bags, and aluminium foil), five explosive-related compounds (*i.e.*, TNT, PETN, RDX, chlorate and nitrate ions), and five detection techniques applied individually then in three distinct sequences (*i.e.*, IND/Zn → NIN → PD; black magnetic powder; CA → R6G). It was observed that explosive residues can still be detected after the application of detection techniques, with varying losses according to the substrate and the applied technique(s). Briefly: magnetic powder showed minimal effect; CA resulted in losses on plastic and aluminium, supposedly through entrapment of the molecules of interest in the polymer matrix; IND/Zn and NIN caused some loss of the organic explosives and nitrate ions, supposedly through mechanical removal during the dipping process or the use of absorbent paper; water-based treatments (*e.g.*, R6G, PD) resulted in a great loss of the considered compounds (especially inorganic ions, TNT, and RDX), only PETN persisted after PD. As a conclusion, it is recommended to limit the number of detection techniques to be applied on an item, and to adapt some application protocols if the recovery, mapping, or analysis of explosive compounds is scheduled.

In the same context, chemical imaging is often considered for the mapping and analysis of explosive residues contained in fingermarks. If many studies consider the use of chemical imaging as a stand-alone technique, using artificially-spiked fingermarks, other explored its application in the frame of realistic handling scenario (372), as well as the impact of conventional detection techniques on the performance of chemical imaging (applied subsequently) (373, 374). In a first study, Kaplan-Sandquist *et al.* showed that artificial secretions (*i.e.*, eccrine and sebaceous pads) are not suitable for the simulation of natural fingermarks but may help in configuring the instrumentation (372). They also showed that handling whole or broken drug pills (realistic scenario) resulted in an insufficient quantity of transferred compounds, which were not detected by MALDI-ToF-MSI, and that the use of drug/explosive powders is consequently still required. Finally, their conclusions met those of King *et al.* (on the persistence of explosive residues) by successfully mapping drug/explosive residues after the application of detection techniques (*i.e.*, black powder and CA). In another study, the same authors evaluated the performance of MALDI-ToF-MSI when

used subsequently to (a) black powder dusting, (b) MALDI matrix spraying, (c) black powder → lifting, and (d) CA → black powder (373). For this study, fingertips spiked with drug/explosive powders (from evaporated solutions) were considered for the deposition of contaminated fingerprints on aluminium. Results showed that powder dusting and MALDI matrix spraying led to the highest average recovery rates (88%), followed by CA (52%) and lifting (18%). It was also shown that the recovery rates were dependent of the targeted compounds. In their study, Groeneveld *et al.* (392) considered 17 drug-related compounds/metabolites, two scenarios (*i.e.*, “handling” and “abuse/consumption”) both based on artificially-spiked fingertips, and different detection sequences based on CA (+BY40) and VMD, MALDI-MSI being applied subsequently to the detection sequences. As a result, it was shown that VMD is much more adapted to MALDI-MSI than CA, which corroborates another study.

2.3.17 C/ Immersed items

Practice-oriented studies: Several studies aimed at determining the possibility to detect fingerprints on items that have been exposed to freshwater (393-395), sea water (393, 396), or to everyday liquids (397) – See details below.

Future prospects: Phase transfer catalyst has been proposed for the detection of fingerprints on immersed items (398).

Used acronyms: **Fe-BPS** (iron oxide-based black powder suspension), **GV** (gentian violet), **ORO** (oil red O), **PD** (physical developer), **PD_{HO}** (UK Home Office formulation of PD), **PD_{Tw20}** (PD based on Tween 20 instead of Synperonic-N), **SB** (Sudan black), **SPR** (small particle reagent), **SPR-B** (black-colored SPR), **SPR-W** (white-colored SPR), **uPVC** (unplasticized polyvinyl chloride)

Freshwater – When items are immersed in water, it is known that conventional reagents (such as amino acid reagents) fail in detecting fingerprints, mainly due to the solubilisation of water-soluble components (such as amino acids). In three different studies, people tackled this issue by either studying the degradation process induced by a prolonged immersion in various water types (393), the choice of the best technique to apply on immersed porous substrates (394), or the possibility to leave fingerprints on immersed items and to detect them afterwards (395). To study the detrimental effect of immersion onto fingerprint constituents, Sutton *et al.* immersed various substrates (*i.e.*, stainless steel, uPVC, and glass) bearing eccrine-rich and sebum-rich marks in three types of water (*i.e.*, lake, river, and sea), under laboratory and field conditions, and for times going up to 14 weeks (393). The marks were then processed using ORO, SB, and GV. As expected, eccrine-rich marks were extremely affected by immersion in water, with little or no ridge details left even after a short immersion time. Quite surprisingly, fingerprints left in field conditions led to few – if no – degradation compared to those immersed in laboratory, which showed a substantial drop in quality with time. This observation should be over-balanced by considering that (a) the field substrates were placed into permeable cases, which could have provided increased protection towards water flow and erosion, (b) the laboratory protocol included a complete change of water every week, which may have caused a detrimental flow of liquid, and (c) laboratory conditions may have allowed the development of a microflora in the water tanks. Finally, in terms of reagents, SB and GV performed equally and were both superior to ORO in the laboratory trial, which could indicate that these reagents target different secretion constituents. In a study aiming at determining the best technique to apply on immersed porous substrates, Simmons *et al.* compared PD_{HO} vs. ORO vs. PD_{Tw20} (394). Three different substrates (*i.e.*, white paper, glossy leaflets, and brown cardboard) bearing natural marks aged from 7 to 28 days were immersed in tap water for one hour. As a result, cardboard and leaflets led to no usable ridge details, with strong background staining

observed for leaflets. On white paper, both PD formulations behave similarly, with >80% of successful detection (including 35-38% of very good ridge details), contrarily to ORO which led to poor results (4.5% of successful detection with no usable ridge detail). A PD formulation based on Tween 20 can consequently replace a Synperonic N one. In their study, Castelló *et al.* (395) studied the possibility to leave fingerprints on items which are already immersed, as well as the chance of subsequent detection. Two substrates (*i.e.*, glass and plastic/photocopy transparency sheets) were immersed in tap water while donors were asked to leave “natural” fingerprints. The marks were left in water for 1 to 15 days before being removed, dried, and processed with different techniques (*i.e.*, dry powders, SB and SPR). Results showed that it is possible to leave fingerprints on immersed items, and that all techniques gave good results for up to 3 days of immersion. Black powder resulted in the best performance with the ability to detect marks after 15 days of immersion on both substrates. Powdered SB and SPR succeeded similarly, but on glass only.

[Note: Both Sutton’s and Simmons’ studies led to the conclusion that ORO performed poorly on immersed items (non-porous and porous), compared to other techniques such as SB, GV and PD]

Sea water – The detrimental effect of sea spray (created by the wind over the ocean) onto items bearing fingerprints has been studied by Goldstone *et al.* (396). Their study consisted in exposing glass panels bearing depletive series of fingerprints to actual sea spray (balcony facing the ocean) for one month before processing them; with one of the two glass panels having already been exposed to sea spray for one week before the deposition of the fingerprints. The authors considered the application of eleven detection techniques, composed of various dry powders and powder suspensions (*i.e.*, Fe-BPS, SPR-B and SPR-W, black and white Wetwop™). They noticed no difference between the two glass panels (*i.e.*, clean glass vs. glass already exposed to sea spray before deposition). If all techniques succeeded more or less to detect marks after a one-week exposition time, only Fe-BPS and white Wetwop™ still succeeded in detecting a significant amount of marks after being exposed for one month to sea spray (all other techniques leading to ~0% of success). However, even for these two techniques, exposition to sea spray caused a serious decrease in detection success rates (*e.g.*, Fe-BPS 96% → 67%, white Wetwop™ 95% → 49%). Finally, the authors found that white magnetic powder can be a valid alternative for marks exposed for less than one week to sea spray (89% of success, dropping to 3% after one month). The detrimental role of immersion in sea water has also been studied by Sutton *et al.* (393) – See “Freshwater”.

Everyday liquids – Glass slides bearing depletive series of sebum-rich fingerprints were immersed in various everyday liquids (*i.e.*, tap water, milk, red wine, soft drink, beer, orange juice, and soapy tap water) for 1 to 24 hours (397). Removed items were then processed with either magnetic powder or SPR – Please note that the items devoted to dry powder were first water-rinsed then dried. No or limited effect was observed for milk, wine, soft drinks, beer, and orange juice. However, soapy water led to a significant decrease in quality after 12 hours and to no ridge details after 24 hours. Finally, the authors observed that powder dusting was slightly more effective compared to SPR.

2.3.18 I/ Photography and forensic light sources

Fundamental studies: The optical mechanisms allowing the observation of latent fingerprints on smooth/non-porous substrates using a RUVIS are extensively described (399); By considering the secretion residue reflectivity and the optical surface roughness (scattering ability), it is theoretically determined that the best illumination angle should be set between 10 and 30° when using the 254nm UV radiation, which is in agreement with experimental data.

Practice-oriented studies: A methodology to obtain improved monochrome digital images from a UV-sensitive camera and reflected UV is described (400); Based on the sensor linear response and the camera spectral sensitivity curves, the method is illustrated by using sunscreen lotion-enriched fingerprints left on an enamelled metallic canister. Focus stacking is proposed to extend the depth of field of images recorded on curved items (401); This technique is based on the recording of a series of images focused from the most distant plane in the curved item to the closest, which are then processed by a raster graphics editor software (i.e., Photoshop® in that case). Finally, other publications described how basic enhancement tools (i.e., contrast inversion, intensity levels, and rotation) may alter – or not – image data (402), or proposed alternative digital enhancement protocols (403).

Future prospects: A portable device taking advantage of the light scattering induced by the secretion residue has been proposed (404); Scattering of light is a well-known phenomenon allowing the contactless recording of latent fingerprints on flat non-porous substrates. Various optical contactless imaging techniques have been proposed to record latent fingerprints: imaging ellipsometry (405), which is based on the induced changes in polarization state when light hits the secretion residues, but is currently limited to flat/specular non-porous surfaces of small size; digital stitching of successive pictures of a rotating cylindrical objects (such as cartridge cases) (340); full-band CCD and UV observation camera combined with a 254nm UV excitation light source (406); 3D confocal laser scanning microscopy combined with a feature extraction algorithm (407); home-made setup facilitating the recording of fingerprints on nonporous curved surfaces (408). Two-photon imaging was proposed to image luminescent marks left on a metallic substrate (253). Finally, HSI in UV (409, 410), VIS (411) and NIR (369) has been proposed to image fingerprints on substrates presenting background interference with conventional techniques.

Used acronyms: **CCD** (charge coupled device), **HSI** (hyperspectral imaging), **NIR** (near infrared), **NPs** (nanoparticles), **RUVIS** (reflected UV imaging system), **UC** (upconversion), **UV** (ultraviolet), **VIS** (visible)

Unconventional imaging techniques – Two emerging imaging techniques are reported in the literature, presenting some advantages compared to conventional imaging: NIR luminescence (207, 253, 290-292, 320, 349, 412) and UC (263, 273-275, 277, 278, 281, 317, 320). NIR luminescence has for main advantage to avoid most of the background luminescence issues. UC allows the observation of a material in the visible range while illuminating it at higher wavelengths (generally in the NIR range) which also provides a way to suppress the background luminescence. However, please note that most of the works dealing with UC imaging are unfortunately based on the dry-dusting of NPs (see remark in section 2.3.4). It is awaited that an increased number of techniques will explore these two imaging modes, for they may offer solutions for the detection of fingerprints on particularly difficult substrates.

2.3.19 I/ Chemical imaging

Imaging of latent secretions: Several techniques were applied to image latent fingerprints, among which DEFFI-MSI (artificial mixture of eccrine and sebaceous secretions; lifting tape) (208), DESI-MSI (sebum-rich marks; glass) (413), LAET-MSI (semiconductor-based substrate) (198), MALDI-MSI (sebum-rich marks; stainless steel) (384), MALDI-ToF-NIMS (porous silicon-based substrate) (200), SiALDI-MSI (silver-based substrate) (201). In the same context, chemical imaging has also been used to study the molecular composition of secretion residues using MALDI-MSI (205, 206) or the aging phenomenon with ToF-SIMS (214).

Imaging of contaminated secretions: Several techniques were applied to image ridge patterns artificially-contaminated with drugs or explosives (handling) or to image endogenous metabolites (consumption), among which DEFFI-MSI (lotion, explosives and drugs; artificial secretions; lifting tape) (208), DESI-MSI (drugs; artificial secretions) (378), DIOS-MSI (drugs and endogenous metabolites) (383), MALDI-(ToF)-MSI (drugs and endogenous metabolites) (204, 374), MALDI-(ToF)-MSI (drugs and/or explosives) (372, 373, 384), SALDI-MSI (drugs) (289), ToF-SIMS (drugs; artificial secretions) (378).

Imaging of processed marks: Several techniques were applied subsequently to the detection of fingerprints, using conventional detection techniques or dual-purpose reagents (reagent allowing the visualization of ridge pattern and participating to the imaging step). In brief: FTIR imaging (CA; fabrics) (252), MALDI-MSI (various techniques; various substrates) (392) – See details below, MALDI-(ToF)-MSI (CA, VMD, or dry-dusting; drug- or explosive-contaminated marks) (372-374), MALDI-ToF-NIMS (dry-dusting with dye-doped porous silicon microparticles) (207), SALDI-MSI (dry-dusting; drug-contaminated marks) (289), SERS (aptamer-functionalized nanocomposites) (321); SKP (VMD; metallic substrate) (414), ToF-SIMS (various techniques; various substrates) (415) – See details below.

Other purposes: Chemical imaging has also been used for determining the chronology of deposition of fingerprints and inks on paper using ToF-SIMS (416) or to test an artificial emulsion composed of eccrine and sebaceous constituents (225) – See section 2.2 for details. Reviews were published about the use in forensic science of FTIR imaging (417), MSI (417, 418), SALDI-MS(I) (419), and SERS (420).

Used acronyms: **BPS** (black powder suspension), **BV3** (basic violet 3), **BY40** (basic yellow 40), **C-BPS** (carbon-based BPS), **CA** (cyanoacrylate or cyanoacrylate fuming), **α CHCA** (alpha-cyano-4-hydroxycinnamic acid), **CV** (crystal violet), **DEFFI** (desorption electro-flow focusing ionization), **DESI** (desorption electrospray ionization), **DFO** (1,8-diaza-9-fluorenone), **DIOS** (desorption ionisation on porous silicon), **Fe-BPS** (iron oxide-based BPS), **FTIR** (Fourier transform infrared spectroscopy), **LAET** (laser activated electron tunneling), **MALDI** (matrix assisted laser desorption ionisation), **MS** (mass spectrometry), **MSI** (MS with imaging), **NIMS** (nanostructure imaging MS), **NIN** (ninhydrin), **SALDI** (surface-assisted laser desorption ionization), **SBB** (Sudan Black B), **SERS** (surface-enhanced Raman spectroscopy), **SiALDI** (silver-assisted laser desorption/ionization), **SIMS** (secondary ion MS), **SKP** (scanning Kelvin probe), **SPR** (small particle reagent), **ToF** (time of flight), **VMD** (vacuum metal deposition), **WPS** (white-colored powder suspension)

Chemical imaging/Hyperspectral imaging – Chemical imaging is part of a wider range of application dealing with the collection of extended spectral information along a scanned area. In this report, the term “chemical imaging” encompasses all the techniques related with the mapping of chemical groups/molecules. MS- or FTIR-based techniques are among the most popular in this field. The term “hyperspectral imaging” (HSI) has been associated with all the other techniques dealing with datacubes, mostly through the use of white light combined with a spectrograph (367-370, 389, 390, 409-411).

Chemical imaging vs conventional detection techniques – It is unfortunate that most of the articles referring to chemical imaging (applied to fingerprints) consist in proof-of-concept studies based on (artificially-)enriched fingerprints left on ideal substrates, with overvalued performances disregarding primary forensic interests, and lacking of practical information (such as the scanning time) or of operational perspectives. For these reasons, we chose to describe only two studies aiming at evaluating the performance of chemical imaging when combined with conventional detection techniques: MALDI-MSI (392) and ToF-SIMS (415). In their study, Bradshaw *et al.* first assessed the range of information that can be gained from

MALDI-MSI compared to conventional detection techniques (*i.e.*, dry-dusting, CA+BY40, DFO, NIN, VMD, WPS), then evaluated the compatibility of MALDI-MSI when applied subsequently to these techniques. Finally, they assessed the possibility to introduce a “dual-action powder” able to visually detect fingerprints and participate to the imaging process. For their study, they considered natural marks, left on a versatile range of porous and non-porous substrates, and aged from 0 to 10 days. For the first trial, it was logically shown that MALDI-MSI can bring chemical information, as a compensation for lower quality ridge details. Interesting results were obtained with DFO and NIN. Indeed, as MALDI-MSI maps several constituents, the “dotty ridge” effect obtained with amino acid reagents was not observed with MALDI-MSI (mainly due to the mapping of lipids). However, once put in sequence, MALDI-MSI suffered more or less from detrimental effects caused by the beforehand application of detection techniques. For example, DFO, NIN, CA prevented MALDI-MSI to image/enhance ridge patterns. The only exceptions were TiO₂ dry-dusting and VMD, the sequence “VMD → MALDI-MSI” giving good results in terms of ridge details and chemical information, mostly because gold can act as a signal enhancer for MS. In their third trial, promising results were obtained from mixing TiO₂ powder or SBB with αCHCA (a powder specifically developed for MALDI-MSI) to obtain a “dual-action powder” which could be dusted on items to detect fingerprints and further analyse them. As a conclusion to their study, the authors provided a proposition of operational workflow integrating MALDI-MSI. In another study, fingerprints left on three different substrates (*i.e.*, aluminium foil, grenade handle, and glass immersed in sea water or buried in soil) were processed using conventional detection techniques (*i.e.*, CA+BY40, CA+CV, VMD, SPR, dry powders, Fe- and C-BPS, BV3) and the results compared with ToF-SIMS (415). It should be noted that it is unclear when chemical imaging was performed (*i.e.*, as stand-alone technique or subsequently to fingerprint detection). It seems that the only application of ToF-SIMS subsequently to a detection technique was for the aluminium foil, for which dotty ridges were obtained with CA while continuous ridges were obtained from chemical mapping of the processed marks. On the other substrates, chemical imaging was supposedly applied as a stand-alone technique and compared to the conventional processes. It is difficult to assess this study, since no split marks were considered and the imaging of ridge pattern was limited to a small area (128x128 pixels), requiring two hours to be processed.

Note: the other articles dealing with the use of chemical imaging subsequently to fingerprint detection techniques are described in section 2.3.16, for they are dealing with contaminated fingerprints.

2.3.20 O/ Fingerprint detection and DNA analysis

“Touch-DNA”: “Touch-DNA” can be defined as the genetic material that is extracted from fingerprints, before or after having been processed for detection. Several studies were conducted in this field and are summarized here-below without being thoroughly described (as it would rather be the scope of a review dedicated to genetic material).

Research was conducted to verify the possibility to extract DNA from latent fingerprints (421) or propose a simplified workflow (422), to compare the DNA-shedding propensity of palms and fingers (423), or to assess the possibility to readily stain genetic material contained in latent fingerprints (424). Other studies aimed at evaluating the impact of fingerprint detection techniques on the subsequent recovery of mRNA and/or DNA (review on this topic: (425)): dry-dusting of latent and/or blood marks (426-428), conventional fingerprint detection techniques (e.g., dry-dusting, iodine fuming, IND, CA) (429, 430), blood reagents (*i.e.*, AB, AY7, LCV) (431), lifting tapes (432), and other emerging detection techniques (*i.e.*, CTF) (429, 433). Finally, the possibility to standardize DNA collection and extraction protocols from glass and metallic substrates was proposed, to suit a military

application context (434), as well as the use of ESDA to collect genetic material from porous substrates (435)

Case report: Mitochondrial DNA extracted from a NIN-processed paper towel (partially burned) (436).

Used acronyms: **AB** (amido black), **AY7** (acid yellow 7), **CA** (cyanoacrylate or cyanoacrylate fuming), **CTF** (columnar thin film), **DNA** (deoxyribonucleic acid), **ESDA** (electrostatic detection apparatus), **IND** (1,2-indanedione), **LCV** (leuco crystal violet), **NIN** (ninhydrin), **mRNA** (messenger RNA)

2.3.21 O/ Miscellaneous (detection) techniques

Fundamental studies: The variability and subjectivity of grading processes were assessed by considering 80 IND/Zn-processed marks assessed by 11 individuals (437) – See details below. Devices were proposed to try reproducing the deposition of fingermarks by controlling the force, angle, and time of contact (105, 438).

Challenging substrates: Three publications reported the best ways to detect fingermarks on rocks and stones, which are known to be challenging surfaces in terms of fingermark detection (439-441); One of the key parameters is to determine the porosity of the material as it will drive the choice for the most suitable detection techniques (e.g., magnetic powder, CA, NIN, silver nitrate), but no real consensus emerged from these studies. The processing of Tyvek Large Pak (e.g., from FedEx) and Padded Pak shipping envelopes for fingermark detection was thoroughly explored (442); Modified black Wetwop™ (composed of Wetwop™ + RO/DI water + black powder) and diluted black Wetwop™ (using RO/DI water) were determined to give the best detection results, respectively, with the possibility to reapply the reagents to enhance the weak marks.

Thermal development: The Thermal Fingerprint Developer (TFD-2; Foster+Freeman, UK) is a reagent-free and contactless device aiming at detecting marks on papers using a monitored application of heat. Its efficiency was compared to fingermark detection techniques and its impact on the subsequent application of such techniques have been assessed by two teams, considering various types of porous substrates (443, 444) – See details below. On a similar aspect, a proof-of-concept study presented the use of microwaves to thermally detect marks on paper (445).

Other miscellaneous studies:

- Use of CWL to image fingermarks left on gloves (446), to study fingermark persistence (447) or following by image processing techniques to enhance the contrast of marks (448).
- Determination of the best technique to detect fingermarks on bird of prey feathers and eggs (449); In that case: magnetic powders;
- Detrimental effect of the use of a liquid bandage (e.g. New-Skin® Liquid bandage) on the deposition of ridge skin details (450);
- Successful application of a dry chemical/powder ABC-type extinguisher to detect marks in a clandestine drug synthetic lab (451);
- Effect of (blood-contaminated) fingermark detection techniques on the subsequent recovery of spermatozoa (452);
- Interaction between secretion residue and easy-to-clean surfaces as a way to improve touch screen technology (453);
- Role of tryptophan derivatives in the autofluorescence of aged fingermarks (454);
- Effect of five CBRN decontamination procedures (physical or chemical) on the detection of fingermarks on glass (455); Decontamination procedures induced a strong detrimental effect on ridge details (bleach presenting the most negative effect), but did

not prevent VMD to detect fingermarks even if a loss of contrast is observed for the decontaminated marks;

- Description of an atomizing device based on piezoelectric vibration to generate a reagent spray (e.g., CA, NIN) which can be applied on substrates bearing fingermarks (456); No results presented and no health and safety considerations;
- Use of a dye-containing substrate (457) or fluorescein-embedded nanofibers (458) to collect rolled fingerprints; Beyond the proof of concept of using electrochromism for such an application, the gain of these two techniques compared to conventional methods (e.g., ink or livescan) are highly debatable.

Future prospects: A range of new fingermark detection reagents was proposed in the literature (unless specified: applied on non-porous substrates and observed in luminescence): oxetane-functionalized semiconductor polymer dots (459); 4-dimethylamino-20-hydroxychalcone (NIR luminescent) (412); perylene derivatives (460, 461); silole derivatives (462); HDDCPU and HDDPU diacetylene copolymers (follow-up study, various substrates) (463), pH-dependent polyelectrolyte (464). Among the various emerging techniques, CTF has been proposed to detect sebaceous-rich and blood-contaminated marks on non-porous substrates (429, 433, 465-471). CTF is a method based on low-pressure vaporization of different materials (e.g., metal, inorganic oxide, glass) which aims at detecting fingermarks by enhancing the topology of the secretions. However, we note that all the publications dealing with CTF originate from one single group of research, and the technique requires specific equipments. No independent validation has been published yet.

Used acronyms: **CA** (cyanoacrylate or cyanoacrylate fuming), **CBRN** (chemical, biological, radiological, and nuclear), **CTF** (columnar thin film), **CWL** (chromatic white light sensor), **pDMAB** (p-dimethylaminobenzaldehyde), **HDDCPU** (2,4-hexadiyne-1,6-bis[p-chlorophenylurethane]), **HDDPU** (2,4-hexadiyne-1,6-bis[phenylurethane]), **IND/Zn** (1,2-indanedione combined with zinc chloride), **NIN** (ninhydrin), **NIR** (near infrared), **ORO** (oil red O), **PD** (physical developer), **RO/DI** (reverse osmosis/deionization), **VMD** (vacuum metal deposition, conventional Au/Zn)

Fingermark quality grading – In a pilot study aiming at evaluating the current practice in fingermark quality grading, Fritz *et al.* (437) showed that independent assessors provided reliable and consistent grading scores. Their study was built on 80 IND/Zn-processed fingermarks, independently evaluated by 11 individuals (differing in their profiles: working institution, geographic location, and knowledge/experience in fingermark grading). The participants were asked to use an absolute ranging scale going from 0 to 4, based on friction ridge detail and contrast. Illustrative pictures were provided for each score. The inter-consistency (between individuals) as well as the intra-consistency (for a same individual; assessed by inserting 20 duplicate pictures in the set of pictures to be graded) were evaluated. Twofold conclusions: (i) 67% of the associated scores were equal to calculated median grade, and 32% within one grade (in other words: 99% of the scores were within one grade), and (ii) 78% of intra-consistency in the grading (meaning that the participants gave a same score for two duplicate pictures), the remaining 22% presenting a difference of one grade. Finally, giving the limited size of the pool of participants, it is difficult to emit conclusions regarding the impact of the participants' experience. This study is supposed to be followed by a larger-scale one.

Thermal development – Two studies aimed at evaluating the performance of the Thermal Fingerprint Developer (TFD-2; Foster+Freeman, UK) (443, 444). In the first study, Fritz *et al.* (443) considered fresh (24-36H) natural and sebum-rich marks, various (semi-)porous substrates among which thermal papers, five detection techniques (pDMAB, IND/Zn, NIN,

ORO, PD) (443). In the second study, Mostowtt *et al.* (444) considered depletive series of fresh and old (>12 weeks) eccrine-rich and sebum-rich fingermarks, various porous substrates, and three detection techniques (*i.e.*, IND/Zn, NIN, PD). The impact of TFD-2 on the subsequent use of detection techniques was considered (*e.g.*, “[TFD-2 →] IND/Zn → NIN → PD”). The conclusions were the following:

- Both studies agreed on the fact that TFD-2 is outperformed by the conventional detection techniques;
- In Fritz’s study, the sequence “IND/Zn → ORO → PD” was compared to “TFD-2 → ORO → PD”. The first sequence outperformed the second, mainly from the performance of IND/Zn. However, ORO gave better results when preceded by TFD-2 instead of IND/Zn. This is explained by the detrimental effect that IND/Zn solvents may have on the lipid fraction targeted by ORO;
- In Mostowtt’s study, TFD-2 had an overall detrimental effect when applied at the beginning of any sequence, especially when considering amino acid reagents. Some positive aspects of TFD-2 were somewhat observed when considering the “TFD-2 → PD” sequence (compared to PD alone);
- TFD-2 can be detrimental to the processed items (especially thermal papers) if the optimized settings were not correctly defined, and is not recommended for wetted substrates (443);
- As a conclusion, TFD-2 should be limited to specific situations (no laboratory facilities or high volume crimes).

3 Miscellaneous marks

3.1 Ear, earprints and earmarks

There is an active community dealing with external ear biometry (472-474), but without strong ties with forensic science and dealing with marks that can be left on scenes. Purkait published a review (475) and researched into the uniqueness of the external ear based on a corpus of 1404 adult male and 1257 female subjects from Central India (476). He also extended the use of ears to familial studies (477). A smaller study on 100 male subjects is due to Verma (478).

Earmark is used as evidence in some jurisdictions (*e.g.* Germany (479), France, Switzerland (480)) but is not getting a lot research attention. During this review period, we note the work by Azadi (481) who implemented a scale invariant feature transform (SIFT) matching technique to compare earmarks to earprints with very low error rates. Also using the FEARID database consisting of 7364 prints of 1229 donors, Morales and colleagues (482) showed the merits of combining local and global features in the matching process. They reported error rates according to the quality according to the quality of images compared. For mark to print comparisons for examples, reported equal error rates were 0.03% (good quality), 3% (medium quality) and 35% (low quality).

3.2 Footprints

Footprints are often a neglected piece of information that can help progress an investigation (483). We report here on some research that came to our attention during the reviewing period.

Podotrack allows easy collection, storage and manipulation of footprint images and can be used to carry out Reel (484) measurements using in forensic podiatry. A Podotrack and an inkless shoe print system were also compared to investigate how often “ghost” images (images giving the appearance that there are “extensions” to some toe pulps) could be

produced (485, 486). Burrow showed that the time of day for the collection of prints does not impact the prints obtained (487).

Nataraja Moorthy *et al.* (488) reported footprint features observed on the prints left by 400 adult Malay participants consisting of 200 males and 200 females. They report on the relative frequencies of local features such as the toes, humps in the toe line, phalange marks, flatfoot condition, pits and cracks.

Kanchan *et al.* (489) reported on the possibility to predict gender based on measurements taken from footprints. Moorthy and colleagues (490) showed how stature is correlated to the dimensions of footprints.

The impact of load bearing activities and walking speed on the size of footprints have been reported by Wall-Scheffler *et al.* (491) in the context of the investigation of human footprint fossils. On a sample of 15 male and 15 female individuals carrying a 20kg pack on their back, they showed that sex, speed and load have effects on the dimensions of footprints.

Kagan (492) has discussed the complexity posed by a forensic examination of where the marks had been left years before the availability of a person of interest. The author is calling for more research investigating the effects of aging on forensic podiatry examinations.

Geometric morphometric methods were applied to study variation of footprint shape in a sample of 83 female individuals, aged between 19 and 36 years (493).

Early results applying image processing techniques and biometric methods on footprint have been reported (494). That includes the possibility of comparing reference footprints against footwear marks left by the person (495).

3.3 Lip prints (cheilosopic patterns)

The study of 60 students (30 males and 30 females) by Kumar *et al.* (496) led them to conclude to the uniqueness of lip prints. Verma (497) or Prabhu *et al.* (498) stated similarly strong conclusion based on the study of 100 individuals. Given the size of the sample, some caution must be exercised. As rightly stated by Dineshshankar *et al.* (499) "The uniqueness of lip print needed to be conformed and accepted." Population studies are reported from Lybia (500), India (501-503) and Egypt (504).

Lip prints have shown ability for gender prediction (474, 505-510). The correlation between lip prints and blood groups has not been established (511-514).

The first studies involving automatic image comparison of lip prints are due to Worbel and his group (515-517). It paves the way towards a systematic understanding of the reproducibility and variability of such prints. Based on a corpus of 120 lip prints, they reported an equal error rate (EER) of 21% (515). In passing, we cannot resist mentioning the very efficient recognition systems developed for cattle identification based on muzzle print images (518, 519).

3.4 Other marks: knuckle patterns, scars, vein patterns

Apart from facial, gait, garment or gender information, CCTV images allow also visualising marks of forensic interest: scars, tattoos, vein patterns or knuckle patterns). Both major and (secondary) minor knuckle patterns can be used in conjunction. Early data suggests that they are these patterns are stable over time and can be used even under unconstrained conditions (520, 521). The modality is still in research stage but a steady increase in accuracy has been achieved (522). Dorsal hand veins patterns (that can also be visible on

images of forensic interest) received also research attention, but mainly in constrained conditions with specific acquisition techniques taking advantage IR cameras (523). At this stage, it is difficult to envisage an application under unconstrained conditions based on query images acquired under forensic conditions.

The use of scars (or other features such as nevi) received renewed interest in forensic science with the proliferation of images showing limited identifying features such as in cases of pedo-pornographic material where only hand on individual may be seen. Assessing these features based only on expert judgment only has shown to be difficult (524) and researchers are striving to acquire systematic data to allow assigning an appropriate weight to these comparisons (525-527).

4 Crime scenes and case reports

We noted in particular the following case reports:

- The report of the development of a mark with cyanoacrylate fuming (CA) on the trigger of a pistol – in this case a Mauser Werke 90 DA (9 mm Parabellum) (528). It is notoriously difficult to develop marks on manipulated firearms.
- The use of a very partial fingermark in association with the print of a person of interest as corroborative evidence, even if an identification couldn't be decided in this case (529).
- The identification of a cadaver through his/her papillary ridges, protected by a latex glove (530). A method to help relax clenched digits from cadavers (531).
- Girelli (532) presented cases of laterally reversed marks and discussed how a thorough ACE-V process could assist in detecting them. Cases of forged identity document using the same fingerprint image are also reported (533). Some of these images can be obtained directly from the Internet and adapted with minimal image processes such as lateral reversal (534).
- The use of skin texture mark from the back of a hand is reported from the UK (535).
- Hays reports on a case where palmar flexion creases have been used to conclude to an identification (536).
- "How long a mark may persist?" or "How fresh a mark is?" are typical questions that ought to be answered with caution. Bunter (222) showed persistence of fingermark over 2.5 years.
- Stones are known to be notorious difficult surfaces to obtain fingermark from. Successes in casework have been obtained with ninhydrin and black powder (441).

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Identification Sciences

DNA and biological evidence, 2013-2016

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1. Introduction

The review focuses on some of the most important developments that occurred during the years 2013-2016 in forensic biology. The selected topics include Rapid DNA analyses, analysis of complex DNA profiles and the development of Next-Generation Sequencing and its application to DNA Phenotyping.

2. Rapid Analysis of STR Markers

The forensic DNA typing laboratories has a core set of markers that are used to generate STR profiles. Currently, the traditional process to generate STR profiles lasts 8-10 hours. This process includes DNA extraction, quantitation, multiplex Polymerase Chain Reaction (PCR) and Electrophoresis Capillary (EC). Multiplex PCR is commonly identified as the bottleneck in the process. The time requirement of up to three hours to complete 28-30 cycles of multiplex PCR for STR genotyping is the largest amount of time required for a single step within the process. Over the last few years, significant improvements have been made to enable the rapid generation of STR profiles to include rapid PCR, direct PCR and development of integrated microfluidic devices.

The goal in developing rapid PCR protocols have been to decrease the time required for amplification to less than one hour ¹. To reach this goal, a first modification is mandatory: the use of alternative DNA polymerases with higher processivities than the commonly used AmpliTaq Gold. The second modification required is to reduce amplification thermal cycling parameters from a three-step protocol to a two-step thermal cycling protocol. Fregeau et al. have conducted the validation of a 26 min amplification protocol using the AmFISTR Identifier primer set, a two-step protocol and the SpeedSTAR HS DNA polymerase ².

In last years direct PCR STR kit formulations have allowed Forensics laboratories to avoid the extraction and quantitation stages in the analytical workflow for reference samples. Direct PCR kit such as Powerplex 18D, Globalfiler Express and Investigator STR Go! have been commercially developed to reach this goal ³⁻⁵. An internal validation of GlobalFiler Express has recently shown that both blood and buccal samples from FTA and non-FTA substrates can produce complete STR profiles with a 40 min PCR ⁵.

Integration of all the stages of the Forensic DNA workflow is a challenging task, but has shown to be possible in recent years. Several biotech companies have developed a device which incorporate all the forensic workflow and utilizes a swab as sample input.

The ParaDNA Screening from LGC Forensics (Middlesex, UK) is a device which is a presumptive DNA test ⁶. This device realizes direct PCR with fluorescent HyBeacon melt analysis of two STR loci (THO1 and D16S539) along with amelogenin and produces a DNA detection score based on total change in fluorescence. This score represents the ability of the samples to produce a STR profile. The ParaDNA device produces results in the form of a DNA detection score within 75 min, thus possibly allowing for improved submission policies to forensic DNA laboratory. The ParaDNA device could be also utilized as a training tool for crime scene officers to check their quality of swabbing before they obtain their qualification.

The ANDE (Accelerated Nuclear DNA Equipment) device, developed by NetBio Inc (Whaltman, MA), provides forensic scientists with a fully integrated device to generate full STR profiles using the PowerPlex 16 chemistry within 84 min ⁷. This device utilizes injection molded biochips and lyophilized reagents, with the capability to run five samples simultaneously on a disposable cartridge. The ANDE device contains an automated allele calling expert system and RFID sample tracking. Evaluation of the success of the CODIS core loci was at 85 % for the 100 buccal swabs tested ⁷. This success rate seems to be low compare to success rate generally observed in reference laboratory closer to 95 %. Despite this low success rate, the ANDE device was the first system to earn the NDIS approval from the FBI in April 2016.

IntegenX Inc. (Pleasanton, CA) developed the RapidHIT 200 integrated device, which utilizes the Powerplex 16 HS chemistry and produces profiles in less than 90 minutes ⁸⁻¹⁰. The RapidHIT 200 allows for 5-7 buccal swabs to be run simultaneously. Success rates was around 95 % which is in accordance with DNA database laboratory success rate. Additionally, an upgraded RapidHIT 200 device is now available with the Globalfiler Express chemistry and the ability to run 7 samples simultaneously ¹¹.

3. Analysis of complex DNA profiles – mixtures and low template DNA

Low-level complex DNA mixtures are often encountered in casework. The statistical analysis of such mixtures is challenging because several genotypic combinations are possible in DNA mixtures. Therefore, it is not straightforward to determine which genotypes contribute to the mixture. This is further complicated when samples have a low DNA content, which makes them prone to PCR-stochastic effects, such as drop-out, drop-in, high stutter rate and imbalanced heterozygotes.

The recommended approach to make statistical evaluation of complex mixtures is to calculate Likelihood ratio (LR). The different models used for LR calculation are typically classified into three groups: i) Binary models; ii) Semi-continuous models; iii) Continuous models ¹². This classification reflects the way peak heights are used. Binary models ignore peak height information completely, and are therefore not suited to interpret low-template mixtures. For this reason this kind of models may be considered obsolete. In semi continuous models peak heights may be used to set the model parameters, while continuous models incorporate peak heights fully. The complexity of LR with semi continuous and continuous models requires the use of specialized software to analyze complex DNA profiles. A number of new software have recently become available ¹³ which are used in conjunction with new high sensitivity STR kits have opened a new era: the era of probabilistic genotyping. Acceptance of such software in the user community, and subsequent acceptance by the court, relies heavily upon their validation.

A key goal of the continuous approach is to describe a DNA profile by modeling all sources of variation and to encapsulate evidence into a single LR. Indeed, it may be tempting to use new statistical methods as a convenient way to generate answers simply by feeding a program with numbers, running the program and reporting the result, but this does not

circumvent a requirement for careful consideration of all of the DNA and non DNA evidence in a case ¹⁴. Numbers of contributors and associated defense and prosecution hypotheses may not be obvious and are subject to debate. There is no reason for numbers of contributors to be the same under alternative hypotheses. In the “exploratory approach” advocated by Haned et al., the biological basis of profiles are evaluated prior to any strength of evidence test ¹⁵.

4. Next-generation sequencing: applications in forensic genetics

4.1 Introduction to Next-Generation Sequencing

DNA sequencing has come a long way since the development of Sanger chain termination method in 1977¹⁶. Sanger sequencing allowed the ability to sequence DNA in both a reliable and reproducible ways, therefore enabling gigantic advances in molecular genetics. Such an innovative method has been the key to successfully complete innovative projects, such as the Human Genome Project, an international research effort (coordinated by the National Institutes of Health) to determine the sequence of the human genome and identify the genes that it contains¹⁷. The Human Genome Project formally began in 1990 and was completed in 2003, two years ahead of its original schedule. However, the many disadvantages of the conventional Sanger sequencing technologies, including low throughput and very high cost have limited its uses to relatively simple genomic applications ¹⁸. Over the past 10 years, the development of high-throughput methods for DNA sequencing, known as Next-Generation sequencing technologies have overcome these issues, becoming a more affordable option for various fields of biology, including disease diagnosis, ancient DNA analyses or forensics.

Next-Generation sequencing technologies refers to DNA sequencing methods which allow multiple parallel sequencing of billions of DNA molecules in one reaction. Two main strategies can be used : shotgun sequencing, which consists in the sequencing of short fragmented DNA sequences (between 50 and 500 bp) without any prior selection of targets and targeted sequencing which involves an initial enrichment step which amplifies the selected regions by PCR or the use of probes to capture the fragments. The latter method is preferred for most applications when the analysis focuses on a specific panel of genes or mutations, including forensics. Using either methods, the fragments are used to generate a library which consist of DNA sequences ligated to generic adapters which will be useful for downstream applications. Additional nucleotide sequences, known as barcode can be added for identification purpose. A normalization step can be added, in order to homogenate the libraries and pool them in equal amounts prior to clonal amplification. Clonal amplification consists of the hybridization of individual DNA molecules to a primer on a solid surface in an isolated reaction. The physical separation of the DNA strands is guaranteed by emulsion PCR or bridge PCR. Billions of DNA molecules are then sequenced in real-time by several methods, depending on the platform used. These methods include sequencing by synthesis (Illumina), sequencing by ligation (Thermo Fisher Scientific), pyrosequencing (Roche 454) or semi-conductor sequencing (Thermo Fisher Scientific). However, clonal amplification is not the only option when performing NGS as several companies. Several companies, including Heliscope Biosciences or Pacific Biosciences, adjust NGS technology to enable single-molecule sequencing, allowing the detection of the sequence of a single DNA molecule instead of a cluster of clonally amplified DNA^{19–21}. The major advantages using single-molecule sequencing is that the original DNA molecules is directly analyzed, therefore eliminating biases generated by capture and clonal amplification steps. Nonetheless, these single-molecule sequencing methods are a lot more expensive to use than clonal amplification methods and the error rate is also much higher. Single-molecule sequencing is not currently used in forensics, but could be implemented within the next few years and could be especially useful for DNA mixture interpretation.

Since 2005, several companies have enlarged the genomic market with different platforms²¹. The biggest advantages of all those solutions is that they provide vast quantities of data, however the associated error rates are higher (between 0.1 to 15%) and the read lengths are generally shorter (30-700bp for short-read approaches) than those of traditional Sanger sequencing platform. However, if NGS has been widely used the clinical field or in academic research for the past 10 years, it has only reached the forensic field for a couple of months. The potential of NGS in forensics is huge, and at the moment forensic labs are still trying to figure out how NGS could be implemented and how it could complement pre-existing genotyping methods, such as Taqman PCR or SNaPshot analysis which have been used in many labs for years. The current capillary-electrophoresis workflow has been optimized for criminal justice over 20 years but unquestionably, NGS display a lot of advantages in Forensic Genomics which relieve many of the limitations imposed by CE. The following part will present the advantages and disadvantages of NGS applied to the forensic field.

4.2 Development of the first commercial NGS kits for forensic genetics

The period 2013-2016 has been the first step for NGS development and validation in the forensic field. Thermo Fisher Scientific launched its first STR assay Precision ID GlobalFiler™ NGS STR Panel containing the same 21 autosomal STRs, along with a Y-indel and two Amelogenin sex markers, found in the GlobalFiler™ PCR Amplification Kit. Thermo Fisher Scientific also launched two SNP typing assays designed for the Ion PGM System : the HID-Ion AmpliSeq Identity Panel for human identification (124 autosomal SNP and 34 Y-STR) and the HID-Ion AmpliSeq Ancestry Panel for ancestry estimation²²⁻²⁴. In the beginning of 2015, the company Illumina launched the ForenSeq DNA Signature Prep Kit containing 27 autosomal STRs, 8 X-STRs, 25 Y-STRs, 95 autosomal human identification SNPs, 56 autosomal ancestry SNP and 24 SNP associated with Externally Visible Characteristics (EVCs)²⁵⁻²⁷. ForenSeq kit is usable on the MiSeq FGx, a desktop sequencer which also has a “research” interphase which is useful for research and development projects. Promega is the latest company to launch NGS kits for NGS with 3 different kits : PowerSeq™ Auto (including 23 STR loci and Amelogenin), the PowerSeq™ Mito System (control region of the mitochondrial genome) and the PowerSeq™ Auto/Mito/Y which combines both sets of amplicons in one multiplex plus 23 Y-STR loci^{28,29}. All the kits exhibit different goals. Thermo Fisher Scientific chose to sell separate kits depending on which type of markers are wanted, whereas Illumina and Promega chose to build all-in-one multiplex. Each forensic labs will have to determine what their needs are and whether or not a global method or a more specific one is preferred.

4.3 Forensic applications to NGS technologies

4.3.1 A wider range of genetic markers

To avoid random match between unrelated individuals, ENFSI and ISFG EDNAP and ENFSI have closely worked together in 2006 to develop scientific recommendations leading to the extension of the European Standard Set of loci by five more STR markers³⁰. Unfortunately, technical limitations of fluorescent-based CE sequencers restrains the maximum number of STR which can be analyzed simultaneously. The addition of new STR markers led to the development of new genetic analyzer platforms with an additional channel for fluorescent detection (6-channel detection systems) where additional STR markers for can be allocated for a simultaneous detection^{31,3}. As more and more countries have their own national DNA database, it is expected that the current number of analyzed loci may increase in the future, and fluorescent-based CE technology may reach its technical threshold. Therefore, NGS technologies appear to be the only option as they offer the analysis of a wider range of genetic markers coupled to an increased resolution at each marker. Other type of significant genetic variants such as single-nucleotide polymorphisms (SNPs) and insertion-deletion (INDELS) can be analyzed³². They are representative of genetic differences both within and

among the different population. SNPs are variation in a single nucleotide that occurs at a specific position in the genome whereas INDELS consist of insertion or deletion of more than one nucleotide. These differences have appeared during evolution creating polymorphic genes, which alleles can be linked to a population or to a particular trait.

Recent NGS kits developed for forensic already have a combination of numerous markers which are useful for various applications. Whereas current CE-based STR kits allow the analysis of less than 30 STR markers, the ForenSeq Library Preparation kit (Illumina) and the AmpliSeq Identity Panel for human identification (ThermoFisher) can be used to analyze up to 235 and 158 markers respectively ^{24,26}.

NGS analysis could also be applied to Y-STR. In general, Y-STR are used to decipher the male component of DNA mixtures whenever a major female profile is present or to analyze paternal relationships between male individuals. Y-STR multiplex kits that include a number of rapidly mutating Y-STRs were introduced and provide substantially stronger discriminatory power than previous Y-STR panels ³³. Meanwhile the maximum number of Y-STR in a CE-based kit is quite limiting, including more and more rapidly mutating Y-STR in NGS kit according to the flexibility of NGS technology could help to enhance discriminatory power, even with close relatives.

Besides the fact that increasing the number of loci leads to more data and can help to reach more conclusive and confident results, the main advantage is the fact that different classes of polymorphism can be analyzed together. Therefore in the future, forensic labs may not have to validate and maintain multiple PCR-based systems. A simple NGS kit will streamline testing by simultaneously analyzing large numbers of globally relevant STR markers and dense SNP sets in a single test.

4.3.2 Increased polymorphism detection

In contrast to fragment length analysis by PCR and CE, sequencing allows to get the full STR sequencing which can contain minor variants within the STR known as Single Nucleotide Polymorphism (SNP)³⁴. These mutations consist of one nucleotide substitution, which means that they can be detected by CE as they won't modify the DNA fragment length. Very recently, previously unknown STR alleles and more overall variability has been found by NGS ³⁵⁻³⁸. Instead of STR composed of pure repeats, some complex STR consist of various sub-repeats. A recent paper has highlighted these new polymorphisms within a data set composed of 183 samples from mixed origin. Within this data set, six loci (D12S391, D2S1338, D21S11, D8S1179, vWA, and D3S1358) showed greater than double the number of alleles obtained by sequence compared to the number of alleles obtained by length³⁵. The highest number to date of mutations found within one STR concerns D12S391 with 53 alleles obtained by sequence compared to 17 alleles obtained by size ³⁵.

Additionally, sequence variations can also be found in the flanking regions of STRs ³⁹. The discovery of internal variants within STR raises the question of STR nomenclature. In 2016, the DNA Commission of the International Society for Forensic Genetics (ISFG) published some recommendations. Indeed, a complex arrangement of repeat motifs in a STR leads to exponential increase of the level of genetic polymorphism and "old" nomenclature is not sufficient anymore. One advice from ISFG consist of a new nomenclature (for example a sequence string format) which could cross data from NGS and from PCR CE, the main advantage being the maintenance of compatibility among established and future data ⁴⁰. Another interesting point using NGS in forensics concerns mixture interpretation. In a recent study, about 30% of the homozygous genotype calls by PCR-CE were found out to be in fact heterozygous after sequencing. Therefore, sequencing of STR with many alleles of the same size can simplify mixture interpretation allowing the right identification of stutter artifact of the major contributor just by sequence analysis ³⁸. It is worth knowing that sequences

from the minor contributor in 1:100 and 1:50 mixtures were detectable by NGS ²⁴ whereas minor contributor less than 1:10 are usually not detectable using traditional methods.

4.3.3 Higher sensitivity

One of the disadvantages of using STR is the fact that combining the size of forensic repeat regions, primer binding regions, and flanking sequence requires amplicons of at least 200 bp therefore limiting the ability to genotype STRs in severely degraded samples ⁴¹. Single nucleotide polymorphisms (SNPs) offer promise for challenging samples as the region being genotyped is only one base allowing for amplicon sizes as small as 50 bases. Several international labs have shown that analysis of SNP genetic profiles and not STR genetic profiles lead to much better results, especially by dropping the allele drop-in rate which are known to be stochastic phenomena related to STR typing of low-template DNA ^{41,42}. As a further proof of the advantage of non-traditional markers for forensic analysis of degraded DNA samples, HID-Ion AmpliSeq Identity Panel assay is able to provide discrimination similar to that of a full STR profile using fragments <150 bp, a level at which an STR profile would show significant degradation ⁴¹. NGS has also been used on very complex such as carbonized corpse whose complete autosomal short tandem repeat (STR) profile could not lead to direct identification ⁴³.

4.3.4 Deeper analysis of mitochondrial DNA

In the past 4 years, next-generation sequencing has also been applied to mitochondrial DNA. It is a time saving and cost-efficient method to analyze the complete mitochondrial genome (mtGenome) compared to Sanger sequencing. In routine forensic applications, the greatest part of analyses have been restricted to the non-coding control region and especially two hypervariable (HV) regions, HV1 and HV2 for legal and technical reasons. Unfortunately, these HV region represent a very small part of the whole mitochondrial genome, therefore when it comes to mitochondrial DNA, only about 4 percent of data is captured. Due to its technical capabilities, NGS allows the entire genome to be analyzed and compared in a case, providing a considerable advancement over the existing capabilities. Whole mitochondrial (mt) genome analysis enables a considerable increase in analysis throughput, and improves the discriminatory power to the maximum possible phylogenetic resolution in many criminal cases, such as the analysis of human skeletal remains, potentially leading to the identification of missing persons ⁴⁴⁻⁴⁶. Moreover, human mtDNA heteroplasmy which is defined as a mixture of more than one mtDNA genome sequence within a cell or between cells of a single individual, can be problematic in mitochondrial DNA analysis in forensics ⁴⁵. The detection of heteroplasmy at the whole mitochondrial genome level supports the advantages of using NGS which can achieve high accuracy and sensitivity, high throughput at lower costs ⁴⁷. Several studies have shown that a mixing ratio of two DNA sources as low as 1:500 can be detected, and maybe even less in the near future ^{45,48}. To conclude, because the developed multiplex PCR system amplifies small-sized amplicons (<250 bp), NGS analysis using the library preparation method described here allows mtDNA analysis using highly degraded DNA samples.

4.4 Ancestry, Appearance and Age: DNA Phenotyping using NGS

One of the major limitations of this comparative approach of DNA identification is that if there is no reference sample in the database to compare the profile to, then the STR profile is not of any value in terms of identifying a person. In the absence of any other information that provides leads for tracing unknown forensic sample donors, investigations can reach a dead end for extended periods of time, before the evidence DNA profile matches with a known individual subsequently added to the growing forensic DNA database or delivered as suspect by police. Therefore, the DNA profile can possibly never match the profile of a known person if this person is never entered in the database.

These limitations of comparative DNA profiling inspired the development of a new method within forensic genetics, the Forensic DNA Phenotyping.

Ancestry prediction has already been used by several labs with traditional methods (CE-based or SNP analysis using SNaPshot) ⁴⁹⁻⁵⁶ but is sometimes not sufficient enough to orientate the criminal case to a restrain number of individuals.

Forensic DNA Phenotyping (FDP) refers to the prediction of externally visible characteristics (EVCs) of unknown sample donors, or unknown deceased persons, directly from biological materials recovered at the crime scene ⁵⁷. FDP outcomes may potentially provide even more accurate informations than human eyewitnesses do. When describing an individual to a criminal sketch artist, victims or witnesses are often shocked by the events and can have memory flaws which can sometimes be contradictory. As such, FDP is expected to assist in the identification of unknown perpetrators, who are not identifiable via conventional comparative DNA profiling by orientating the case towards a more specific type of individual, therefore designing a priority list of suspects that meet the general prediction. FDP is also expected to be useful for missing person identification, in cases where reference DNA profile from putative ante-mortem samples, or from putative relatives are unavailable, and for mass disaster victim identification (natural cataclysms, accidents or deliberate acts such as terrorism). This application of DNA marks a change in the forensic use of genetic material rather than that of current DNA profiling presented in the courtroom.

FDP studies started in the early 2000s and first progressed very slowly. The main reason for the relatively late introduction of FDP is the limited knowledge about the genetics of most human EVCs ⁵⁸. Another reason is that some EVCs are indeed correlated to genetic factors but also to environmental factors which makes the prediction less accurate. For instance, a characteristic that beholds a lot of interest is the height, but it appears that this EVC is related to 424 gene regions and that more or less 9500 nucleotide variations (SNP) only explain 29% of phenotypic variance ⁵⁹. Indeed an important part of human height relies on environmental factors such as dietary deficiencies in childhood, genetic diseases or hormonal defects such as growth hormone deficiency.

From a genetic point of view, the variety of phenotypes is mostly caused by multiple polymorphisms in genes. In the literature, EVCs are mostly identified through Single Nucleotide Polymorphism (SNP), DNA sequence variation in which a single nucleotide (A, C, T or G) in the genome differs between two different individuals. They are the most abundant form of genetic variations in the human genome (around 90% of all genetic variation between humans).

SNPs are stable and uniformly distributed along the genome. Most of them are silent and have no impact on gene function or phenotype; however some SNP however can have functional consequences and may contribute to phenotypic changes and therefore diversity between population groups.

There are a lot of advantages to use them in forensic science. First, their size is very small in comparison with STRs; therefore the polymorphism can be analyzed from a small DNA fragment amplified by PCR (50 to 150bp) and can be especially used on samples containing degraded DNA ⁴¹. Secondly, SNPs have a really low mutation rate making them stable in time (10^{-9} mutation per base and per generation for the SNPs against 10^{-3} for the STR in human nuclear DNA). A great example of their stability can be observed when analyzing the world map of human migrations while studying SNP in mitochondrial DNA among populations. Those mutations stay within groups of population (known as haplogroups). Haplogroups are used to represent the major branch points on the mitochondrial

phylogenetic tree. Thirdly, the majority of the SNPs are bi-allelic polymorphisms as they are due to unique mutational events and without recurrence since their mutation rate is low. Lastly, there are freely online databases of SNPs which contain precious informations such as the location of the SNPs, the DNA sequence framing the SNP of interest, the probable genotypes and allelic frequencies in different populations. Since there are thousands of SNPs, it is challenging to find the ones which could be statistically associated with one or several physical traits.

Among the several analytical methods which can link a genotype to a phenotype, Genome Wide Association Studies (GWAS) are mostly used in the literature. GWAS consists of the analysis of many common genetic variants in several individuals in order to check whether or not any variant is statistically associated with a trait. GWAS has revealed a few genes and SNPs, among them which are associated to pigmentation^{27,60}. There are two types of melanin in the epidermal cells of mammals, the eumelanin (pigment of brown-black color) and the pheomelanin (pigment of yellow-red color). In human, skin color is mainly determined by the number, the size, the type and the method of distribution of melanosomes. The differences of skin pigmentation in human are not based on the number of present melanocytes but in the type of melanin and level of activity of melanocytes.

Some SNPs can impact the determination of eye, hair and skin color. As presented on figure 4, the ASIP (Agouti Signaling Protein) gene is involved in melanogenesis regulation, OCA2 (Oculocutaneous Albinism II) is required for the transport of tyrosine, the precursor to melanin synthesis, within the melanocyte and TYRP1 (Tyrosinase-Related Protein 1) encodes a melanosomal enzyme that belongs to the tyrosinase family and plays an important role in the melanin biosynthetic pathway. To date, around 59 SNPs have been linked to pigmentation and can potentially help predict pigmentation⁶¹⁻⁶³.

Male pattern baldness is a type of androgenetic alopecia that affects susceptible individuals and progresses with age to form a key human externally visible trait. GWAS has significantly associated baldness with AR (Androgen Receptor) gene and EDA2R (Ectodysplasin A2 Receptor). Mutations in EDA2R give rise to a clinical syndrome characterized by loss of hair, sweat glands, and teeth. The AR gene provides instructions for making a protein called an androgen receptor which allows the body to respond appropriately to dihydrotestosterone and other androgens. Studies suggest that variations in the AR gene lead to increased activity of androgen receptors in hair follicles^{64,65}.

Another interesting EVC studied is the presence of freckles which are mostly related to BNC2 (Basonuclin 2) gene and IRF4 (Interferon Regulatory Factor 4) gene. BNC2 gene influences saturation of skin color and is responsible for freckling. IRF4 is associated with the combined trait of sunlight sensitivity, brown hair, blue eyes and freckles^{66,67}.

Other physical traits have also been studied during the past 4 years including scalp hair shape and facial hair (beard thickness, eyebrow and monobrow thickness), with the identification of 10 novel associations⁶⁸. Even huge progresses have been made on very complex traits, such as face morphology. Indeed, in 2016, 5 new genes have been associated with nose shape and chin protrusion⁶⁹.

The last four years also help to progress in the understanding of age prediction^{70,70,71}. The analysis of DNA methylation markers seems to be giving more consistent results compared to quantification of T-cell specific DNA rearrangements or telomere length markers⁷². The expected window of prediction could be reduced to plus or minus two years with an increasing number of markers.

At the time, only the ForenSeq Library preparation kit from Illumina proposes DNA Phenotyping analysis using a NGS workflow, based on the 24 markers developed in the previous Hirisplex multiplex from Manfred Kayser^{27,60,73}. However, it is expected that companies developing solutions for NGS technology will start to develop specific DNA Phenotyping kits which could help to predict a vast majority of physical traits which would overcome traditional identikit based on witness testimonials. However, it is worth keeping in mind that DNA phenotyping is not legal in the great majority of European countries (or at least, the constitution or legislation does not specifically deal with this issue). It also raises several ethical questions including confidentiality and risk of racial stigma^{57,74}.

4.5 Limitations of NGS for forensic DNA Analysis

As we discussed, Next-Generation Sequencing is a very promising tool for forensic labs. However, some points need to be address in order to give access to this technology to labs that are not familiar with the technical part and especially the way to handle a huge amount of data.

First of all, there is a real need that the forensic community decide how to deal with sequence variants. ISFG proposition for a new STR nomenclature which would maintain compatibility among established and future data is a first step that need to be discussed further⁴⁰. There is also the need of a community consensus on the types of markers and regions that can be interrogated. Indeed, as we discussed, several countries forbid the use of ancestral or phenotypic markers⁷⁴. Even if it is not directly related to science, the question of the cost is also important, as NGS analysis are considered more complex and more expensive that CE-based method. However, it looks like a NGS workflow would be completely compatible with a traditional CE workflow and could be only used, at the time, for complex DNA sample (degraded, mixture, low DNA amount...). It is however expected that reagent costs will drop in the near future. Until the beginning of 2008, the cost of sequencing a genome was closely in-line with Moore's law but the sudden and profound out-pacing of Moore's Law beginning in January 2008 help reaching the symbolic bar of \$1000 for a whole genome sequencing⁷⁵. Forensic DNA analysis based on NGS should then adapt as well, enabling more and more forensic labs to implement this technology. As every new technology that can revolutionize a field, the feedback from the community, from Forensic DNA academic research labs to governmental lab with high-throughput DNA analyses, will be important to draw the future of NGS in the DNA forensic field.

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Identification Sciences

Questioned documents,

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INTRODUCTION

This paper is an exhaustive review of the latest technical advances and latest developments concerning documents examination, including handwriting comparison, since the 17th INTERPOL Forensic Science Symposium in 2013. This review is based on articles mostly published in the major forensic or generalist science journals, but also on presentations at international forensic meetings during the period 2013-2016.

The aim of this work is to identify all the relevant work in the field of document over a 3 years period, going from the second half of 2013 to 2016. This is to improve existing technologies, but also to implement new development in the forensic laboratories working on documents. This work can also help determine future axes of research that would try to answer real cases problematics. Although every effort has been made to cover all developments about document examination in this review, some omissions might occur.

It is important to notify that in this paper, two different kinds of publications are referenced: forensic and generalist publications. For this work, only forensic publications are commented upon, the others are included as background information only.

The different areas of analysis concerning questioned documents being numerous, it was decided to group the results of bibliographic research around key topics: handwriting comparison, ink composition, inkjet and laser print analysis, crossing lines, analysis of paper, indented impression, altered documents and security documents.

SOURCES OF REFERENCES

References presented in this work come either from the scientific literature (forensic or not), or publications from various international meetings. Posters are not included. Different journals viewed as Forensic Science International or Spectrochimica Acta and the various meetings concerning documents such as American Academy of Forensic Sciences (AAFS) or American Society of Questioned Document Examiners (ASQDE) are listed below.

Specialised references in forensic :

- Forensic Science International (FSI)

- Science and justice
- Egyptian Journal of Forensic Sciences
- Forensic Science International : Genetics
- Australian Journal of Forensic Sciences
- Research Journal of Forensic Sciences
- Arab Journal of Forensic Sciences and Forensic Medicine
- Problemy Kryminalistyki
- Austin Journal of Forensic Science and Criminology
- International Journal of Law and Forensic Sciences
- Malaysian Journal of Forensic Sciences

Specialised references about the forensic analysis of document:

- Journal of the American society of questioned document examiners (ASQDE)
- American Academy of Forensic Sciences (AAFS)

Scientific references:

- Spectrochimica Acta Part B: Atomic Spectroscopy
- Analytica Chimica Acta
- Applied Radiation and Isotopes
- Microchemichal Journal
- Pattern Recognition Letters
- Journal of luminescence
- Journal of Chromatography A
- Journal of cultural heritage
- Chemometrics and intelligent laboratory systems
- Dental materials
- Resources, conservation and recycling
- Progress in organic coatings
- Journal of Applied Research and Technology
- Vibrational Spectroscopy
- European Association for Signal Processing (EURASIP) Journal on Advances in Signal Processing
- International Journal of Computer Applications
- International Organisation of Scientific Research (IOSR) Journal of Applied Chemistry
- Analyst
- Universitatea Politehnica Bucuresti (UPB) Scientific. Bulletin, Series B
- Nanoscale
- International Journal Of Advanced Studies in Computer Science and Engineering
- http://www.fosterfreeman.com/download_application_notes/ECCO-printer_toners.pdf
- Asian Research Publishing Network, Journal of Engineering and Applied Sciences
- Journal of Applied Physics
- Der Pharma Chemica
- Journal of analytical chemistry
- Color Research & Application
- Journal of the Brazilian Chemical Society
- International Journal of Advanced Research in Computer and Communication Engineering
- Denim - Manufacture, Finishing and Applications
- Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy
- Talanta
- Procedia Computer Science

- Springer Science+Business Media New York
- Anal. Methods
- International Journal for Pharmaceutical Research Scholars
- Journal of American Science
- World Journal of Pharmaceutical Research
- Microchemical Journal
- Applied Soft Computing
- International Journal of Electronics, Communication and Soft Computing Science & Engineering (IJECSSE)

Meeting, symposium, forums:

- 24th Danube Adria Association for Automation & Manufacturing (DAAAM) International Symposium on Intelligent Manufacturing and automation – Procedia Engineering
- 10th European Network of Forensic Handwriting Experts (ENFHEX) conference and business meeting
- Non Impact Printing (NIP)-Digital fabrication and digital printing
- Systems Conference (SysCon)
- 2nd International Workshop on Automated Forensic Handwriting Analysis (AFHA)
- Proceeding of the 2015 ACM Symposium on Document Engineering
- 14th International Conference on Frontiers in Handwriting Recognition (ICFHR)
- European Document Experts Working Group (EDEWG)-8th Conference
- 13th International Conference on Document Analysis and Recognition (ICDAR)
- 11th International Association of Pattern Recognition (IAPR) International Workshop on Document Analysis Systems
- Intelligence and Security Informatics Conference (ISIC)
- Association of Forensic Document Examiners (AFDE) Symposium
- American Society of Questioned Document Examiners (ASQDE) annual meetings
- American Academy of Forensic Sciences (AAFS) annual conferences

STATE OF THE ART OF THE EQUIPMENT

To begin with this study, it is crucial to identify the whole array of equipments, old and new, that are used, or have been used in documents forensic analysis. Some techniques such as Thin Layer Chromatography or High Performance Thin Layer Chromatography have been in use since the beginning of ink analysis.

Some of them were improved and new technologies were developed. They can be more efficient in the study of ink composition (dye or other compounds such as volatile solvents used in dating), ink discrimination or in the study of paper.

Although non-destructive techniques such as the Raman spectroscopy are still the preferred ones, a growing number of almost non-destructive techniques are being used (damages to the document are kept at a minimum).

Depending on the object and/or matter of the analysis (ink, paper...), a large choice of equipment is available. The analytical techniques available to us are listed below:

- Raman spectroscopy
- Laser-induced breakdown spectroscopy (LIBS)
- Laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS)
- Scanning electron microscopy-Energy dispersive X-ray spectroscopy (SEM-EDS)
- X-ray fluorescence (XRF)
- UV-Visible spectroscopy

- Micellar electrophoretic capillary chromatography with UV/Vis diode array detection (MECC-DAD)
- Laser desorption ionization mass spectrometry (LDI-MS)
- Thermal desorption gas chromatography mass spectrometry (TD-GC/MS)
- Fourier transformed infrared spectroscopy attenuated total reflectance (ATR-FTIR)
- Pyrolysis gas chromatography mass spectrometry (Py-GC/MS)
- Capillary Electrophoresis
- Direct analysis in real time mass spectrometry
- Thin Layer chromatography (TLC)
- High performance thin layer chromatography (HPTLC)
- Nanospray mass spectrometry
- X-ray microtomography
- Ultra performance liquid chromatography (UPLC)
- Paper spray mass spectrometry
- Liquid chromatography diode array detection Orbitrap mass spectrometry (LC-DAD-Orbitrap)
- Capillary electrophoresis electrospray ionisation time of flight mass spectrometry (CE-ESI-TOF-MS)
- Nuclear magnetic resonance spectroscopy (NMR)
- Surface enhanced resonance Raman scattering (SERS)
- Fourier transformed infrared spectroscopy
- High performance liquid chromatography (HPLC)
- High performance liquid chromatography mass spectrometry (HPLC-MS)
- Isotopic Ratio Mass Spectrometry (IRMS)
- Direct analysis probe nanoextraction coupled to nanospray ionization mass spectrometry (DAPNe-NSI-MS)

In addition to these techniques, one may add all those routinely used by FDEs such as the Video Spectral Comparator which does not need to be presented.

Overview about ink and paper analysis, including datation and crossing lines

Due to the large number of analytical methods available to Forensic Document Examiners (FDEs), we may find it difficult sometimes to determine what kind of material/method to use in order to resolve a given problem. This state of the art article may allow for some perspective on what is done nowadays and serve as a basis for future research.

M. Calcerrada and al [1] set out a literature review (years 2000-2014) about the forensic analysis of documents, including the dating of inks, pen ink analysis, printer ink analysis, paper analysis and other topics such as crossing lines. In this study we will discuss the results of all the analytical techniques that are currently in use, and present advantages and disadvantages of each.

BALLPOINT, GEL, FOUNTAIN, MARKERS, PEN INKS AND STAMP INK

Because of their wide use and accessibility, writing inks remain a popular topic for FDEs. Forty one references are presented here, stressing the interest about this subject: The references from 2 to 41 are sorted by the analytical technique used for ink analysis.

1. HPTLC-TLC [2-9]

This method is one of the oldest available. Thanks to the simplicity of its implementation, it's still used today in the analysis of many types of inks. *D. L. Feraru and al* [2] confirmed the interest of TLC when used in combination with several analytical techniques to discriminate pen inks. According to *N. Sharma and al* [3] [5] and *H. Bhardwaj and al* [8], TLC is used to separate the components of the ink which are then identified by other analytical means. This

technique can also be used independently for the discrimination of pen inks [4] [6] [7] [9]. Old databases exist but the user must adapt to using new plates which can raise problems of repeatability and reproducibility that endanger the relevance of the original database

2. LIBS [10-11]

LIBS is a method requiring no sample preparation and which is quick to implement, hence its increasing use.

A. Kula and al [10] evaluated the discriminating power of LIBS on blue, black and red inks. They showed that the discrimination power obtained is important for blue and black inks. For them, it is sometimes possible to discriminate inks originating from the same manufacturer. The discrimination power of this technique, which is based on the variation of the wavelength of the incident laser, was also studied by *N. Elsherbiny and al* [11].

3. Spectroscopy: UV-Visible, IR-FTIR, Raman and Luminescence [2] [11-22]

Since the video spectral comparator is a very common equipment in our laboratories, it is used in several studies.

V. Da Silva and al [12] [18] identified blue ink ballpoint pen manufacturers through statistical processing (PLS-DA) data obtained by VSC.

S. Alzaabi and al [13] studied the discriminating power of VSC6000HS in reflectance, infrared and visible lights, as well as luminescence on black and blue inks from ballpoint pen. They got 92% of DP for blue inks and 94% of DP for black inks. B.

Usman [16] created a database of different brands of ballpoint pens (black and blue inks) using both UV-Vis and IR spectroscopy.

G. Mark [17] raised the problem of "inkless" pens that deposit a thin layer of metal that can fluoresce under VSC.

Raman signature of inks from several brands of blue ink pens was studied by *A. Braz and al* [19]. Results showed that, regarding oil-based inks, the most commonly used pigment was the Crystal Violet, making their discrimination difficult. In contrast, since there's a greater diversity of dyes in the composition of gel and liquid inks, discrimination is easier. *M. Asri and al* [21] realised similar studies but used FTIR spectroscopy and chemo-metric tools. *M. Halim and al* [22], used FTIR spectroscopy too, but with ATR, a non destructive technology.

4. LDI-MS [23]

X.F. Wang and al [23] applied the LDI-MS to the analysis of inks from stamps. This technique allows the identification of the present dyes and their classification. Finally, this allows a rather powerful discrimination of inks.

5. Capillary Electrophoresis [24-26]

M. Krol and al [24] described the use of both micellar electrophoresis capillary chromatography and capillary zone electrophoresis coupled with mass spectrometry for the analysis of stamp inks. This allows the discrimination of green, violet and blue stamp inks. The contribution of mass spectrometry is essential for the characterisation of dye molecules. Discrimination of black pen inks is achieved in the work of *V. Bohme and al* [25] by using capillary electrophoresis. The various components of ink were separated by capillary

electrophoresis, and electropherograms were processed by statistical tools so as to obtain groups based on the origin of pens.

6. IRMS [27]

The isotopic ratio of nitrogen, carbon, hydrogen and oxygen is measured by IRMS in order to study the isotopic variation of these elements in blue ballpoint or gel pen inks. According to *A. Lesley and al* [27], this would help to distinguish between several inks with very similar chemical compositions.

7. HPLC or UPLC and LC-DAD-Orbitrap MS [19], [28-30]

Gas chromatography is a destructive technique often used for ink analysis (sometimes, coupling it to a pyrolyzer may be useful). HPLC, as for it, is a widely used technique in analytical laboratories that can replace the HPTLC. Its major advantage is its coupling with a fairly easy MS.

X. Ying-Jian and al [28] studied the red dyes present on the one hundred Yuan Chinese banknote by HPLC /MS, in order to identify them. Obtaining the “chemical signature” of these dyes, would permit to differentiate the genuine banknotes with the right molecules from the false ones.

L. Chuen Lee and al [29] confirmed the use of UPLC to discriminate black inks from ballpoint pens. The chromatograms were analysed using statistical tools, PCA and ANOVA, in order to create different groups according to the origin of the inks. The discriminating power obtained for 12 pens is 100%. In another study, *L. Chuen Lee and al* [19] also used UPLC on blue inks from ballpoint pens but by analysing at the same time by UV-Vis spectroscopy. Again, it proves that UPLC offers the strongest discriminating power.

The analysis of acidic and basic dyes from blue pen inks applying the LC-DAD-Orbitrap technique is realised in the publication of *Q. Sun and al* [30]. In this work, ten types of roller pens and twenty ballpoint pens were studied. The discrimination then becomes possible from the identification of present dyes.

8. Paper spray mass spectrometry (PS-MS) [31]

The Paper Spray method is a new way to introduce samples into a mass spectrometer in a simple manner. This technique combines a substrate made of chromatography paper with electrospray ionisation to enable the direct analysis of dried fluids like inks.

This was applied by *P. Da Silva Ferreira et al* [31] to the study of ballpoint pen inks. No sample preparation is required and discrimination of inks is effective.

9. Miscellaneous [32-41]

This section deals with publications on various topics based on a common object ie the analysis of dyes.

In order to identify pen manufacturers, dyes have been analysed by surface assisted laser desorption/MS in the work of *S. H. Bahng and al* [32] and by various chromatographic methods in *D.L.Feraru and al* [33].

Other publications relate to the analysis of old documents inks [34], [37-38], or to the authentication of tax stamps on bottles of alcohol by LIBS and statistical tools [36].

M.A. Taha and al [35] raised the issue of erasable ink pen by friction, in order to highlight the features left by this type of pen.

C.R. Appoloni and F.L. Melquiades [40] used the portable XRF technology to perform an elemental analysis on various colored regions of banknotes.

Finally, *V. Huynh and al* [41] tested a method of nanoextraction coupled with nanospray ionisation on iron gall ink and modern ink. This allows local analysis of the present inks on a document, with the ability to analyse ultra traces. This analysis generates almost non-existent damages, which could be useful for FDEs.

INKJET AND TONER PRINTING

Technological development and low prices are making these 2 printing techniques more accessible to the public, especially for laser printers (an increasing number of cases related to this technique are treated at the IRCGN).

Analytical methods can vary according to whether the inks contain dyes or pigments.

1. TONER [42-47]

Despite the presence of more and more questioned documents printed by laser printers, only a few publications deal with this subject.

However, *K. Herlaar* [42] showed that toners can be discriminated by their magnetism. Discrimination is important between single-component toner containing iron and bi-component toner with no iron. The question has risen whether quantitative measurements of magnetism can also be used to discriminate between printers which use magnetic single-component toner.

78 samples from twenty-three different printers using magnetic toner were analyzed with a Regula Magmouse. Algorithms were used, based on a Bayesian Likelihood Ratio framework, to discriminate the printing, depending on the printer used.

In another context, *T. Rottes* [43] deals with the insertion of toner mentions on an already printed document. The author studied the effect on toner of a second passage in a laser printer by microscopic observations and LIBS analysis. The use of LIBS for toner analysis was also presented by the company Foster and Freeman [44], which showed that 5 toners from 5 different printers could be 90% discriminated using this technique.

The interface between the paper and the toner can be studied too. *M. Myllys and al* [45] studied this by positron RX and laser ablation, in order to obtain the depth profile. In this study, positron RX was used to produce a 3D structure of the paper and laser ablation, combined with optical means, revealed itself useful when building the 3D structure of the layer of toner. All this was done to investigate the factors influencing the penetration of toner into the paper.

To increase the security of documents, *M. Ataefard and al* [46] developed a fluorescent toner by eco-friendly emulsion aggregation. This toner possesses characteristics comparable to an industrial one.

Although toners are composed of pigments, *K. Saini and al* [47] carried out analysis by TLC and HPTLC. Chloroform was used to extract the components of inks, and two elutions with two different mobile phases were performed. The results show that it was possible to differentiate between two toners by color and RF of some spots revealed on the plate.

2. INKJET [48-54]

The share of documents printed by inkjet in cases of threatening / anonymous letters or in counterfeit documents is very important. Therefore it is essential for FDEs to study this topic.

Raman spectroscopy is a non-destructive method often found in inks analysis. This technique, which requires no sample preparation, can be used to create databases. *F. Herry* [49] used the Raman in this purpose. This database, composed by around 700 samples of inks allows the identification of cartridge reference used to produce an inkjet printing, thanks to the profiling of Raman spectra of colors found in a questioned document.

Sometimes, Raman spectroscopy can be coupled with another destructive method in order to accurate the discrimination of inks. It is developed by *M. Krol and al* [48] who analysed several cartridges by Raman spectroscopy. In parallel, the application of capillary electrophoresis allows more accurate group identification, by the separation and comparison of the various components of inks

Statistical analyses may also be performed on the Raman spectra obtained, as showed in the work of *P. Buzzini and al* [50] who performed a PCA within each colour, to assess inter-variability. Spectra are then classified based on the model and the reference of the printer.

B. McKechnie and al [51] studied reflection spectra of cyan, yellow and magenta inks to determine whether the standard terms of security documents have been printed by one or more printers, with the aim of identifying possible forgeries. Inks discrimination can also be done by Raman, by reducing the interferences due to fluorescent inks, thanks to the SERS method [52].

Regarding some cases related to inkjet printer inks, some destructive methods can be used for discrimination if it is deemed useful and relevant by the community. *R. Sharma and al* [53] realised the extraction of primary colours from inkjet printing on a questioned document, and analysed them by FTIR to identify functional groups of dye molecules.

To conclude, *A. Kula and al* [54] applied the CE-ESI-TOF-MS to determine the chemical composition variations of inks between different brands. The mass spectra of certain additive molecules are characteristic of some manufacturers, which may allow a group identification of inks.

3. ANALYSIS OF BOTH TONER AND INKJET [55-60]

To finish with this paragraph, several publications relate to the analysis of both inkjet and toner.

One of the major information contained in this section is that laser ablation techniques such as LIBS and LA-ICP-MS are used more and more, demonstrating their efficiency for the analysis of printing inks. Not only the results obtained are very useful for the discrimination of inks that come from different cartridges, but damages on the questioned document are kept at a minimum.

T. Trejos and al [55] used the coupling between a LIBS or a LA-ICP-MS with a SEM for the discrimination of black toner and inkjet. 27 samples from different manufacturers and different batches are analysed. Results show that using the SEM-EDS technique on its own gives little discrimination, but when coupled with the laser ablation, it becomes possible to achieve 89% of discrimination with LIBS and 100% with LA-ICP-MS. These two methods are also used by *K. Subedi and al* [56] for discrimination of inkjet and toner inks, but also for

discrimination of offset and intaglio inks. The combination of the two methods for the analysis of one type of ink would increase the discriminating power.

A multi-technique approach has been chosen by *R. J. Williamson and al* [58] for the characterisation of inkjet and toner inks. Thanks to FTIR analysis, 75 samples of toner were divided into 12 groups, based on the polymer used for their fixation. In contrast, paper makes the analysis of inkjet ink impossible. With the help of Py/GC/MS and DART/MS, components of the two types of ink are characterised. To conclude, these three complementary methods can help us discriminate inks coming from different sources and to make connections.

As in their previous study [55], *T. Tréjos and al* [59] realised a new multi-technique approach to the development of a database of inkjet, toner offset and intaglio inks. Six analytical techniques were used. The obtained results were then presented according to their best discriminating power (resulting from the coupling of some techniques). They concluded that the coupling of LA-ICP-MS and FTIR seemed to be the most relevant approach for all types of inks.

INK AGING / DATING

This topic remains a challenge for FDEs despite the various studies that have been conducted for many years [61-75]. A consensus between the different forensic laboratories seems to have been reached, and the methods begin to be standardised. However, ink dating remains controversial, especially regarding the reliability and repeatability of results. In addition, the document storage conditions remain a hugely influencing factor in the methods currently in use.

Several analytical approaches may be employed, such as solvent evaporation or dye degradation.

B. Li [61] used the method of dissolution-diffusion over different black inks of gel pens. A dissolving-diffusion agent is spilled on the studied ink, and photography is taken under the microscope every 10 seconds. The grey levels of the obtained pictures are then measured in Photoshop, and a grey level curve representing a function of time is plotted.

A. Koenig and al [62] [70] proposed a chemical method by focusing on a solvent present in pen inks, the phenoxyethanol (PE). Four parameters were studied by GC/MS : the amount of PE, the relative intensity of PE peak, the solvent loss ratio and the solvent loss ratio calculated with relative peak area. Three parameters that may affect the aging of ink were also taken into account: the writing pressure, the composition of ink and the storage conditions. In conclusion, the measure of solvent loss ratio with relative peak area seemed to be the better way. The drying process of solvent can also be an approach for ink dating as shown by *A.A. Cantu* [63].

As presented in the introduction of this section, several factors influence the aging of the inks. Daylight is one of the most important. For instance, crystal violet pigment (CV), common in pen inks, deteriorates under light in methyl violet and tetra methyl para rosaniline (TPR). *D. S. Islek and al* [64] have studied the evolution of the degradation of CV in two residues under UV lighting for several hours. They showed that the amount of CV decreased significantly after a 13 hours exposure. This approach may be useful to artificially age the ink of a pen that's been used on a questioned document, in order to obtain a specific dating curve. However, there is the issue of the storage conditions that are often unknown and will influence the result. *B.H. Gungor and al* [65] were also interested in the deterioration of the CV under illumination by determining the amount of the different molecules by HPLC.

Other authors were not interested in solvents or dyes for ink dating, but in resins and binders present in various inks. *A. Hahn and al* [67] [69] tried to date ballpoint pen ink by analysing resin and binder by HPLC-ESI-HRMS. A slow degradation of resins can be observed, and some can still be detected after 10 years. However, other resins show faster decomposition and after a few years they are no longer detectable.

The FDE deals not only with ink analysis, but with the aging of paper too. *M. Zhaoyang* [68] used biotechnology to date paper. The determination of the age of paper is realised by sugar analysis and by determining the degree of polymerisation of paper. An analysis of bacterial colonies on paper is also performed. In terms of resins, some degrade slowly (possible detection after 10 years) while other ones degrade quickly (few years).

B. Li and al [71] and *A. Koenig and al* [72] have addressed the major problem regarding the accuracy of results achieved through datation, ie storage conditions of the document until analysis.

For *B. Li and al*, calibration curves were obtained by determination of the volatile compounds on documents stored in different conditions (under light or in the dark). The results show that the GC method can be effective for the relative dating of inks under certain conditions. *A. Koenig and al* highlighted the major influence of storage conditions. They studied the implementation of a method of ink dating by thermal desorption, and analysis by GC/MS in a new laboratory to verify the reproducibility of the results. A probabilistic approach seems preferable.

PAPER ANALYSIS

Compared to the previous review, only 7 articles have been published in different journals since the last review. [57] [76-80]

Isotopic ratio mass spectrometry (IRMS) is one of the methods used by several scientists for paper discrimination. *K. Jones and al* [76] used this technique to measure the isotopic ratio of carbon 13. In this publication, they studied the influence of the source (raw material), the manufacturing process and the effects of printing on the isotopic ratio of carbon 13. A light carbon isotopic fractionation was observed during the pulping step and during the first stages of bleaching. On printed documents, it appears that laser toner printing has a greater impact on this ratio than the other printing techniques. To conclude, if the paper is to be discriminated by IRMS, we must be careful to analyse non-inked areas, and to confirm the absence of toner particles by microscopy. One advantage of this technique is that it can be applied on paper, even after it has been treated with indandione in the case of fingerprint revelation.

In a second study, *K. Jones and al* [77] have pushed further the study of the influences of printing, fingerprint and DNA record. This experiment was then renewed on isotopes of oxygen. Results obtained were compared with those obtained by conventional methods of paper comparison (basis weight, paper density...).

LIBS which is already used for technical analysis of inks, can also be applied to the analysis of paper. *A. Metzinger and al* [57] showed that elemental analysis using LIBS provided an effective, practical and robust technique for the discrimination of document paper and inks with minimum damage to the document's substrate.

Putting chemical analysis aside, *W. Mazella and al* [78] explained that it is possible to discriminate sheets of paper by their size and colour. In their study, two hypotheses are

considered. The first one is that during the production process, paper sizes vary within a single ream and this can be helpful when working on a case of page substitution. The second one is that paper colour varies between different types of paper, allowing discrimination. The first results show the possibility of differentiating the sheet from several manufacturers according to size and colour. There is also a possibility to observe a predictable variation in the width and height between different sheets. This method is fast, simple to implement and non-destructive.

To finish with this section, page substitution is generally treated by non-destructive approaches by looking for inconsistencies between different sheets of paper. For this, one can observe paper fluorescence, printing processes present on different sheets, layout, stapling, any present watermarks... Sometimes, the analysis can be destructive just as paper micrography which aim is the identification of the fibers constituting paper. In addition to these methods, a new approach which involves the analysis of mineral fillers present in the paper, is being developed. *J. Green* [79] suggested the use of X-ray fluorescence to metering the amount of minerals in the paper.

Let's end this chapter with *W.F. Rowe and al* [80], who presented the application of Fourier Transform in two dimensions (FFT2D) for the discrimination of paper sheet. Several non-destructive methods for the analysis of paper (thickness, weight, colour...) exist. However, paper making process involves repetitive patterns (mesh) on the paper surface. Applied to these patterns, FFT2D produces an image made up of dots, representing the periodicity of mesh and their direction. 16 sheets of paper from different reams were analyzed using this method. The FFT2D is achieved through Image J with paper images taken with a 14M pixels camera using transmitted light. The figures obtained are reproducible and each leaf has its own image.

DETERMINATION OF WRITING OR PRINTING SEQUENCE

Just like the dating of inks, determining printing or writing sequences remains a major challenge for document examiners [81-89]. Some spectroscopic (Raman, Infrared) or optical (microscopy, 3D profilometry) techniques are used by experts in questioned documents.

Before presenting the use of advanced analytical techniques in other fields of document analysis such as inks or paper, *K. Y. Lee and al* [81] presented a novel approach to determine the order of printing sequence between a wet stamp and printing text using adhesive tape. To do this, authors took pictures of the crossing area before and after affixing tape. Then they observed the pixels that changed colour in order to deduce which ink was removed with the tape. Experiment results showed that this method successfully discriminates the sequence of seal impression and printed text for different types of ink cartridges and seal inkpads, under various storage conditions, enabling forensic investigation in cases of forgery.

Other publications deal with stamps ink. *T-Y. Kang and al* [82] used the atomic force microscope to study crossing between wet stamp and toner printing. This technique is based on the fact that the toner particles can be moved by the microscope probe to see what is present below. This method has the advantage of causing minimal damage and proved to be conclusive on the cases studied by the authors.

Putting aside chemical techniques, *N. Ozbek and al* [83] had the striking idea to cut the crossover region with a scalpel and to observe the slice under a microscope. Thus one is able to observe and delineate the different layers of inks, and therefore to deduce the order of apposition. This technique has 2 major disadvantages: it's destructive, and you need a suitable microscope to apply it.

The applicability of Raman imaging for non-destructive and rapid analysis of blue crossing ink lines was investigated by *A. Braz and al* [84]. Using Raman spectroscopy, each ink was analysed on its own, and then both were analysed at their crossing. The predominant Raman signature at the crossing location was used to infer the order of application. Different types of pens have been tested on different types of papers. On the 90 Raman spectra analysed, correct order of apposition could be determined in 60% of the cases. The remaining cases could be attributed to a non-homogeneous distribution of inks. This non-destructive method is promising and could probably be improved by the use of statistical tools.

L. Vanco and al [85] examined surface enhanced Raman maps of blue writing inks. Inks were deposited on a rigid silver-nanocrystalline diamond-silicone heterostructure which constitutes an active substrate providing a possibility to write directly on the surface and to record Raman maps. By this mean, authors could successfully distinguish the order in which the cross lines of inks were deposited on the substrate.

U. Konarowska and al [86] and *L.A. Mancebo* [87] presented the observations made when a document already containing pen ink is then printed by a laser printer. Results are based on microscopic observations and the study of specific characteristics.

Sometimes the order of deposition between pen and toner can be determined without having line crossing. Experts in questioned documents have to observe whether the toner particles are present in pen-made writings and to determine whether these particles are present above or below the ink pen. Several techniques can be used for this. *F. Delavalle* [88] used the observation at high magnification (x1000) under visible light, NIR and UV. NIR and UV were used especially in the case when the toner and the pen ink were black.

To finish with this subject, *S. Wang* [89] used tomography microscope to determine the writing or printing sequence between fountain pens and laser printing. Several factors, such as the types of ink and laser printer, the writer and the class of fountain pen, were analysed to determine whether they might affect the formation of characteristics. The results showed that there were many differences among the samples resulting from different sequences. This method can be applied to determine the sequence of laser printing and fountain pen writing.

DOCUMENT SECURITY

An increase in publications related to document security has been noted in the review period, with 35 references. These can be conveniently classified under the following categories. Some references had already been cited in other sections like ink analysis by chemical method or by forensic tools.

1. Fraud and forensic intelligence: [90-93]

The general principle of forensic intelligence is described by *M. Morelato and al* [90]. When a false document is detected and is made with sufficient quality, it may be interesting to operate in this field. The modus operandi of the forger can thus be traced. A set of characteristic signs will be drawn from the false document and will thus define a particular profile (printing techniques, security elements, errors in the MRZ band...). Then profiles are integrated into a database and compared with each other.

S. Baechler and al [91] also considered the forensic intelligence and made a study about visual characteristic on false documents.

M. Auberson and al [92] proposed a method based on computer vision to analyse and compare images of false identity documents. This part deals with the problematic of conditions for the acquisition of pictures and the analysis of relevant regions for the purposes of forensic intelligence.

K. Saini and al [93] explained the detection of forgeries by common photo editing software on different documents.

F. C. Shiver [94] presented a real case of falsification of a quitclaim deed. He showed how the simple technique of “cut and paste” can be used on a document that is not the original one. In this case, the questioned signature could only be revealed as a copy of an upstream signature, thanks to the presence of the original document.

Still starting from with real casework, *H. M. Hoover* [95] presented cases dealing with forgeries of securities present on US secured polymer documents (laser engraving, embossing, hologram). The development and the availability of techniques such as inkjet, laser and thermal printing allowed for an ever increasing quality of the imitations.

2. New solutions to increase security levels: [96-101]

This paragraph deals with the latest technologies and the latest research regarding the safety associated with secured documents. *J.A. Zlotnick and al* [96] showed the use of half toned picture as a security. This technique is used to simulate a continuous tone using a limited number of colours, usually by transforming a continuous image by a set of microscopic dots which vary in size, colour, spacing... The frequency and modulation of amplitude of these dots were used in ink-jet, laser toner and offset in order to tighten the security of these images. Security is also added by using inks different from those found in retail in order to prevent forgers from getting a hand on them.

A. Hodgson [97] discussed the applications and hardware currently used for security printings and threats from new digital printing technologies that might supersede conventional printing techniques.

Regarding the development of new techniques, *M. You and al* [98] presented the impression (ink-jet technology) of a high resolution, highly luminescent anti-counterfeiting figure. Nanoparticles were used and they reacted differently depending on the excitation wavelength they were subjected to. This reaction allows the printing of two elements at the same place and their observation, one after the other. Also in the field of printing, *M.J. Meruga and al* [99] explained the development of an ink containing different percentage of poly-aniline to print security documents figures. Thus it is possible to measure the difference in conductivity between the unprinted area and the printed one.

In any other field, *S. Ambadiyil and al* [100] addressed the thorny issue of breeder documents. They are the basis of the request for a security document, and false ones can be used to obtain an undue document. This paper proposes a method to create a unique identifier. The idea is to convert the fingerprint of the person holding the document in a QR code that is then printed on it.

Finally, *T. Wagler and al* [101] presented the moiré technique used to hide information on a security document. Their work focused on incorporating two moirés or more at the same level, aiming at making counterfeiting more difficult.

3. Document analysis by chemical methods or analysis by traditional forensic tools: [102-120]

If possible, like at the IRCGN which is a multidisciplinary laboratory bringing together experts in document fraud and chemists within the same unit, more advanced chemical and physical tests should be done if the case requires them. *J. Takalo* [102] presented a method based on the study of the paper fibers constituting banknotes to quickly authenticate them. He used a curvelet type algorithm to measure the fiber orientation, and quantify anisotropy. By optimising the parameters of the method, it was possible to distinguish between genuine banknotes from false ones. Also in the field of counterfeit money, *S. Sugarawa* [103] conducted a study on scanner identification based on the study of the scanned holograms present on forged banknotes. Two protocols were studied. The first was based on the distribution of brightness, in order to identify the scan parameters. The second studied correlations in the colour distribution. The identification of a model out of a database is thus made possible. Researchers should now focus on the influence of the printing of the hologram.

More advanced analytical capabilities can be used to study counterfeit banknotes. For example, *E. M. Schmidt* [104] used the ambient EASY spray MS ionisation technique for analysing Brazilian banknotes at the molecular level in order to obtain a specific signature, which constituted the genuine bills. The molecular profiles of counterfeit notes will then be different from the genuine ones, making counterfeit detection possible. This technique is quick and almost non destructive. *X. Ying-Jian and al* [28] used liquid chromatography to obtain a chemical signature, characteristic of the Chinese banknotes. FTIR spectroscopy can also be used for the same purpose as the previous studies. *E. Sonnex and al* [105] used this technique, which is based on the presence or absence of a characteristic peak of counterfeits documents to 1400cm⁻¹, to authenticate English banknotes.

The ever growing use of plastics in the manufacture of titles contributes to an increase in the number of papers dedicated to the topic. *S. A. Kingsbury and al* [106-107] presented a synthesis of plastics used for security documents, and the effective analytical tools for characterizing them. Among them, various non-destructive tests can be used such as digital microscopy or FTIR spectroscopy. Also in the field of IR, hyper-spectral images taken in the near IR were used by *C. S. Silva and al* [108] to detect document fraud. Different types of fraud were tested : the changing and/or the adding of words and the crossing of lines. The image acquisition was within the range of 928-2524nm. Chemometric tools like PCA were used. Other techniques such as atomic force microscopy or Raman spectroscopy can also be used [109].

In addition to these chemical methods, more conventional optical examinations were made on several subjects. For example, *P. Chi-Ming and al* [110] discussed about the various factors affecting the authentication of stamps. *R. Bertrand and al* [111] presented a system to detect document fraud based on the intrinsic characteristics of the document. *Z. Luo and al* [113] used an algorithm to detect a variation in the ink proportion between the original entries and those that were added. Entries were observed through hyper spectral images.

S. Elkasrawi and al [116] presented an automatic identification method of the type of printing used on a document. This method is based on the analysis of the noise specific to each printing technique. This noise can also be studied to identify a brand as thin differences are attributable to the manufacture of the machine, making specific identifications possible. *S. Eskenazi and al* [117] wrote a publication about applications in the field of automatic security document authentication. Some parts of the questioned document were analysed by some algorithms and were compared with those of a genuine document. This was made by using the Delaunay triangulation.

To end this section, some publications are about automatic document control, such as the ones that exist in some airports. *M. Gschwandtner and al* [118] provided a system for controlling the automatic terminal using an optical document simulator. *S. Stolc and al* [119] are concerned with the influence of the degradation of paper on the effectiveness of the controls carried out on the automatic terminals.

HANDWRITING

As in each triennial review, handwriting comparison is widely represented [120-201]. Topics covered here are so vast that it is difficult to deal with all of them in this work. Nevertheless, major trends can be identified and can be classified as follows:

1. Skills and knowledge: [120-140]

A. Devlin and al [120] conducted a visual approach to determine the writing direction in ballpoint pen. They achieved that by observing the distribution of ink on the paper fibres. The type of pen strongly influenced the resulting observations, and those were made more difficult when the ink covered fibres.

In another area, some studies were carried on the importance of training experts in writing comparison to make correct conclusions. *T. N. Dewhurst and al* [121] performed a blind test between Forensic document examiners and uninitiated people. One of the tested parameters was the distribution of rewards to the group of uninitiated people when they gave correct answers. No difference was obtained by this type of motivation on the number of correct answers.

The quality of writing in reports was addressed by *R. N. Morris* [122] and one of his arguments was that all carried examinations have to be presented and explained.

The conclusion in particular must present clearly the logical approach and the elements that helped to achieve it.

The education of the American FDEs, their membership in a professional organisation... were investigated by *V. Springer and al* [123]. This study also focused on the type of laboratory in which experts work and on their experience. *V. B. Dahir and al* [124] discussed the importance of training for the quality of the published findings. Many studies have shown that FDEs are more effective in comparing and correctly identifying or excluding signatures than untrained people. Although FDEs and untrained people sometimes find the same characteristics in some writings, experts always give better results due to their training.

With the current issues of terrorist threats towards Western countries, it will be interesting to see if experts in writing comparison will be able to solve cases in a language that is not their native one. *A. Al-Musa Alkahtani* [127] conducted a study to determine whether handwriting experts with no mastery of the Arabic language could resolve cases in Arabic. The results obtained by these experts are compared with those obtained by Arabic-speaking experts. The findings of this study show that non-Arabic speaking experts sometimes take several months more to make their conclusion than Arabic-speaking experts. However this study is incomplete because only the appearances of signatures were taken into account and only four items were studied. *M. Van der Hammen* [128] presented a work on writings made in the Latin alphabet by different ethnic groups in North Africa. Discussion was held to assess the common characteristics, identified or not.

Al-Hadrahmi and al [129] tried to assess whether national characteristics were discernible in Arabic writing. From each of four Arabic countries Morocco, Tunisia, Jordan and Oman, 150 participants produced handwriting samples which were examined to assess whether it was true or not. The results showed that it is possible to group countries by region (North Africa and Middle East) based on the observed characteristics.

Jinwoo and al [130], as to them, were interested in finding common characteristics between Chinese and Korean characters.

In another area, *Y. Seki* [131] made a database consisting of several characters in different alphabets or writing systems (Japanese, Chinese, Latin and Arabic). Two-hundred and sixty-eight subjects participated in the data collection. A digital pen was used for the data acquisition and x-y coordinates of the pen tip were acquired online. So, this database had x-y coordinate data and handwriting image data. A writer identification from a handwritten text was made using this base. This identification was accomplished using characters that were different but having the same component characters for questioned and known handwriting. Two characters per construction type were selected for the examination. First, anyone sample of character number one was selected as the questioned handwriting and all sample of character number two was defined as the known handwriting. Then, the reverse experiment was done. The results showed that the average correct identification rate was 58%. The comparison of the questioned and known handwriting using only a part of character was not the good way.

The topic of handwritten signatures is at the heart of several studies [132-135].

T-Y. Kang and al [136] presented a blind test that proved that the exchange of opinions between handwriting experts is vital to reduce the risk of error in the conclusion of a report.

2. Environmental influence: [141-149]

The writer's environment can influence greatly the findings of a handwriting examination; that's why this issue was raised by several experts in the field. *K. Fazio* [141] studied the influence of an external force on one's signature. Different types of constraints (without constraint, on a given line width, in a frame...) have been imposed to writers of all ages (18-83 years old). The different signatures were collected using a digital tablet, in order to treat the data with statistical tools (ANOVA). The conclusion of this study is that the imposed constraints altered the obtained signature, which is to be considered during an exam.

M. E. Durina and al [142] researched the different ways one can disguise his or her handwriting and the strategies used.

The results showed that the most used technique involved distorting the lower case letters. Writing entirely in a different style is also an option. The second part of this study focuses on whether a person disguises his or her handwriting in the same way every time. 71% of studied people used the same disguise between two sample of written collected.

In order to study the influence of the educational level on one's writing, *A. T. Szymanski* [143] observed the development of the design features from three members of the same family separated by two years in age. Depending on the education received and the time spent learning writing, their classroom level, it was found that children develop more or less individual characteristics (letter basic construction, alignment, punctuation...).

J. A. Lewis [144] examined the impact of modern writing tools on handwriting, working on the writings of one writer, from 2011 to 2014.

C. Fernandes [145] studied the influence of Alzheimer disease on the drawing of signatures. Differences were found in several features such as quality of the baseline, spacing between word. However, the writing speed and pressure do not seem to be affected by this disease. On the other hand, letter repetitions, omissions and substitutions are observed.

3. Bayesian approach: [150-152]

The use of Bayesian statistics in forensics is increasingly studied, especially in handwriting comparison. In Bayesian statistics, probabilities are interpreted as a degree of belief rather than the limit frequency of a phenomenon. The model parameters are modelled by probability distributions and parameters, and are then refined after taking into account new observations.

F. Taroni and al [150] have created a database by analyzing the contour of the loops of the characters “a” and “d”. The contours were analysed by image processing by means of techniques based on Fourier analysis. The ultimate goal of this study is to differentiate populations of male and female writers, right or left handed people. To do this, the authors use a Bayesian approach.

R. Marquis [151] presented a case illustrated through the use of a Bayesian approach in order to say if a person in particular has produced the signature or not. The probability was 50% at the beginning of the study and by applying a likelihood ratio of 1/70, the final probability that the suspect did not produce the questioned signature was 98.6%. This method allows the use of statistics to strengthen one's conclusion, but it can also be helpful when the results are presented on the basis of numerous data.

4. Digital document: [153-156]

M. Abel [153] and *M. Pertsinakis and al* [154] were both interested by the issue of digital signatures used to authenticate electronic documents. The authors presented the difference between the written and the digital signatures. They studied the latter through software that retrieves data directly from the tablet used to collect signatures.

J. Zimmer [155] and *T. Dziedzic* [156] are also addressing the issue of digital signatures. The first study shows the three digital signature systems used in the Czech Republic in several services (banking, telecommunications...) and how this issue is addressed by experts. The second article describes an experiment conducted to determine if a visual feedback when drawing a signature affects its characteristics. Some signatures are collected directly on a digital tablet, and others are collected after a sheet of paper was placed between the pen and the tablet. The results showed that a higher pressure is exerted on the tablet when the paper is not present.

5. Automated comparison: [157-201]

This last section deals with automatic comparison methods. These methods mostly use a probabilistic approach with different criteria chosen as a function of the conducted studies.

R. Mandal and al [157] presented an identification signature model in different languages (English and Indian dialects) by using an algorithm. *S. Yin Ooi and al* [158] also compared signatures excerpted from images and used several statistics models (DRT, PCA, and PNN) to study them. This makes the differentiation between genuine signatures and imitations possible.

X. Chen [159] created a database comprised of several Chinese signatures. Features like character height, gray level, and alignment among others were identified and extracted automatically. A correlation coefficient was generated to assess the degree of

correspondence in the comparison of two signatures. To validate the results, a MANOVA analysis was performed.

E. Griechisch and al [160] present off-line signature verification methods by analysing images and forms of signatures. The method chosen by the authors verifies the signatures and is based on similarities measured by comparing 2D characteristics of the shape of the signature's skeleton.

To close this section, *A. Parzale and al* [169] suggested a novel definition of stability regions and a new method for detecting them from on-line signatures. The stability regions are defined as the longest similar sequences of strokes between a pair of genuine signatures. Shape and temporal information from genuine signatures were captured by a pad. The results obtained by using simple criteria, in order to stress the importance of stability regions on assessing the authorship of a signature, confirmed that this method captures the subject's behaviour.

INDENTED IMPRESSION

This simple technique, recognized and used by many laboratories, is not at the cutting edge of technology [202-207]. Nevertheless, *N. Hanarine and al* [202] described the factors influencing the revelation of indentations on paper. Several factors were studied (type of pen, paper type, humidity...).

Some articles deal with DNA sampling on the results achieved by electrostatic revelation. *M. Holt and al* [203] studied the influence of the DNA sampling by putting adhesive tape on a document. This method degrades the surface of the paper. This has the effect of producing a noise which interferes with revelation on indentations; at worst, the surface of the paper may be torn. In conclusion, if this technique has to be applied, revealing traces of indentation has to be achieved first, knowing that sometimes, DNA is transferred onto the plastic film [204]. It is the expert's role, in agreement with the investigator, to determine priorities and to decide which type of analysis comes first.

D.T. Plaza and al [205] used the ESDA to reveal all fingerprints on a document, in order to be able to collect the DNA on them with a dry cotton swab. This non-destructive analysis is not performed on paper but on the plastic film itself.

In another field, *K. Butler* [206] has addressed the issue of traces of indentation on crumpled documents. Revelation on these types of documents can never be optimal since the toner cannot stick. To improve this situation, the author recommends to humidify and to flatten the document. Various tests were performed with multiple parameters (with and without weight, humidification only...); better results were obtained using this technique.

QUALITY ASSURANCE

Only a few publications deal with quality assurance. Quality assurance in regard to results is such a given today among laboratories that the issue is less and less addressed in the bibliography. However, it is possible to find articles addressing the theme of interlaboratory or proficiency tests. The latter are an essential aspect of quality assurance. They cover all areas of forensics. *C-K Li and al* [208] organised a proficiency testing programme on Chinese handwriting and signature examination. They assumed that Chinese handwriting is used among a quarter of the global population and that the possibility exists that FDEs might work on cases involving Chinese language. A total of 23 forensic laboratories were registered for the programme and 19 of them returned their examination results. Among the registered labs, 11 came from North America, 5 from Europe, 1 from Africa and the rest from Southeast Asia, Australia and New Zealand. Most of the examiners who did the test did not

know the Chinese language. All the reported results were satisfactory (text and signature). To conclude, this programme underlined the fact that properly trained FDEs, applying sound scientific principles and following a proper methodology can reach the proper conclusion, even if the text is not written in their native language.

C-K Li and al [209] wrote a paper about the work practices and the quality control procedure for Chinese handwriting examination in accordance with ISO/IEC 17025, an international standard on general requirements for the competence of testing and calibration laboratories. With the support of a quality assurance management complying with the ISO standard, FDEs are compelled to address the criteria as stated in the quality assurance plan, and to undertake scrupulous efforts in delivering the scientific work at all stages throughout their examinations. By doing this, personal bias and other subjective factors would be kept to a minimum.

MISCELLANEOUS

We've listed 45 papers in this section that were difficult to classify in the previous categories [210-255].

1. Altered documents: [210-212]

M. Abd-ElAziz Abd-ElZaher [210] studied different inks which have the property to disappear after application. His study presents means to reveal these inks. This can be done by analysis under incident light, by using VSC-6000 and thanks to the use of chemical products such as sodium hydroxide.

Er. Waheeda Dhokley and al [211] showed the use of an algorithm that can reconstruct a document from torn pieces. This technique is called "Image Mosaicing". *L. A. Olson* [212] also presented a methodology to reassemble shredded documents. This was developed during the examination of a torn document found in a bag:

1. Sorting papers by colour and type as well as trademarks,
2. Taking measures of the different pieces,
3. Aligning the pieces in the direction in which they were torn,
4. Creating an assembly model and a grid to align the pieces,
5. Creating a grid of the figure torn on the whole document.

2. Expert in court law: [213-214]

One article deals with an effective way to expose the examinations on a case to a jury [213]. Taking the example of a real case presented to a jury, question and comparison signatures are enlarged 4X and displayed on a board. Question signature was retrieved from an altered document and was then expanded and printed on paper and projected. The use of images is important to present a case and convince a jury because it helps support identified elements. Finally, the presentation must be adapted to the audience.

G. A. McNally [214] explains how to present the analysis of documents to uninitiated people. The expert should speak about real cases to captivate the audience, including the circumstances of the analysis and the resolution of the case.

3. Document analysis : [215-225]

These papers covered wide-ranging topics including the use of radiography to visualise watermarks in documents, the detection of hidden correspondence, the analysis of document securities, the authentication of some documents [215]. The identification of a thermal sublimation printer based on the analysis of a specific machine and on specific cards [220]. The examination of the cards revealed a misalignment in the images. The

comparison of the cards and ribbon under UV light revealed voids resulting from the incomplete transfer of the protective layer. Physical match allowed concluding that the tape had been used to print images on the cards.

4. Intersection between fingermarks and ink : [226]

N. Attard-Montalto and al [226] explored the use of TOF-MS for determining the deposition sequence of fingermarks and ink on a porous paper surface. By mapping selected endogenous components present in natural fingermarks, it enables the observation of friction ridges on a laser printed surface, and this only when a fingerprint is deposited over the layer of ink. Further investigations have shown limited success on ink-jet printing and ballpoint ink.

5. Effect of ninhydrin treatment about document analysis: [227-228]

R.H. Negherbon and al [227] used the VSC to analyse documents after fingerprints examination. Ninhydrin is commonly used to reveal fingerprints, which renders any ink analysis impossible afterwards. The aim of this presentation is to highlight the effects of ninhydrin on ink. It's an update on what is possible to do by VSC analysis when the document is treated. This was inspired by a real case in which mentions were revealed by the VSC under several conditions after a treatment by this chemical compound.

H. Itamiya and al [228] were also interested in the effects of ninhydrin and printing on the examination of paper (change of weight / thickness, composition). Thickness and weight are increased by two-sided printing. The ninhydrin only affects the thickness of the paper substrate.

6. New developments in printing techniques: [229-236]

This part deals with new developments in the fabrication of toner and the influence on the final printing, the application of flexography on plastic, the presentation of ink drying mechanism and its influence on printing quality... These articles are too disparate to warrant comments.

7. Miscellaneous: [237-255]

These articles deal with the application of 3D printing [237-238], review and application of functional data analysis to chemical data [239], the collecting and saving of data in thermal ribbon [243], the method to detect fraud in a casino game with cards [249], script format document authentication scheme based on watermarking techniques [253], currency security and forensics [254].

CHALLENGES

Trends and future challenges concerning document analysis are turned to the conception of databases, these being incremented by information obtained thanks to various analytical tools available to FDE. These databases can be based on the use of a single analytical technique or more, each giving different but complementary information. The ultimate goal is to provide useful and relevant response to investigators such as identifying an ink (trademark, manufacturer...) or a machine. The correlation of data from several laboratories seems to be an efficient way to quickly develop such bases, cause of the lot of work!

Regarding document fraud, the new challenge is in the forensic intelligence. Indeed, the collect of relevant information regarding the making of false documents is the best way to identify a possible counterfeiter's connection. For this, the creation of technical platform combining technical expertise of forensic laboratories and the work of investigators in order to dismantle these connections is an effective way.

Moreover, having within the same organization, document fraud experts and chemists is a real asset. Indeed, chemists are able, thanks to databases mentioned in the first paragraph, to provide additional information about tools used to design false documents. It's a real advantage for forensic intelligence.

Finally, for several years, researches on handwriting comparison have focused on the use of automatic and statistical tools. The fact that these works are still in process is the proof that implementing of such devices is very challenging.

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Forensic Science Management, 2013-2016

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1. Introduction: Are We on the Path Forward?

The National Academy of Science's (NAS) 2009 report *Strengthening Forensic Science in the U.S.: A Path Forward* [1] is a milestone, altering the way in which the industry and outside stakeholders view its existence, including laboratory managers, medical examiners, teachers, members of law enforcement, the legal profession, academic scholars, and the community at large. This report was surprisingly critical in the eyes of many forensic scientists working in the field at the time, describing the forensic science system as having "serious problems," stating "significant improvements are needed," and referring to cases of faulty forensic analysis leading to "wrongful convictions of innocent people."

Understandably, many forensic scientists, and even police and prosecutors, were defensive [2,3], and in some cases rightly so. Houck [4], for example, points out that some commonly used medical tests performed in clinical laboratories have lower accuracy rates than those seen in many forensic science disciplines, yet it seems forensic science takes a disproportionately large amount of criticism.

The ultimate purpose of the 2009 NAS report was not to criticize, but to offer, as in its title, a path forward by systematically outlining challenges and areas for improvement both broadly and discipline specific. Integrated governance, improved data management, developing best practices, oversight, accreditation, increased research, education, and training have all been stressed as essential goals for continuous improvement. Indeed, these formally stated goals indicate an ongoing coming of age for forensic science, goals which every mature and developed industry uses as a way to reflect by collecting and analyze performance data, creating action plans when things go awry, and following up on outcomes in a continuous feedback loop necessary for continuous improvement. To what degree and in what way the forensic community responds remains to be seen.

2. Themes in Literature

Seven years have now passed, with limited assessment on whether or not the 2009 NAS report has inspired improvement in the areas of its recommendations. Unfortunately no guidance on measuring outcomes or what success would look like was provided by the NAS. In addition, assigning a grade, number, or label to the level of success the industry achieved, although a potentially worthy endeavor, would necessarily ignore the complexity and extensiveness of issues faced by forensic science [2]. Operating as a system within systems, the laboratories have operational, political, and procedural relationships with police, attorneys, the courts, and other criminal justice entities. The *17th International Interpol*

Forensic Science Management Symposium in Lyon, France, held in 2013, offered the first formal qualitative assessment of forensic science management broadly, a driving theme behind the changes in the science in forensic science the 2009 NAS report called for. Forensic science is majorly a governmental endeavor, supported by public-provided resources which must be managed effectively and efficiently to meet the desired goals. Therefore, any changes demanded of a science must come through, at least operationally, changes in managerial strategy, policy, and practice. The current review continues where the last review [5] left off and covers how the literature has progressed from late 2013 to the middle of 2016 [6].

The categories and themes for this review have necessarily changed from the last review, reflecting the dynamic nature of the industry and perspectives of those who work within or with a forensic science service provider. Five categories have been chosen to represent the current landscape of issues in the forensic science management literature, being:

- partnerships,
- best practices,
- ethics,
- third party perceptions, and
- the future.

The themes from the last review have not stopped being important and can be found in the literature represented in this review, either explicitly or implicitly. Management and managers should be responsive to changes in their environment and this review reflects that responsiveness. Although not newly important, these updated topical themes, in the eyes of the authors of this current review, appear comparatively more in the literature of the last three years as industry practitioners and scholars adjust their research focus. What has stayed the same is the difficulty in writing a review of this type, given how broad and overlapping the topics are, as well as the review's emphasis that it is not only *what* is produced but *how* it is produced that is important. Therefore, readers interested in any topic here are encouraged to delve deeper into the original literature cited for a more thorough treatment.

3. Partnerships

The words “partnership” and “collaboration” appear often in the literature, covering university, intergovernmental, and law enforcement partnerships; the word “team” occurs even more, reflecting an organizational psychology view of forensic laboratories. This prevalence is evidence that the industry finds partnerships an important issue in both a positive and negative way. It is true that the industry does not, and like most industries cannot, operate in isolation, and therefore has interactions with the nonprofit sector, private sector, and government sector; the use of federal funds by state laboratories to pay for private laboratories to analyze sexual assault kits is an example. That this topic has increased in emphasis in the literature is probably no coincidence; as budgets dwindle and demand for services increase, reaching out to partners to share resources and distribute burdens is a natural remedy.

3.1 Nonprofit Sector Partnerships

Although universities are not the only nonprofits to work with forensic science laboratories—for example, forensic science can be used for wildlife conservation efforts [7]—they are the dominant ones. Universities with resources to support meaningful research, and the academics who work within them, are an integral part of the continued development of the forensic science industry, not only in research, but in curriculum design and university

program accreditation (not to be confused with laboratory accreditation). The specter of too-little research, a point strongly made by the NAS and others, continues to haunt this relationship.

Communication with academics at conferences leads to information sharing, where theory and practice complement one another and lead to new applications that otherwise would not occur [3]. One study gives a more formal example of this, where, by using the theory of constraints (similar to Six Sigma or lean management), key improvement areas in the throughput of a Finnish forensic DNA laboratory were identified and acted upon to increase efficiency and reduce backlogs [8]. This process, which is in the domain of industrial engineering not forensic science, may not be known to laboratory managers without this type of academic communication. Other examples exist elsewhere but do not routinely appear in peer-reviewed literature; if at all, they seem popular with the press at large, perhaps signifying a need to polish the laboratory's reputation and justify improvements made.

Finding and qualifying for many grants or other financial resources, government sponsored or otherwise, may not be possible without a university both as a partner and in many cases as the principle investigator in a given research study. Some research solicitations require a university faculty as the principle investigator. Human decomposition facilities are an example of research that would be difficult outside of a university setting. In 1980, the first such facility in opened at the University of Tennessee, Knoxville, USA and since then only five other universities have followed suit, something unsurprising given the unique difficulties these facilities face [9]. Secured indoor and outdoor spaces, mental health resources for negatively affected observers, and sizeable budgets for research and security are rarely found at operational forensic laboratories. Most if not all forensic science laboratories would find it challenging, if not impossible, to conduct this type of research given their primary mission. The industry benefits by partnering with universities and learning from their research conducted by professors, students, and practitioners.

Curriculum development, program accreditation, and continuing education are other important ways laboratories can partner with universities or other nonprofits. Ultimately, the laboratories will determine the skill sets needed of recent graduates and a feedback loop of what is working and not working in educating these students is necessary to improve university academic programs. In too many cases, students are perceived to be not prepared to work in a laboratory or crime scene investigation unit after graduation [2,10]. Although some training period is needed in most industries, the average training period laboratories demand for new forensic scientists is excessive. Several scholars explore this issue by developing a new curriculum design in forensic DNA education at Fayetteville State University using Collaborative Testing Services' proficiency kits [11]. By tracking outcomes for three consecutive years, this curriculum redesign was shown to reduce training time and substantially decrease cost both budgetary and to society through increased number of submitted DNA profiles. The risk here, of course, is that the wider and higher goals of education become reduced to a vocational check list.

The challenges that laboratories face with new hires are part of the feedback loop that laboratories should use to communicate to universities, either directly or through accreditation. Through working with and potentially for such organizations as the Forensic Science Education Programs Accreditation Commission (FEPAC), the industry should be able to guide university programs to the educational level, quality, and focus desired. Although not all university programs are accredited, they do respond to market pressure. Demand from employers for high quality students translates into demand by students (or parents of students) for accredited programs. Through these market pressures,

accreditation should grow as universities wish to remain competitive. Continuous improvement, similar to laboratory accreditation, is an essential component of academic degree program accreditation and should be embraced by any competitive university who has their students' interests at heart. As a complement to university program accreditation, some argue for required continuing education and licensing requirements to practice forensic science, given its importance and effect on people's lives [3]. These requirements or similar already exist elsewhere, like in the U.K.

3.2 Private Sector Partnerships

The topic of privatization and independence from government control is a controversial one. Most laboratories are publicly owned and only a few are privately funded [3]. However, complete privatization has not been embraced by the industry. The U.K. Home Office has been under harsh criticism for several years now after an uncoordinated closure of the FSS and attempted pseudo-privatization of forensic science services in 2012 [12]. It is clear that whether or not a laboratory is privately or publicly controlled, benefits from partnering with private firms and incorporating traditionally corporate strategies into a laboratory management framework have benefits, some of which were laid out in the last review [5]. In this way, a public laboratory can still capture some benefits experienced by privatization, without the politically and operationally difficult process of privatizing the forensic system.

Incorporating new technology is one way to partner with private firms. As the needs of the industry change, communication with suppliers of forensic science resources and consumables will be a continued priority. Use of radio frequency identification devices (RFID), rather than barcodes, in property and evidence tagging is one recent and successful example [13]. Since RFID tags contain informational capabilities not seen in barcodes (like carrying far more detailed information about the object tagged and touchless screening), laboratories can work with manufacturers of these devices to develop the systems they desire, such as those that monitor temperature, light, movement, and those that record data in specific ways or on specific devices. Shipping technologies or courier systems are another example of technology [14, 15]. These systems, which need not be internally provided, have the potential to reduce backlogs and increase competition if evidence can be shipped in a budget-friendly manner to laboratories that have capacity to handle larger levels of evidence processing. There are some challenges to implementing this technology, including refrigeration and storage; however, shipping regulations and federal, state, and local laws are perhaps the largest barrier. [16] describe shared services by public health laboratories, and provide an example of cost savings and reduced processing time for tuberculosis testing in a sharing system between Rhode Island State Health Laboratories and the New York State Department of Health Wadsworth Center. Although cause for celebration, the legal challenges in such systems, which include the U.S. Constitution's Compact Clause limiting the power of states to form agreements, varying and in some cases contradictory state laws, and transportation of infectious substances [17]. Others [18] provide a third applied technological example, being aerial vehicles, which can be used to diagram scenes of traffic accidents rather than using hand drawn images. Finally, [19] describe potential benefits of partnering, either directly or indirectly, with social networking companies such as Facebook, Twitter, and Google, as well as the experts who work with these technologies to aid in criminal investigations involving digital footprints.

The private sector offers more than technology but also management methodologies and process improvement designs. The private sector must weather market pressures unknown to the public sector and, therefore, must develop fast, cheap, and effective solutions for survival, while the public sector can limp along with "good enough" solutions that satisfy rather than excel [56]. The section found below titled *Best Practices* contains many

examples of these, so to avoid excessive overlap and repetition, the interested reader is directed to that section.

3.3 Government Partnerships

Partnering with government, which comes in many forms, can be a double edged sword. Supportive legislation and grant funding, such as the Coverdell National Forensic Science Improvement Grants, was created to reduce backlogs. Although not exclusively meant to reduce DNA backlogs [20], Langenburg et al. [21] argue that DNA has been prioritized in funding, leaving many of the other disciplines, such as latent print examination, with increased work without a proportional rise in resources or personnel. This sentiment is reflected more broadly by [10] and [2], who see a poorly supported research program and funding framework; that is, no national research strategy for forensic science. Robertson, et al., [10] are encouraged by the internationally recognized need for this increased financial support of the industry but sees no evidence that there will be any progress here in the foreseeable future. The 2015 Department of Justice \$41 million grant initiative to reduce backlogs of untested sexual assault kits signifies some gains, but is still a narrowly focused pet project of Vice President Biden, and does not come close to systematic change in how the industry is funded as a whole [3]. Developing quality control legislation is another way that the industry has the potential to partner with government. Sweden, for example, has seen an overhaul of their national DNA elimination database legislation with the goal to increase DNA analysis' effectiveness and reduce false exclusion [22]. Doleac [23] has shown that DNA databases have significant cost and crime reduction benefits, where national and even international cooperation increases the societal benefit due to economies of scale. Crime knows no borders and forensic science must be nimble enough to reach across those borders, as well.

Partnerships with police are a special type of government partnership, and vary from being contained within a police unit to complete autonomy. Samarji [24] argues that such partnerships are often strained and when a laboratory is controlled by a police unit, an "unholy marriage" is created. The source of this strain comes from the differences between a para-military culture and a scientific culture. A strong research and peer review culture is necessary for an open and adaptable discipline that unbiasedly solves crimes [24, 25], and when a laboratory is not independent from police, this flexibility does not always occur. Bruenisholz et al. [26] describe another angle of partnership with police through standardized data collection, in this case for solving repetitive deliberate fires. The challenge of tracking deliberate fires is partially vocabulary, where a fire department, police department, and forensic science laboratory may have various ways to record an event. The police, for example, may categorize the event of a deliberate fire as vandalism, which may never register as a fire event for a fire department or forensic science laboratory. By coordinating language, better communication and by extension criminal investigation should occur [26].

Finally, partnerships and interactions within the industry among scientists, both domestically and internationally, have the potential to help the industry and reduce the effects of fragmentation [3]. Wright et al. [15] describe the Thai tsunami victim identification operation. This 2004 disaster resulted in the largest disaster victim identification operation in history. Due to the size of the disaster, varying demographics of victims, and limited operational capabilities of local Thai forensic facilities, several outside international DNA laboratories were contracted to help. The ability of outside vendor laboratories to help when the capacity of laboratories nearest a disaster is less than necessary is an important planning issue. King and Wells [27] also look at a small country, Trinidad and Tobago, and analyze the use of forensics ballistics imaging technology to reduce crime. By partnering with and treating Trinidad and Tobago as a case study, King and Wells show that effective communication

networks between investigators and police can link crimes and reduce violence, something that is often lacking in developing nations.

4. Best Practices

It is not just *what* is produced, but *how* it is produced that determines the effectiveness and the total benefit to society of forensic science services [6]. The *how* is improved by implementing best practices, or improving the framework in which they are produced. The 2009 NAS Report stated that “most disciplines still lack any consistent structure for the enforcement of ‘better practices,’ operating standards, and certification and accreditation programs.” Forensic science has made progress in this area, through developments such as ASCLD/LAB-*International* accreditation and ISO/IEC 17025:2005 standards which outline organizational requirements of a forensic science laboratory, as well as the new U.S. National Commission on Forensic Science (NCFS) and its allied Office of Scientific Advisory Committees (OSAC) administrated by the U.S. Department of Justice (DOJ) and the U.S. National Institute of Standards and Technology (NIST). However, the sentiment of many forensic scientists suggests that compared to older and more clearly defined industries, commonly accepted best practices in several areas of forensic science are still in their infancy or developmental stage [2, 3, 28, 9], especially at a national level [1]. Highpoints of important topics related to best practices in this review include data management, accreditation, and management education. These topical headings contain within them issues of efficiency, effectiveness, and continuous improvement among others. Much of the last review was also implicitly dedicated to best practices [6], including a section on leadership and organization and a section on business realities faced by the industry.

4.1 Data Management

Collecting data on significant aspects of a laboratory and its employees is important to not only understand performance within a particular laboratory, but also to benchmark performance to the industry at large, which both allow for critical decision making. There has been great progress over the last several years in collecting business data on forensic science laboratories for a variety of key indicators. Speaker [29] analyzes data from project FORESIGHT, which standardized many definitions and metrics of laboratory assessment measures [30] and provides a univariate and multivariate analysis of certain business metrics of laboratory performance, such as return on investment, productivity, and average total cost of case processing. Similar to FORESIGHT, Brown et al [14] explore an end to end identification process for fingerprint and DNA analysis in Australian burglary cases in order to benchmark current business processes at multiple stages of the investigative process in order to improve national performance. This project follows the United Kingdom’s Scientific Work Improvement Model (SWIM), which recognized the crucial but sometimes forgotten fact that one stage of the investigative process impacts the next. Peltokorpi and coauthors [8] concur, and through an application of the theory of constraints, identify bottlenecks, or resource constraints, in the output of a Finnish forensic DNA laboratory by collecting and analyzing information management system database data and interview data. In the case of the considered Finnish forensic DNA laboratory, the resource constraints came from inefficiency processes in the registration phase of samples, the use of daily quotas, and an autonomous working culture.

Analyzing data goes beyond specific laboratory performance metrics. Wright et al. [15] analyze identification data collected after the 2004 Thai Tsunami, which clearly shows identification rates vary by demographics, specifically nationality and age. For example, children were most likely to be identified with DNA, whereas adults were most likely to be identified with dental records. Additionally a Thai adult was most likely to be identified through fingerprints, whereas an adult victim from Norway who was vacationing in Thailand

at the time was most likely to be identified with dental records. This sort of information could result in improved triage methods, where victims of smaller length may be prioritized in sending DNA samples for analysis. Samarji [24] also explores data that goes beyond laboratory performance, and delves into industry perceptions and culture. By interviewing forensic scientists, sworn police members, and members of the judiciary in Australia on their perceptions of independent versus police managed laboratories [24], a clear coexistence problem between the scientific culture of civilian employees and military culture of police has been demonstrated. Police are significantly less likely to believe that they have significant influence on the practice and identity of forensic science compared to civilian employees, and are also significantly less likely to prefer an independent management structure for a laboratory.

By analyzing data such as that described above, comparisons and benchmarking can be done to understand where a particular observation, such as a laboratory, stands compared to the larger picture. Speaker [29], for example, uses multivariate analysis to explore the quadratic relationship between average cost and output for FORESIGHT laboratories. Due to economies of scale, the relationship seen is nonlinear and U-shaped. Thus a simple univariate numeric comparison is not appropriate as average total cost is dependent upon output. McAndrew and Roth [28] propose an extension of the FORESIGHT project through benchmarking division of labor and its impact on output which may also be a quadratic relationship. Brown and Ross [14] propose similar benchmarking within a national Australian laboratory system. [15] discusses whether events such as the September 11, 2001 World Trade Center attacks or 2002 Bali Bombing, and the identification operations that followed, can be used as benchmarks on performance. Complexities and overlooked realities of each event compared to the 2004 Thai Tsunami identification operation made previous experiences in large identification operations an imperfect if not inappropriate benchmark. Finally Oorschot and colleagues [31] discuss not benchmarking but rather comparisons of data within DNA contamination minimization monitoring programs. By collecting DNA profile data on laboratory employees, police, and others who come into contact with evidence samples or work areas, and comparing these DNA profiles to those in an active case, incorrectly linking cases or false exonerations should be reduced.

By properly collecting performance data, and benchmarking or comparing this data to reference points, managers can make decisions and form arguments for improvement. Langenburg et al. [21] make the argument that there is a need for technology and training in the area of latent palm print identification, as a significant number of identifications through latent print analysis use palm prints. There are still many agencies without these resources. Dawley and coauthors [32] through the creation and use of survey data, argue that women place a statistically significant lower importance on salary than men in their decision to enter into and stay in the field. A regression model was used [33] to argue that reduced labor density in a laboratory increases the time needed to complete forensic exams, something expected as reduced labor density would decrease the likelihood of crowding and interruptions. Reavy[34] came to a similar conclusion anecdotally with the new Utah state crime lab, where a better organized and larger facility is seen to increase efficiency and reduce backlogs. There are many other recent papers that make arguments with data, including the comparison of error rates of forensic science to clinical laboratories [4], the exploration of improvements in ballistics imaging technology and effectiveness in Trinidad and Tobago [27], and improvements in arson identification and ultimately reduction [26].

4.2 Accreditation

Accreditation is an external check of minimum standards of operation for an organization by an outside professional body. Accreditation also creates standardization and professional mobility within a country or globally. Robertson et al. [10] suggest that accreditation is one of

several keys to create a professional structure for the industry, and ultimate acceptance by the criminal justice community. This issue is therefore deeply important to the profession's development. Although achieving appropriate outcomes is important for accreditation, the process of creating these outcomes and ability to work towards continuous improvement is the emphasis. *How* the results are produced is the key, not only for the immediate cases but for those to be reviewed later by others.

Accreditation within the field of forensic science is relatively new, with many practitioners who worked in the 80's and even 90's remembering a field where accreditation was absent [2]. The recent push to increase the number of accredited forensic laboratories and the move of accreditation programs to ISO 17025-based accreditation largely coincided with the 2009 NAS Report [3]. The 2009 NAS report revealed that few formal standards of operational procedures or a common language among laboratories existed at a national or state level, and gave recommendations for improvement. Many of these recommendations overlap or complement accreditation requirements [35]. A panel of forensic professionals [2], much like the 2009 NAS report, also see the industry as fragmented and not communicate well, and state accreditation and continuous improvement are part of the path forward.

Whether through outside demands from policy makers or through internal efforts, the literature suggests that accreditation and certification will become mandatory for all forensic laboratories operating in the United States within the near future. The importance of proper evidence processing and analysis for courtroom testimony is too important, and accreditation will provide some level of guarantee and safety from error. Although accreditation does not completely prevent errors in evidence processing or improper interpretation of results, it ought to reduce the likelihood of these events.

A need was expressed for INTERPOL to create an inventory list of accredited laboratories that are able to perform post-mortem DNA analysis within a standardized framework [15, 36]. Given that an estimated 250 million people are affected by natural and manmade disasters annually, there is increased global expectation that someone is responsible for a timely and professional response [36]. After large disasters with many casualties, most laboratories nearest the disaster would be overwhelmed and require help from other laboratories with excess capacity. By having an international inventory of accredited DNA laboratories, a standardize language can be used across jurisdictions which allows for an efficient system without multiple databases, languages, and report formats which would be burdensome.

As stated, continuous improvement is a major goal in accreditation. The private sector has embraced this for centuries as a part of the competitive market process, something laboratories do not have to face [37]. Public agencies do have to face decreasing budgets and rising pressures from stakeholders, making continuous improvement and efficiency necessary [56]. The National Institute of Justice's project FORESIGHT [30] and subsequent data analysis point to the need for increased transparency and benchmark comparison as a way for accredited laboratories to see where they stand compared to industry norms in order to improve. A similar analysis using data collected from Australian burglaries and subsequent DNA and fingerprint analysis for the purpose of benchmarking, creating a national model, and identifying areas of improvement at each stage of the overall case process [14].

Transparency, although complementary to accreditation, is an "unsettling" issue for many laboratories when corrective action follows. Until the criminal justice system at large uses transparency or benchmarking for purposes of improvement and not "badgering, belittling, and beating, the three B's of those who practice in the field," transparency will be a delicate and controversial issue [2]. Indeed, police are seen as having a military culture that does

not allow true partnerships with laboratories or scientists, leading to any increased transparency as a potential for criticism and increased control and oppression of the industry by others in the criminal justice system [24]. “Mistake” to a para-military group means something very different than it does to a science organization.

4.3 Management Education

It was pointed out in the last review that the Interpol International Forensic Science Managers Symposium now includes management as a topic, and has renamed itself to include the word “managers” [6]. Although this signifies recognition of its importance, the level of managerial education, training, and performance of laboratory leadership is often questioned. The state of management within the industry has been laid out nicely by [2]. Most managers of laboratories have simply been appointed based upon seniority and are thus likely scientists by training. Although many laboratory directors are intelligent individuals with a Ph.D. in a given field, they are often not formally trained in management. ASCLD, West Virginia University, and UC Davis have made progress in offering leadership and management courses, but the scope of the skill gap among leaders is still present.

There are several challenges in closing this gap. First is the problem that when a narrowly focused, highly trained scientist moves from laboratory work to organizational leadership, what they are trained in is no longer the most important skill to be effective. People skills and organizational knowledge is separate from technological and scientific knowledge. It is also something that can be deeply complex, as in industrial engineering or economics, and its challenges sometimes ignored or underestimated. In other industries this problem has begun to be addressed at a university level with accredited academic programs. In health care, for example, health care management degrees and MBAs with a focus in health care are offered by many universities. There is no reason why this cannot happen in the forensic science industry with proper curriculum design and instructional resources.

Autonomy is the second challenge that managers have faced in two separate ways. First, as in many high skills fields, there is a certain level of professional autonomy for employees who, although there is a leadership structure, make decisions for themselves such as work schedule [8,10]. In an application of the theory of constraints, Peltokorpi marks this issue as one constraint that can lead to poor coordination and prioritization of cases that contain multiple transitions between analysis phases. The second way autonomy is a challenge is through laboratory independence. Whether or not a laboratory is controlled by a police organization or is independently operated determines power structures, laboratory culture, and potentially how cases are prioritized [10,24].

Finally, although the word “challenge” is perhaps not quite right here, gender differences are another issue that laboratory managers must be aware of and make decisions about [32]. [32], by collecting and analyzing survey data, describe many of the gender difference between why men and women in why they enter the forensic science industry, and why they stay. Unsurprisingly there were statistically significant different levels of stated importance in what determines career satisfaction by men and women, such as salary, new products and technologies, interest in field, etc. Dawley and colleagues [32] argue that managers must be aware of what makes their scientists happy, and create an organizational culture that rewards employees based upon what the employee finds important, for example working on specific types of cases that interest them.

Many other managerial techniques or skills exist that laboratory directors can be trained on to improve laboratory performance. Having a case manager as a middle man between police and laboratory scientists can reduce bias [3]. Stimson [38] shows the power of regression analysis within the context of an autoregressive time series model that can detect

process failure so corrective action can be taken. Bytheway and coauthors [9] provide an overview on how a human decomposition facility should be organized and managed. Finally, proper data management on the operations of a laboratory, described in a previous section, is critically important.

5. Ethics

It seems only natural that ethics would be a core issue for the forensic science industry and criminal justice system more broadly, given that many of the criminal justice system's activities seek to create a moral and well functioning society. Furthermore, if it is an ethical imperative to work towards the goal of a moral society, and if we can do something about it such as implement a new technology to reduce gun crime, decrease recidivism, or decrease negative testimony due to evidence contamination [26, 27, 31], ought we? Unfortunately, forensic science has been under scrutiny, whether deserved or not, for falling short in this. Contextual information and interaction between forensic scientists and investigators is a key ethical concern that has the potential to cause bias and error, a concern that the industry has been and will continue to address. Although [21] provide limited and statistically informal evidence that context and interaction has no measured effect on the identification or exclusion of individuals using forensic evidence analysis, this study has been heavily critiqued by many [39, 40] for severe methodological flaws. Indeed, the "emerging consensus" is that "observer effects are a real and substantial problem," which can only be directly observed with controlled experiments that measure an analyst's state of expectation, desire, and the ambiguity of evidence [39]. One would hope that any observed error and bias would be unintentional; however, the psychological effects caused by framing a case for even seasoned forensic scientists still have an effect on analysis and thus potentially someone's life [3].

The industry is working towards developing and implementing ideas that decrease bias and error, in order to reduce wrongful imprisonment, as well as wrongful exoneration or exclusion [39], [40], [35], and [22]. These include, creating a culture of integrity at the managerial level [2], introducing double blind proficiency testing [10], [40], [31], and [39], sequential unmasking [21], [39] and [40], reporting measurement uncertainty [3], [35], a forensic voucher system for the defense [21], removing professional indemnity for errors by expert witnesses [10], and having a case manager handle all interactions with police as a way to shelter analysts from contextual information [3]. As these changes are and continue to be implemented, controlled experiments suggested by Koppl et al. [39] would go a long way towards measuring improvement, something accreditation demands.

More broadly there are several other things the industry can do in order to improve ethics within laboratories. Dudani [41] suggests that teaching social psychology rather than ethics will lead to greater measured improvement in behavior. We should know why people behave poorly and design systems to reduce their negative effects. Siegel [42] and recommendation 11 of the 2009 NAS Report call for a national code of ethics, which includes punishment for violators. Making the code of ethics national and incorporating a licensure component, similar to the chartered financial analyst (CFA) designation seen in the finance industry, reduces the possibility that a forensic scientist who resigns from one laboratory over ethical concerns can simply start over at a new laboratory. Little has been done towards developing [42]. Finally improving the perception of outsiders such as trial lawyers would increase the effectiveness of forensic expertise and diminish obstacles such as the weak evidence effect or exoneration by negative testimony [3], [35],[43].

6. Third Party Perceptions of Forensic Science

CBS's *CSI: Crime Scene Investigation* television series, one of the most successful nighttime crime dramas, has aired 797 episodes within four separate franchises to date. Many video games, toys, novels, and other merchandise have also come out of this television show and its kind. It is clear then that society's interest in forensic science is strong. Although interest in forensic science is to be encouraged, the impression programs like *CSI* give to the public, and potentially others working in criminal justice system, of the how laboratories operate is frankly unrealistic and potentially dangerous [44]. Nonetheless, how forensic science is perceived is, unfortunately, mostly out of the control of the industry.

The most influential element in determining the perceived identity of forensic science is the media in the eyes of those working within the legal system [24]. In many cases the expectations that the media creates for the public and family members of victims are inconsistent with the speed and scientific reality of the present state of the industry [15]. University students who see the excitement on television and in the news might become mistakenly interested in forensic science because of this, but may not fully appreciate the educational background and serious effort needed to work in the field [2]. A panel of forensic professionals [3] argued that those students who are influenced by the *CSI* effect are exactly the ones you do not want working in a laboratory. With the televisions focus on nabbing the bad guy rather than objective science, error and bias could be increased by training and hiring *CSI* fans rather than true scientists.

The media's continued dramatic presentation of forensic science failures have also not helped in outside perceptions of the industry [45]. It is not obvious why the errors of forensic science, although an important issue, are focused on in a way that seems excessive compared to the errors observed in other industries [2,4]. Perhaps the fear of being wrongfully imprisoned or wrongfully executed by forensic science errors simply makes for better television than medical malpractice. However the mere existence of the 2009 NAS report which was critical of the entire industry shows that it is more than just that. As Houck et al. (2014) state, "what you don't see is these reports addressing the entire medical profession" or other fields ([2], page 54). It is unclear whether any error rate beyond zero would be acceptable by the media.

There are some ways that the industry can improve their media perception. [22] for example provide details on DNA elimination database legislation that ought to reduce error rates and negative testimony, something which should increase public trust. Byethway et al. [9] provide best practices for human decomposition facilities including how to handle the media. Reducing the chance that photographs are taken, especially with cell phones, to preserve the dignity of the dead is one of the most important issues in maintaining good relations with the public through media.

The perceptions police have of and create for forensic science is another important aspect to third party perceptions. The political influence and budgetary control of laboratories by police units are common. Being a strong stakeholder, the police undoubtedly want to direct the image of forensic science, and currently do in a way that dominates the influence of forensic science practitioners and educators. Furthermore, the police's interest in solving crimes creates the incentive to manipulate the science and interpretation of evidence to bring about prosecution [24]. Koppl and Saks [46], for example, find that many laboratories are funded in part by the number of convictions obtained, a poor incentive structure if objective science justice is the goal. [The use of certain words, such as "we are looking for the suspect's DNA" rather than "we are looking for the assailant's DNA" which would reduce police influence [3]. When mistakes happen and are detected by the media, whether the fault of police influence or not, the image of forensic science is damaged.

Reducing the negative effects of police influence is possible, even short of creating independent laboratories. For example, a panel of forensic professionals [3] suggests that lawyers ought to have specialized qualifications to handle criminal cases involving forensic science analysis, much like some jurisdictions require special training by lawyers to handle death penalty cases. This training would allow lawyers to see more clearly how powerful or weak forensic science evidence is in a courtroom. Additionally [35], [47], [27], [3], and [26] argue that improvements in laboratory reports would better communication, and thus perception of the industry, among outsiders. Most forensic science communication occurs through written reports, so its readability and language are perhaps just as important if not more important than courtroom testimony. Howes et al. [35] suggest standardized organization, standardized terminology, a glossary of terms for non-scientists, and a statistical expression of uncertainty so outsiders can understand the power of analyzed evidence. Morrison and Stoel [47] suggest that a statistical expression of uncertainty is preferable to expert opinion since it is more objective and less easily manipulated for reasons described in the previous paragraph.

As a final point, third party perceptions within the industry could also be improved. Forensic science is a very fragmented and multidisciplinary field [2,10] often requiring multiple scientists to process one case. As such, quality communication across the disciplines ought to occur, but unfortunately often does [2]. Houck et al. (2014) state:

“toxicologists do not always know what the chemist are doing, even though the toxicologists are seeing the same drugs that the chemist are seeing they’re just in a different environment. I don’t think that we communicate well enough and I think there is so much going on in the U.S. and I hate to say but a lot of forensic labs are almost like a factory environment, there is so much coming in and so limited resources that its just like what can I do to get the cases done as quickly as possible.” ([2], page 55)

Improving communication within the laboratory could go a long way towards bettering the perception scientists have of each other, and bettering the perception that outsiders have of the industry as a whole.

7. Looking to the Future: Forensic Science’s Coming of Age

A panel of forensic professionals discuss the biggest changes that will affect forensic science in the next 5 years, how will the environment change, how will society change, how will the profession change, including police and courts? [2] Several potential changes are offered, such as arrestee DNA legislation, rapid DNA, increases in the caliber of new personnel, improved focus on the sciences, training, independence, and transparency. Others changes have been offered, such as [10] who describes a coming of age within the discipline, [48] that forecast increased use of forensic intelligence or big data, and [3] who hope for increased standardization at the national level. Although there is no crystal ball, none of these potential changes listed are completely new or unexpected. Their prominence within the industry however is mounting.

As forensic science experiences its coming of age and defines its professional status [2,10] several issues must be clarified. What is the difference between a technician versus scientist, how do we view education versus training, are our managers scientists first or visa versa, do we create national systems or maintain fragmentation at least in the United States, is forensic science a discipline or a group of disciplines such as chemistry, anthropology, archaeology, and biology, and what are our ethical codes [10, 2, 3, 8, 30]? Forensic science has been and needs improvement [2] to advance its professional status, and the way in which the industry clarifies these points will determine the direction it goes. Non-scientists do have a voice in defining the industry and the direction it goes, but one hopes that they are

not the dominant voice [10]. Embracing accreditation, implementing laboratory independence broadly, developing a research culture, developing a code of ethics, and establishing standards grounded in empirical data will all go a long way towards improving forensic science and increasing the voice of the profession over outside stakeholders. If not, there is the danger of “falling apart, spinning out of control into a thousand sub-sub-sub disciplines, and with that goes any hope of lobbying in our favor” ([2], page 68). Four of these issues will be addressed separately, including scientific validation, national standards, forensic intelligence, and laboratory independence.

7.1 Scientific Validation

In September of 2016, the U.S. President’s Council of Advisors on Science and Technology (PCAST) discussed a draft report, *Forensic Science in Criminal Courts: Ensuring Scientific Validity of Feature Comparison Methods*, that made several significant recommendations, including,

- The combined-probability of inclusion (CPI) method for evaluating complex DNA mixtures (more than 2 donors) has no valid scientific foundation. The Council also recommended that probabilistic genotyping software must be properly validated by independent third parties before being routinely used in criminal cases.
- The following comparative methods fail to meet scientific standards and should be inadmissible in criminal courts;
 - Footwear (except for size and make)
 - Bitemarks,
 - Tire tread,
 - Microscopic hair
- Fingerprinting has made significant advancement and is “heading in the right direction”; the Council had high praise for the FBI’s recent “black box” studies.
- Firearms comparisons require more explicit explanations to juries and lack a suitable foundation as a science.

This comes from a pre-release draft version of the report that, while open to public comment, was voted on favorably by the Council “without any substantive changes” in an August XX, 2016 public conference call. The political posturing began immediately with the National District Attorneys Association [49] crying foul:

The forensic science disciplines that the PCAST authors attack are (and have been) reliably used every day by investigators, prosecutors, and defense attorneys across the United States to aid in both exonerating the innocent and convicting the guilty... Engagement with recognized subject matter experts would have led PCAST to compelling foundational support for the overall reliability of these forensic sciences. Notwithstanding the lack of qualifications, PCAST has taken it upon itself to usurp the Constitutional role of the Courts and decades of legal precedent and insert itself as the final arbiter of the reliability and admissibility of the information generated through these forensic science disciplines... The determination of whether scientific evidence is reliable and therefore admissible must remain with the judiciary -- while the determination of how much weight will be given to any particular piece of scientific evidence must rightfully remain with the jury. ([49], pages 1-2). The last sentence is telling of the persistent cultural split between law and science: The prosecutors see the judges as arbiters of science and the juries as the deciders of significance, leaving the role of the scientists completely unstated. Hyperbole aside, the PCAST does carry the weight of top scientists and the imprimatur of the White House (until the November Presidential elections, at least). Forensic science, policy, and politics will be interesting in the years to come.

7.2 National Standards

As a profession, forensic science is fragmented, with each discipline often operating within their own separate “silos,” with poor communication even at the laboratory level, and certainly at the national level [48], [2], [3], and [10]. This can create a situation where we have scientists performing as if “a bunch of cowboys” [3], working as “journeymen” [50] within a laboratory, where professional autonomy goes too far and cohesion is lost. The ability to standardize the industry then is an important task, which accreditation has made some progress in solving. Standardization creates a system that is more easily understood by outsiders, and allows the transference of skills across laboratory systems and geographic borders by employees [10]. As the world and criminals become more global, this standardization will need to encompass more than one nation [15], [48], [33],[35], and [27].

Some progress has occurred in the creation of common languages, including the National Institute of Justice’s project FORESIGHT, the United Kingdom’s SWIM study, and subsequent Australian application of SWIM, and Standards Australia [14], [30], [29], [35]. By creating a common language, questions on the industry are possible, which otherwise would not occur. By creating nationally integrated and standardized data systems in which data definitions mean the same across jurisdictions [48], the full power of potential evidence can become realized. DNA databases of former criminals [23] as well as DNA elimination databases [22] are great examples that benefit from economies of scale and the standardization of data across a wide geographic area. Forensic reporting is another application which [35] outline. Using Australia as a case study, [35] suggests a standardized reporting format, glossaries, expressions of uncertainty, standard terminology, etc. as a way to improve communication and standardization across forensic science. This model could become an adopted model internationally as there is currently no international standard in forensic science reporting. Short of creating a truly national system as seen in New Zealand [51], standardization will create some of the benefits of a national system while still maintaining distinct laboratory systems and jurisdictional independence, for better or worse.

7.3 Forensic Intelligence

“The most valuable commodity I know of is information; wouldn’t you agree [52]?” Although forensic scientists certainly handle, create, and interpret information through data, they are not specifically trained as data scientists. Machine learning, econometrics, geographic information systems programming, etc. are skills that are in the domain of a variety of fields including economics, industrial engineering, statistics, and computer information science, but are often not in the skill set of even highly trained forensic scientists [48]. However, these skills are becoming increasingly and deeply important for improving laboratory performance and solving crimes, and will continue to do so well into the future. Bruenisholz et al. [48] have labeled this type of advanced data analysis within the industry as forensic intelligence, which they state “is in its infancy as the focus of forensic science has traditionally been the court and the resolution of crime.” As the discipline moves forward, further sub-disciplines within forensic science may be developed to address this new area of forensic science, by hiring or consulting professionals who are skilled in data analysis and pattern recognition.

To a degree this is already happening. DNA matching software [15], DNA criminal databases [23], DNA elimination databases [22], laboratory information management systems [8], applications of autoregressive regression models [38], and data collection on performance standards [30], [29], and [14] are all developments in data science and information analysis that most forensic scientists who began their careers in the 80s or even 90s did not have available in any meaningful way. As computers continue to become integrated into the workplace, the data they generate and interpret will have value that will certainly improve the criminal justice system in ways that we can only just now imagine.

7.4 Laboratory Independence

The ability of a forensic laboratory to do work based in science and generally accepted industry norms is undeniably important, and creates an environment of objective justice. However, when outsiders maintain a degree of control over a laboratory, this environment can become contaminated as third party objectives are often separate and mutually exclusive of the scientists' and objective justice [24]. The issue of laboratory independence was therefore presented in the 2009 NAS report, which served to legitimize and create more value for arguing for independence moving forward [3]. The political difficulties in achieving independence however are still not small. Police, prosecutors, judges, and even mayors have a vested interest in maintaining control of the political process [3,53]. In the context of a police controlled laboratory, these interactions and pressures would be greater, thus increasing bias and error.

Creating independence from police would be a step, but not the only one needed to achieve true independence. The District of Columbia created a new forensic agency separate from police in 2012 but efforts at functional independence were derailed by political pressure cloaked in scientific critique [54]. The Houston, Texas, Forensic Science Center continues to battle for its independence from the city police department. As Thompson noted, when the top management of the DC Department of Forensic Science was removed for disagreeing with Federal prosecutors' experts about DNA methods (which were not standardized at the time and a codified method was later offered by some of the agency's critics [55] but disavowed by the White House PCAST),

...this [the removal of management] sends a strong message to laboratory directors nationwide who come into conflict with local prosecutors. The message is be afraid, be very afraid. That, in itself, is a serious setback for efforts to protect the scientific independence of crime laboratories. [54]

In an article describing Jay Siegel's recent resignation from the Scientific Advisory Board for D.C.'s Department of Forensic Science after the firings and resignation of top management at D.C.'s Consolidated Forensic Laboratory [53], it describes a situation where hopes of laboratory independence were dashed by city government in order to maintain the status quo and political control of criminal justice. In the article, Fatzick quotes Siegel, who states that the firings were "hasty and unwarranted" and were not related to scientific quality within the laboratory. This is evidence enough to suggest that independence from a police unit is not necessarily true independence, if a job requires something other than scientific objectivity by a third party such as a city. Whether forensic science independence continues as a political issue remains to be seen but the chances for the U.S. remain slim.

8. Conclusion

The answer to the title's question is that we will always be on the path forward, if continuous improvement is a part of our culture and a requirement for accreditation. As noted by Houck, despite some research improvements to the science, the current state of encroaching Federal oversight is the profession's own fault: "Paradigm shifts are inherent to science; we may be in the midst of one right now...the shift may occur and bypass us without our ever having been actively involved. We can change our science and profession or it can be changed for us. One is more beneficial for us in the long run." ([4], page ii). The 2009 NAS report served to push forensic science onto the path and keep it from drifting, but we should not think of this path as leading to a destination if the industry is to remain a dynamic and adaptable part of the criminal justice system. New technologies, new criminal techniques, or

a shifting legal environment will make the managerial process and continuous improvement evermore important.

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