FINGERMARKS AND OTHER BODY IMPRESSIONS – A REVIEW

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Table of content

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 introductive 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 Purkynje 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 Associated 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).[1]

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, especially in the USA. The public attention was especially turned to bitemarks with a series of articles initiated by the investigation of Radley Balko of the Washington Post.[2] 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 Dandridge[3] 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

[1] See : http://www.nytimes.com/2016/01/23/science/who-ever-said-no-two-snowflakes-were-alike.html?

[2] https://www.washingtonpost.com/people/radley-balko

[3] https://www.law.umich.edu/special/exoneration/Pages/casedetail.aspx?caseid=4768

fingerprints.[4] 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 evidence.[5] The Statistical and Applied Mathematical Sciences Institute (SAMSI) offered from 2015 a program in forensic science).[6] 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 fingerprints.[7] When you combine all these efforts (in the US mainly) with the Organization of Scientific Area Committees for Forensic Science (OSAC)[8] 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)[9] 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:

- 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).

[4] https://www.aaas.org/page/forensic-science-assessments-quality-and-gap-analysis

[5] http://forensic.stat.iastate.edu

[6] https://www.samsi.info/programs-and-activities/year-long-research-programs/2015-16-program-on-statistics-and-applied-mathematics-in-forensic-science-forensics/

[7] 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

[8] http://www.nist.gov/forensics/osac.cfm

[9] https://www.justice.gov/ncfs

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* of *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".[10] 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.

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).

[10] *Illinois v. Christopher Robertson*. Motion to Exclude Statements Officer *XXX* Claim of Fingerprint "Identification", Circuit Court of Cook County, 15 CR 7788, filled 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, filled June 13, 2016.

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-clock wise 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 reseachers 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 studies 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 reports[11] 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.

[11] 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 fingermark 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 fingermarks (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 fingermark aging has been published (209), encompassing the different existing methods and compounds of interest identified so far. Legal considerations of fingermark age determination received also a deserved attention (221).

Persistence: The question of persistence of a fingermark when it is exposed to (detrimental) environmental elements has been addressed (222), including recommendations regarding some common assumptions linking the age of a fingermark with its easiness of detection. The influence of light on fingermarks left on brass was also briefly explored (223).

Artificial secretion: The use of artificial secretions is sometimes presented as a reproducible way to leave fingermarks presenting similar chemical composition.

The efficiency of (commercially-available) artificial mixtures has been evaluated and compared to actual fingermarks 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 fingermark 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 fingermark detection. In this context, Moret *et al.* (210) conducted a study based on the optical/microscopic observation of different types of fingermarks (*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-polaryzation) 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 fingermarks behave when processed with conventional detection techniques (*i.e.*, IND/Zn, NIN, ORO, PD), in regards with actual fingermarks (natural and sebum-rich). Difference of behaviour were observed: (a) sweat/eccrine pads seem to contain more amino acids than in an actual fingermark, 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 fingermarks, 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 fingermarks. Both these studies show that the consideration of artificial secretion is still of interest in the field of fingermark 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 Fingermark 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 fingermark 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 surprizing 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 fingermarks.

IFRG guidelines – In an attempt to provide guidelines for people interested in performing research in fingermark 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 fingermark 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/fingermark considerations. As a reader, the combination of the following elements should raise concerns: (a) exotic technique barely applied for fingermark detection, (b) small-scale study including only a couple of donors, sebum-rich and fresh fingermarks, 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.

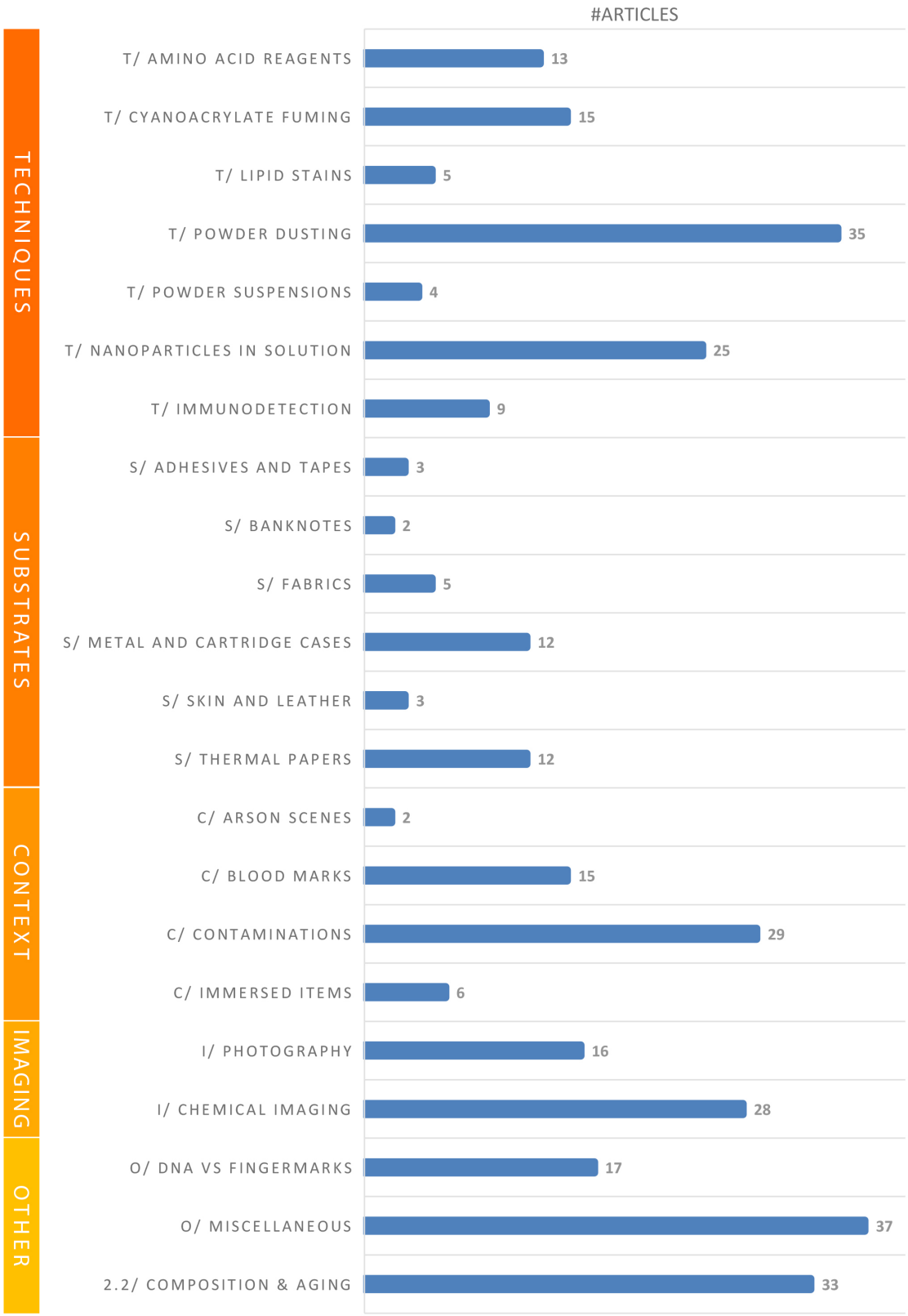

Figure 1 *– Number of articles per defined category (please note that some articles can be present in two categories if they present more than one main scope)*

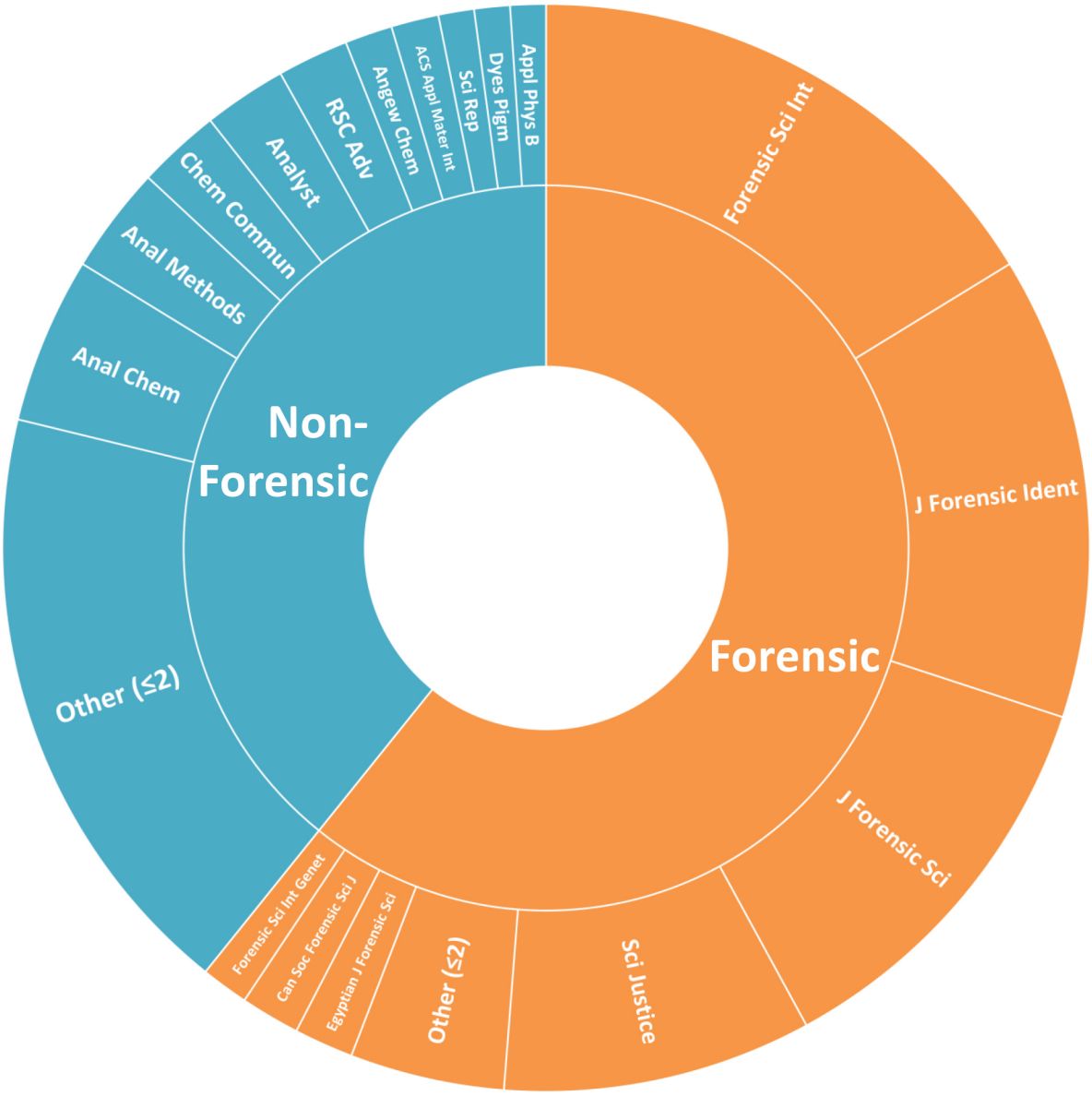

Figure 2 – *Sunburst representation depicting the number of articles per journal, for all contributions cited in sections 2.2 and 2.3. The sub-category "Other (≤2)" contains all the articles having been published in journals in which a maximum of two articles dealing with fingermarks were published in 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 fingermarks 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 fingermark 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: **^{13}C-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 fingermark 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

<u>One-step luminescent CA</u>: 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 fingermark 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 comparaison 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 fingermark 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 fingermarks 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 fingermark 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 fingermarks, 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 fingermarks (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 fingermarks, 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 Fe_3O_4-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; (b) 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 57-67µm.

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., TiO_2) 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, MoS_2-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 $ZnCO_3$-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 fingermarks 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 fingermarks (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 fingermarks 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), Cu_7S_4 nanocomposites (316), lanthanide-based upconversion NPs (317), lanthanide-doped silica NPs (272), and ZnO-SiO_2 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 fingermarks 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) SiO_2 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 fingermarks 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 fingermarks 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 fingermarks, 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 fingermarks 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 fingermarks 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 fingermark 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 fingermarks 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 fingermarks 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 fingermarks]*

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 fingermarks 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 procotol 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), **H_2O_{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, H_2O_{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 fingermarks 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 fingermark 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 fingermark 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.*, $AgNO_3$, 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 fingermarks. 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/m^2 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 $ZnCO_3$ 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 fingermark 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 fingermarks exposed to (detrimental) environmental elements (222).

(Illicit) Drugs (handling): Fast Blue B was proposed as a new reagent to detect THC-rich fingermarks, 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 fingermarks + the link with the activity of handling cannabinoid may not be straightforward. Chemical imaging of contaminated fingermarks 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 fingermarks (379). Aptamer-functionalized NPs were used to detect cocaine-contaminated fingermarks (320). Other studies involved the analysis of drug-spiked fingermarks: oily marks using DAPNe-NSI-MS (380), sebum-rich fingertips using DART-MS and MALDI-MS (375).

Endogenous metabolites (drug consumption): Fingermarks 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 fingermarks using LC/MS (376, 382); The main outcome of these studies is to evaluate how fingermarks 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 fingermarks: 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 fingermarks 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 Cu_7S_4 nanocomposites) (316). Explosive residues (i.e., ammonium nitrate, black powder, smokeless gun powder, dynamite) were detected/mapped in finger-/hand-marks 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 fingermarks 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 fingermarks 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 fingermarks 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 fingermarks, 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 fingermarks on immersed items and to detect them afterwards (395). To study the detrimental effect of immersion onto fingermark 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, fingermarks 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 fingermarks 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" fingermarks. 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 fingermarks 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 fingermarks has been studied by Goldstone *et al.* (396). Their study consisted in exposing glass panels bearing depletive series of fingermarks 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 fingermarks. 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 fingermarks 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 fingermarks 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 fingermarks 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 fingermarks on flat non-porous substrates. Various optical contactless imaging techniques have been proposed to record latent fingermarks: 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 fingermarks 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 fingermarks 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 fingermarks on particularly difficult substrates.

2.3.19 I/ Chemical imaging

Imaging of latent secretions: Several techniques were applied to image latent fingermarks, 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 be 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 fingermarks, 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 fingermarks 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), **αCHCA** (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 fingermarks) consist in proof-of-concept studies based on (artificially-)enriched fingermarks 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 fingermarks 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 TiO_2 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 TiO_2 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 fingermarks and further analyse them. As a conclusion to their study, the authors provided a proposition of operational workflow integrating MALDI-MSI. In another study, fingermarks 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 fingermark 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 fingermark detection techniques are described in section 2.3.16, for they are dealing with contaminated fingermarks.

2.3.20 O/ Fingermark detection and DNA analysis

"Touch-DNA": "Touch-DNA" can be defined as the genetic material that is extracted from fingermarks, 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 fingermarks (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 fingermarks (424). Other studies aimed at evaluating the impact of fingermark 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 fingermark 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 (cheiloscopic 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 marls 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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