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## Interpol review of fingermarks and other body impressions 2016–2019

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## ABSTRACT

This review paper covers the forensic-relevant literature in fingerprint and bodily impression sciences from 2016 to 2019 as a part of the 19th Interpol International Forensic Science Managers Symposium. The review papers are also available at the Interpol website at: <https://www.interpol.int/content/download/14458/file/Interpol%20Review%20Papers%202019.pdf>.

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## 1. Introduction

Ten years after the report of the National Research Council that highlighted a dearth of fundamental research in forensic science and especially in the forensic identification fields [1], much has been done, but the task is daunting. We are happy to report on the main research initiatives published during the review period.

This review period (2016–2019) has been very rich in publications and it is obvious that this review cannot be exhaustive. We will focus on papers and reports published in peer-reviewed journals or books mainly.

A few monographs have been published during the period, dealing with either detection or identification. For detection, we recommend the book by Bleay, Croxton and de Puit [2] as it covers extensively the area of fingerprint detection, including: formation mechanisms, composition and properties of the secretion residue, optical methods, detection techniques and sequential processing. A book by Kasper covers fewer techniques [3]. For identification, we note the release of the second edition of the books by Moses Daluz [4,5].

**Publication trends on detection issues** – The combination of

the articles covered in Sections 3.1 (Fingerprint composition and evolution with time) and 3.2 (Fingerprint detection and imaging/recording) results in 393 articles published in 128 different journals. When distinguishing these journals by their main editorial scope (i.e., “Forensic” and “Non-Forensic”), it appears that the number of articles published in “Non-Forensic” journals (235) surpasses those published in “Forensic” journals (158), as illustrated in Fig. 1. This is the first time that such an inversion occurs, and it must raise questions about the targeted readership, the technological drift, or the loss of forensic interest for researchers related to fingerprint composition/detection.

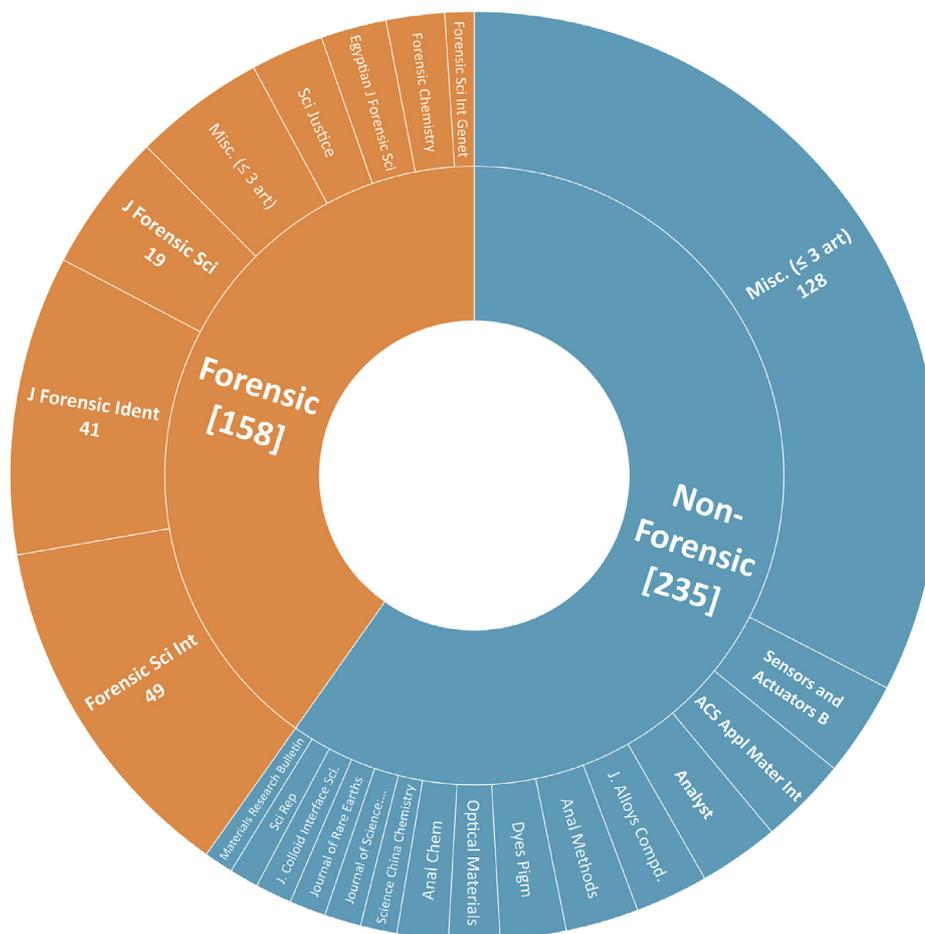
Beyond absolute numbers, a clear difference in publication behaviour appears when looking at the distribution of journals. In Fig. 1, only journals that have published at least four relevant articles in the covered period appear by name; otherwise, they are grouped in the “Misc ( $\leq 3$  art.)” category. With regards to “Forensic” journals, the three most popular are *Forensic Science International* (FSI – 49 articles), *Journal of Forensic Identification* (JFI – 41), and *Journal of Forensic Sciences* (JFS – 19). Individually, they represent 12%, 10% and 5% respectively of all articles that were published. But together, they encompass approximately 70% of the articles published in “Forensic” journals. This means that these three journals still enjoy a great popularity and represent an anchor for people wishing to target the forensic readership. When considering “Non-Forensic” journals, almost all of them are chemistry-oriented. The three most popular are *Sensors and Actuators B* (13 articles), *ACS Applied Materials & Interfaces* (12) and *Analyst* (11). Altogether, they

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**Fig. 1.** Sunburst representation depicting the publication portfolio associated with the contributions cited in Sections 3.1 and 3.2. The sub-category “Misc. ( $\leq 3$  art.)” groups all the articles that have been published in journals associated with a maximum of three articles for the covered period.

represent 9% of all articles and 15% of the articles published in “Non-Forensic” journals. This means that there exists no reference journal for people wishing to publish chemistry-oriented research linked to fingerprint composition or detection. In our 2013–2016 report [6] (hereinafter the 2013–2016 report), *Analytical Chemistry*, *Analytical Methods*, and *Chemical Communications* were the three major “Non-Forensic” journals, confirming the fact that no chemistry-oriented journal can be labelled as a reference one so far. Also, the number of articles published in journals encompassing three articles or fewer during the covered period increased to 128, which represents 33% of all articles and 54% of the articles published in “Non-Forensic” journals. This trend has worsened compared to 2013–2016 and may reflect a lack of relevant journal options or may be a consequence of successive rejection decisions from the most famous forensic or chemistry-oriented journals. The publication trends will be further discussed in Section 3.2.

## 2. Friction ridge skin and its individualization process

We note a few publications describing the practice in their jurisdiction, e.g. in Switzerland [7], or Poland [8]. Sometimes the analysis comes with a strong call for change in the practice, typically in France where the 12-point rule is still a dogma [9]. We remain aware and alert to the fact that fingerprint evidence may be faulty; it is worth repeating that the number of wrongful convictions in which fingerprint evidence played a key role is limited

[10,11]. The profession has to minimize these occurrences but more importantly increase its transparency.

### 2.1. Statistical studies associated with friction ridge skin features

#### 2.1.1. Level 1

Pattern types received some effort in the community mainly by comparing pattern frequencies between populations. We note a study of Middle Eastern populations [12] showing small differences between Iraqi and Afgani populations.

Level 1 features (general patterns and associated measures such as ridge counts and tracing) can also help in determining the finger number of the source of a given mark. The direction of whorl slants are good indicators to distinguish right from left hands [13]. Ridge densities have still received interest in their capacity to predict gender [14–20] or stature [21].

One work stands out due to its depth and applicability. De Jongh et al. [22] presented the results of a survey of primary class distributions in a Dutch fingerprint collection (24 104 fingerprints). Their classification scheme comprises 8 subclasses of arches, 8 for loops, and 20 for whorls. The added value of the study lies precisely in the number of subclasses considered. This data serves as the basis to assign evidential weight to fingerprint general patterns when visible on marks. They compared their results with legacy data (with less subclasses) including a recent study involving Algerian prints [23].

### 2.1.2. Level 2

Gender determination based on general pattern or ridge density is not new, although the power of these inferences is rather limited when dealing with partial, smudged marks recovered from crime scenes. The frequencies of minutiae have been added to the set of predictors recently [24]. Differences in number of minutiae and type have been reported in the past as well. The predictive ability of these level 2 features remains unknown.

Swofford et al. [25] published a paper presenting the development and validation of the software FRStat used by the U.S. Department of Defense. From a similarity value (called GSS) obtained between two sets of features (typically between a mark and a print), the software derives a statistical measure that indicates how often such a GSS would be obtained in cases where marks and prints come from a common source compared to cases originating from different sources. This software is now used operationally by the fingerprint experts of the Defense Forensic Science Center, and their reports account for that statistical value [26]. Such tools respond to the call for objective measures in forensic science. The computed metric reduces a large source of variability – inter-examiner variability in decision-making – in the latent print analysis process [27].

The performance of a score-based likelihood ratio system (using the algorithm of an AFIS system) has been studied [28]. The research team showed that there is substantial evidential strength for same source comparisons that currently do not meet the Dutch twelve-point standard. The general principles to move from a biometric score to a likelihood ratio are given in Ref. [29]. Note that Meuwly et al. [30] published an excellent guideline to help scientists with the validation of such statistical based methods. The fingerprint data used to illustrate the application of the guideline have also been published [31].

Lee et al. developed an effective system based on distortion modelling to allow an early detection of potential mis-attribution or mis-pairing occurring during the comparison process [32]. It is based on a computed similarity score associated with a set of paired minutiae between a mark and a print associated with a score associated with the proposed pairing of minutiae. If the score obtained in the case at hand is an outlier compared to scores obtained from cases originating from the same source, it would trigger an alarm forcing the fingerprint examiners to review the case and potentially articulate the reasons such a level of distortion has been observed.

### 2.2. Measuring suitability of fingerprint marks

We will detail later the research from this review period that showed that fingerprint experts may vary substantially on their decision to declare a mark as being “of value”, even when they are presented with the same images more than once. The need for objective ways to assess suitability cannot be overstated. In that context Neumann et al. [33] presented a statistical model that can be used to facilitate the latent print examination workflow by predicting whether a mark should be searched in AFIS or not based on the quantity information (number of minutiae and specificity of the configuration) and quality of information (latent quality metrics available in the ULW software) available in the mark.

### 2.3. Reporting fingerprint evidence in court

In our previous Interpol review reports, we outlined the regular discussion and evolution regarding the nature of conclusions that fingerprint experts may provide [34]. There is a general call for more modest claims. Some will go as far as saying that all conclusions must be expressed in probabilistic terms [35].

A model forensic scientist [36] would be expected to be fully transparent in his report and testimony highlighting the uncertainties and limitations associated with his/her conclusions. Individualization (claimed of an identification to the exclusion of all others) is not dead yet, but a few additional nails have been added to the coffin. As Eldridge puts it: as least in the U.S. “There is a shifting landscape of latent print testimony” [37]. In his formal review of fingerprint evidence, Kadane concluded that: “there is no scientific basis for a source attribution; whether phrased as a “match,” as “individualization” or otherwise” [38]. Not only does it apply to the fingerprint fields, but all areas of forensic science dealing with identification issues including forensic medicine [39].

In the McPhaul case in North Carolina (State v. McPhaul, 808 S.E.2d 294 (N.C. Ct. App. 2017)), the state appellate court ruled in favor of the defense and identified a reliability problem with latent fingerprinting. Garrett [40] highlighted that judges, as in this case, will ask more for a transparent testimony of what process has been followed and how the conclusions have been reached. Testifying by *ipse dixit* to an identification will no longer suffice.

In September 2016, the U.S. President’s Council of Advisors on Science and Technology issued a report [41] with an addendum [42] [hereinafter the PCAST report] that impacted most forensic fields including fields dealing with pattern impressions. Very critical of bitemark evidence, footwear marks, and firearms, the report praised the effort made in the fingerprint field to measure error rates using black box studies. Overall, the report led to the often-heard call for more metrology in forensic pattern evidence disciplines [43], and for more science and statistical research in forensic science [44–46]. Reactions were also numerous in the legal community, for example [47–49]. On one side, the PCAST report is viewed as an effective tool to exclude, limit, or debunk an expert’s opinion [50]. On the other side, it gives the agenda for further research and educational efforts [51].

A complete list of reactions can be found on <http://for-sci-law.blogspot.com/2016/11/index-to-comments-and-cases-discussing.html>.

The PCAST committee warned against the use of terms that convey explicitly or implicitly absolute certainty and suggested to adopt the term “possible identification”:

*“Because the term “match” is likely to imply an inappropriately high probative value, a more neutral term should be used for an examiner’s belief that two samples came from the same source. We suggest the term “proposed identification” to appropriately convey the examiner’s conclusion, along with the possibility that it might be wrong.” (pp. 45–46).*

A report [52] prepared under the auspices of the American Academy for the Advancement of Science (AAAS) is an excellent overview of the current state of affairs in the fingerprint field. It is a must read in our opinion. It also warned against making overly bold claims, for example:

*“Latent print examiners traditionally claimed to be able to “identify” the source of a latent print with 100% accuracy. These claims were clearly overstated and are now widely recognized as indefensible. While latent print examiners may well be able to exclude the preponderance of the human population as possible sources of a latent print, there is no scientific basis for estimating the number of people who could not be excluded and, consequently, no scientific basis for determining when the pool of possible sources is limited to a single person.” (p.71).*

The American Statistical Association (ASA) “strongly

discourages statements to the effect that a specific individual or object is the source of the forensic science evidence. Instead, the ASA recommends that reports and testimony make clear that, even in circumstances involving extremely strong statistical evidence, it is possible that other individuals or objects may possess or have left a similar set of observed features" [53]. There is a strong push from the scientific community towards the adoption of a transparent, data supported, probabilistic reporting framework instead of the traditional categorical reporting.

Swofford and Cino [54] reported that 71% of the 300 potential jurors surveyed interpreted expert testimony containing the word "identification" (or "identified") to imply a single source attribution "to the exclusion of all others". Kadane and Koehler [55] showed how much weight a conclusion of "identification" may convey to lay jurors, regardless of the limitations raised during cross-examination. Thompson et al. also investigated lay people's perceptions of the relative strength of various conclusions that a forensic scientist might present about whether two items (fingerprints, biological samples) have a common source [56]. They compared various ways to report source conclusions (likelihood ratios, strength of support statements, random match probabilities, likelihood of the observed similarities, source probability statements, and categorical statements). The results suggest that statements involving numbers are perceived as very powerful. For a fingerprint comparison, the random match probability (RMP) of 1 in 100 000 was as strong as the categorical statement that the examiner had "identified" or "individualized" the print. This shows that lay people are not expecting an association to the exclusion of all others, regardless of the certainty proclaimed by the expert. Garrett et al. [57] showed similarly that the traditional categorical approach to fingerprint evidence carries great weight with laypersons, but also that a strong probabilistic statement about the likelihood of a match carries similar weight. Finally, Ribeiro et al. [58] showed that laypersons gave fingerprint comparison a mean accuracy rating of about 88%, much lower than the actual empirical data (obtained through black box and white box studies).

To the question "How Should Forensic Scientists Present Source Conclusions?", Thompson [59] responded by reviewing various reporting options and by delineating two conditions that any source identification report ought to meet:

- (1) whether the reported conclusions can be justified logically and empirically; and
- (2) whether the reported conclusions will be understood and used appropriately.

The traditional way to present fingerprint evidence (and other impression evidence as well) in the form of categorical opinions fails the first condition and additional work is required to make sure that forensic examiners would [59] "present their findings in a manner that causes people to respond to the evidence in a manner commensurate with its probative value". It is a new era in which statistics will inevitably play a greater role [60]. But we have to recognize that communicating statistically based conclusions to jurors is a complex exercise [61] and individuals tend to fail to incorporate the probative value of the statistical evidence [62]. For a full review of juror comprehension of forensic expert testimony, refer to Eldridge [63].

The U.S. Department of Justice issued an approved uniform language for testimony and reports (ULTR) for the forensic latent print discipline [64] that defines source identification as:

*'Source identification' is an examiner's conclusion that two friction ridge skin impressions originated from the same source. This conclusion is an examiner's decision that the observed friction*

*ridge skin features are in sufficient correspondence such that the examiner would not expect to see the same arrangement of features repeated in an impression that came from a different source and has found insufficient friction ridge skin features in disagreement to conclude that the impressions came from different sources.*

*The basis for a 'source identification' conclusion is an examiner's decision that the observed corresponding friction ridge skin features provide extremely strong support for the proposition that the two impressions came from the same source and extremely weak support for the proposition that the two impressions came from different sources.*

*A 'source identification' is the statement of an examiner's opinion (an inductive inference that the probability that the two impressions were made by different sources is so small that it is negligible. A 'source identification' is not based upon a statistically-derived or verified measurement or actual comparison of all friction ridge skin impression features in the world's population.*

The ULTR also states that latent print examiners shall not "individualize" or assert that forensic latent print examination is infallible or has a zero-error rate. The ULTR has been received with mixed feelings in the legal community [65]. Indeed, Cole judged that the proposed categorical reporting framework with three categories—identification, inconclusive, and exclusion—even if defined differently than in the past, perpetuates the status quo. Reporting a fingerprint comparison as "inconclusive" when no categorical conclusions can be articulated is a very common traditional approach. Cole argues that the ULTR is no different as no nuances or shades of gray are associated with inconclusive decisions. Inconclusive decisions have been discussed through the use of a cognitive model which takes into account that decisions are an outcome of interactions and intersections between the actual data and human cognition [66].

A coherent analysis, based on decision theory, of the ingredients of the decisions taken by fingerprint experts is found in the work of Biedermann and colleagues [67,68]. They highlight that experts are guided by decision-making goals that need to be articulated. Two of their publications aimed at conveying these concepts in a less formal or mathematical approach [69,70] and include an discussion of the inconclusive decisions [71,72] in response to Refs. [66]. They also suggest that identification decisions should be the remit of the factfinder and not be taken by experts [73].

Another aspect of fingerprint reporting is related to the ability of experts to opine in relation to the activities that led to the deposition of the detected marks. De Ronde et al. have shown the position, direction, area, and location of marks can be used, helped with a Bayesian network [74]. Such anatomical features of detected marks on pillow-cases assisted in determining if they were deposited while smothering or by a regular changing activity [75].

The retrial of the Canadian case against Timothy Borneyk, reported by Wilkinson et al. [76], is a good example of the impact of a report such as the PCAST report in the courtroom. To meet the recommendations of PCAST for the introduction of pattern evidence testimony, Wilkinson introduced basic information on the structure and results of the two black box studies of fingerprint comparison accuracy that PCAST used to establish the foundational validity of the discipline. In addition to proving an overview of the studies, it was necessary to include discussions of both the calculation error that was made by Miami-Dade (and overlooked by PCAST) in their reporting of the false positive error rate, and a discussion of the appropriateness of PCAST's choice to report error rates as 95% one-sided confidence intervals and the language used to discuss such. PCAST arguments are appearing more frequently in

court and examiners should be fluent in these discussions. Details of the Miami-Dade calculation error can be found in Appendix 2 of [72].

#### 2.4. Quality assurance issues

Proficiency tests (PT) allow the assessment of the state of a discipline. Results from Collaborative Testing Services (CTS) have been subject to highly sophisticated statistical modelling using Rasch models [77]. Mitchell and Garrett [78] have shown the impact on American adult mock jurors of the disclosure of the error rates associated with fingerprint evidence. As somewhat expected, the provision of error rates associated with PT had an impact on the weight assigned to the evidence. Participants gave greater weight to fingerprint testimony from the more proficient fingerprint examiners. These results speak in favor of greater transparency in fingerprint testimonies, highlighting potential limitations as well. The ENFSI fingerprint working group has published the results of their annual testing programs [79–81]. For the 2015 test [79] for example, this group reported a false positive rate of 1% and a false negative rate of 4.4%. In line with Koehler [82], we cannot insist enough on the need to implement a mandatory proficiency testing program carried out under casework conditions with cases that vary in difficulty. Koertner and Swofford [83] have investigated how well the quality of latent print proficiency test samples represent those encountered during routine casework using objective quality measures (LQmetrics associated with the ULW software). The results indicate that the marks used in commercial proficiency tests are generally higher quality and less complex, compared to actual recovered marks from scenes and do not represent the quality levels observed in routine casework.

We are indebted to Cole and Scheck [84] for a complete analysis of fingerprint errors in U.S. cases with a matrix defining each error type. This allows us to realise that fingerprint evidence may be in part at least the cause of miscarriages of justice such as the case of Mr. Dandridge, convicted of murder in 1996 in Alabama that we reported upon in our last report.

Technical review is a good procedure to detect errors before being reported out by a laboratory. A compilation of the technical reviews from 3599 cases (between 2012 and 2015) allowed the detection of 90 cases with significant errors [85]. Although ground truth was not known, there were differences in the opinion of examiners on 14 identifications and 9 exclusions in the verification phase. All cases resulted in an inconclusive conclusion after the conflict resolution process. If all of these 14 differing opinions on identification were truly false positives, the false positive rate would have been 0.52%.

The verification stage and the resolution of conflicting opinions are important parts of a quality management system. It is fair to say that the landscape of conflict resolution procedures is diverse and rather unknown [86]. In Ref. [87], the review of two years of casework at the Houston Forensic Science Center (HFSC) have shown that important changes of conclusions may occur following consultation. Cases with differing conclusions represented roughly 7% of all cases. Verification regimes may vary between agencies and can take various forms: 100% verification, blind verification, identification verification, and suspect only verification. It is important that agencies decide which verification regime fits their activities [88].

How to estimate error rates (typically the false positive rate) has been discussed by Ausdemore et al. [89] in the context of the study carried out in 2014 by the Miami Dade Police. The paper is complemented by a series of commentaries by leading authorities.

Introducing a statistical model (such as FRStat) as an additional tool offered to fingerprint experts to form their conclusion is

bringing some new and interesting challenges in terms of quality assurance. How to resolve conflicting opinions between model and expert judgement is a challenging area for the future [90].

#### 2.5. Experts' performance and expertise

White box studies allow us to gain an understanding regarding the features and processes used by fingerprint experts while undertaking fingerprint examinations. Ulery et al. [91,92] added to their previously published results and showed how examiners may vary in their markup in both the analysis and comparison stages. The variability is mainly due to the clarity of the images. On high clarity images, a larger consensus is achieved compared to low quality images (or areas) where larger inter-examiner variations have been observed. For a global presentation and discussion of these studies, the reader can refer to Hicklin's PhD thesis [93]. Ulery et al. [94] identified factors that may lead to false exclusions. They are the quality of the latent, the value determination, the minutia count in analysis, the perceived comparison difficulty, and the presence of cores or deltas. Errors were typically made under the following circumstances (quoted directly from Ref. [94]):

- Misinterpreted pattern class due to distortion, inadequate overlap, or insufficient area (indicated by examiners citing pattern class differences, or core or delta differences);
- Incorrect anchoring (“corresponding” minutiae in the wrong regions, or incorrectly rotated images);
- Incorrect ridge counting or misinterpretation of distortion resulting in false “discrepancies” (only portions of the image have markup in agreement with other examiners); or
- Inappropriate use of the “one discrepancy” rule (exclusions made despite high numbers of corresponding minutiae, e.g., nine or more).

Gaze behaviour, measured by eye-tracking devices, assisted in further understanding how examiners are conducting comparisons. For the “find the target” task, research has shown [95] using 675 trials conducted by 117 participants that the presence or absence of context (i.e. mark presented as a whole in its context or cropped to a small target area, hence out of context) notably affected the areas viewed and time spent in comparison. It confirms that the comparison process is a holistic process by which objects are perceived as a whole rather than a compilation of individual features [96].

Research experiments aiming at comparing fingerprint experts and novices have been pursued [97–100]. These showed that experts outperformed novices on recognition tasks. These results were somewhat expected (or hoped). However, they have shown that fingerprint perceptual expertise did not generalize to an unfamiliar class of stimuli [99]. This means that just because an examiner is efficient at fingerprint recognition does not mean that this ability will automatically transfer to other forms of recognition such as face recognition. By examining the performance of trainee examiners over their first 12 months, it has been shown [97] that their accuracy improved considerably within the first three months, then plateaued after this time. In our opinion, this is evidence to support much shorter and more targeted training schemes than the multiple years before being formally signed off practiced in many agencies.

It is fair to say that the selection of personnel for pattern recognition tasks such as fingerprint recognition will require special attention in the future. Much has to be gained from collaboration with psychologists and borrowing from other disciplines. The excellent proceeding of a workshop sponsored by the National Academies of Sciences, Engineering, and Medicine helps to realise

how multidisciplinary and complex the problem is [101].

The use of scars in the fingerprint identification process never receives a lot of attention in the forensic literature. Schreel et al. [102] gave a reminder of wound healing and scar formation. They also surveyed 29 examiners to identify how they use scars in practice. All of the participants would give weight to scarred features under high-clarity conditions. The weight to be assigned was dependent on the clarity and complexity of the features of the scar. The examiners also noted that the features were easily recognized and designated as scars on a mark when the corresponding print is also available. This type of circular reasoning is unfortunately not at play only for scar features.

## 2.6. Contextual and cognitive bias

Cognitive and other forms of bias still received a lot of attention during the review period [103–113]. Specifically with regards to fingerprint evidence, Stevenage and Bennett [114] presented a solid state of the art complemented by a study on 48 participants conducting 72 trials (36 “matching” and 36 “non-matching”) with and without time pressure. Participants were not fingerprint experts and received a brief training under the guidance of fingerprint experts. They conducted analyses and comparisons under the influence of three types of contextual information in the form of additional DNA evidence that may be consistent, neutral, or misleading with regards to the fingerprint evidence at hand. The results showed a clear demonstration of cognitive bias when participants were aware of accompanying DNA results even without time pressure. Gardner et al. [115] showed that a lot of agencies are cascading down to their examiners task-irrelevant information that has a high potential to bias, such as type of offense or the name of the suspect or victim. They invite the latent print community to design processes to avoid examiners being exposed to such information.

In their overview focused on fingerprint evidence, Nawrocka & Kiejnisch suggested three realistic measures: (1) train fingerprint experts on cognitive bias; (2) limit contact between experts and investigators in charge of the inquiry; and (3) limit the information provided to task-relevant information [116]. Bunter adds another solution: adopting a “linear” approach to the ACE-V examination process [117,118], i.e. a documented analysis of the questioned mark before unmasking the prints.

## 2.7. Permanence and persistence of friction ridge skin

We take for granted the aspects of persistence associated with the friction ridge skin pattern. As long as the dermis was not altered (e.g. by scarring), it is generally accepted that the structure of friction ridge skin is persistent. An important and welcome study by Monson et al. [119] brings new empirical knowledge to the topic. The authors distinguished the permanence of friction ridge skin when referring to the observations of the skin itself from the persistence when looking at prints left by friction ridge skin. They summarized their main observations as follows (from the abstract):

*“Within all the periods of observation, level 1 detail was permanent and persistent. Persistence, but not permanence, was supported for level 2 detail. Notably, the small changes observed were only in appearance; there were no changes in the presence of new, or absence of existing, minutiae. Level 3 details of ridge edge shape and pore presence were neither permanent nor persistent. Ridge width was permanent and persistent. Incipient ridges were neither permanent nor persistent. The authors are pointing out the need of care when considering level 3 details as their permanence and persistence is not established.”*

Differences in ridge densities between male and female prints tend to diminish with time [120] and age as well [14].

Drahansky and his team provided an extensive collection of fingerprints from individuals suffering from skin diseases [121] and described how the resulting appearance represents a challenge for biometric acquisition and matching [122–124]. The known adverse effect of chemotherapy on friction ridge skin has been further documented [125,126]. Adermatoglyphia will impact modern societies where biometric techniques based on fingerprints play a larger role [127].

## 2.8. Fingerprint matching, biometry and presentation attacks

Covering the literature in relation to automatic processing of fingerprint images in the context of AFIS (ABIS) systems is beyond the scope of this review. The chapter by Maltoni et al. [128] sets the scene regarding AFIS systems and their use in forensic science.

We came across a selection of papers that we cite here as a starting point on the following themes:

- Measurement of image quality using crowd-based learning [129–131].
- Image quality and its impact on the accuracy of matchers [132].
- Latent print matching using minutiae [133], sweat pores [134], pores in conjunction with ridge skeleton [135], extended minutiae types such as enclosures and crossings [136], improving on the minutiae matching algorithms [137], dealing with overlapping marks [138] or taking advantage of SIFT [139,140] or deep learning techniques [141,142].
- A review of the minutiae-encoding systems for palm prints [143].
- The effect of distortion on AFIS fingerprint matching [144] and the development of a distortion detection and rectification algorithm [145]. Distortion is such an important perturbation factor when conducting fingerprint matching either manually or algorithmically but paradoxically, we noted only one publication [146] on the subject in the forensic literature.
- Methods to compute from a query image an expectation of a match against a database and also an expert’s assessment of suitability [147].
- image processing techniques applied to low quality images [148,149], including the use of deep convolutional neural networks [150].
- Use of deep learning techniques to classify the general pattern of fingerprints [151].
- The use of deep learning techniques for minutiae extraction [152,153].
- Improvement on the computation of orientation fields of fingerprint patterns including marks on complex backgrounds [154–156] and palmar impressions [157].
- Automatic segmentation of fingermark images against complex backgrounds [158–161], including overlapping marks [162] or using deep learning techniques such as convolutional neural networks [163]. For a review of segmentation methods, refer to Ref. [164].
- Fusion of data from multiple sensors [165], the matching [166] of subsurface fingerprint images acquired using optical coherence tomography [167].

We note the work by Guan et al. [168] who showed that latent fingerprint image preprocessing (such as colour and greyscale adjustments) results in a statistically significant increase in fingerprint information and quality.

One area where we felt it was important to report to the forensic

community is linked with the risk associated with the presentation attacks of biometric devices (typically liveness devices) using spoofed fingers [169–171]. Not only spoofing materials have progressed, but also the arrival of 3D printers offered new avenues to produce fake fingers that could be used on biometric sensors [172]. Also there are recent advances in solvent-assisted molding of plastics that allows the creation of high quality 3D replicas of fingerprints [171]. We note the technological advances to mitigate presentation attacks [173,174] and the improvements observed at each Fingerprint Liveness Detection Competition [169]. Dedicated protocols to test biometric presentation attack detection solutions are now available [175]. The ability of examiners to distinguish between true latent marks and fabricated marks (obtained using a lift from a ten-print card) or marks produced with forged prints (mimicked with a stamp or a cast) have been shown to be limited [176].

AFIS systems are expected to take a larger part in the establishment of identities in our modern societies, not only in law enforcement or migration contexts, but also to allow the appropriate provision of state or health services. Krzeminska [177] showed how AFIS developed over the years in Poland but also the development within the European Union to support the management of migration, asylum and visa entry and exit systems. Joint Research (JRC) Center of the European Union has published an excellent study showing that the AFIS technology associated with marks (or latent prints) and palmar impressions is ready in terms of accuracy, availability and interoperability to be added in the next Schengen Information System (SIS) [178]. Dealing with young individuals has always been a challenge for AFIS systems, especially when there is an age difference between the enrollment and the identification transaction of interest. The JRC team showed that from an age of 13 years old there is a matching score loss of around 1.5%–3% for each year between the two collected samples [179]. They also showed that a linear isotropic growth model (from the fingerprint core towards its periphery) can handle the issue of fingerprint template aging [180,181]. These efforts have led the European Commission to issue a proposal for amending the regulation by either lower the minimum fingerprinting age requirement for children from the current requirement of 12 to 6 years, or to removing the fingerprinting age limit to include all ages.

We were also particularly impressed with the work by Jain et al. [182,183] on adapting sensors and matching algorithms to allow the identification of very young infants. The contexts for application include child tracking, vaccination campaigns, missing children, or newborn swaps.

### 3. Fingerprint composition and detection

#### 3.1. Fingerprint composition and evolution with time

**Preliminary/Pilot studies:** The articles below refer to preliminary/pilot studies dedicated to the analysis of fingerprint composition. Given that they refer to unconventional approaches or are based on limited sets of fingerprints, caution should be taken with regards to some expressed conclusions.

**Donor profiling** – The following studies aim at providing additional information from a fingerprint, other than the ridge pattern: a new biometric identification tool built on an amino acid-based chemical assay [184]; donor-related information (i.e., gender, ethnicity, and donor age) obtained from secretion residue lipid profiling (technique: DESI-MSI) [185]; impact of gender and ethnicity on the lipid composition of residues present on an individual's fingertips (technique: HPLC-ACPI-MS) [186]; donor gender determination using an amino acid-based chemical assay [187] or by specifically targeting the

chromosomes X and Y contained in nucleated cells (technique: fluorescent in situ hybridization) [188]; donor characteristics and behavioural information (e.g., gender, ethnicity, diet, occupational activities, use of hand sanitizers) gained from bacterial profiling [189]; impact of donors and secretion types (i.e., eccrine, sebum-rich, and natural) on secretion residue composition (technique: MALDI-MSI-based metabolomics approach combined with chemometrics tools) [190].

**Evolution of secretion residue with time** – Surface adhesion monitoring and topography variation (technique: PeakForce QNM AFM) [191]; molecular composition variation (e.g., carotenoids, squalene, unsaturated fatty acids and proteins – technique: Raman spectroscopy) [192]; migration imaging of endogenous fatty acids contained in sebum-rich fingerprints (technique: hyperspectral SRS) [193]; topological modifications (e.g., decrease of ridge height from 200 nm to 100 nm over three days – technique: AFM) [194]; thermal degradation of sebum-rich fingerprints (technique: FTIR microspectroscopy) [195]; intermolecular interactions between lipids (technique: FTIR microspectroscopy) [196], physical modifications of fingerprints left on metallic substrates (technique: EIS) [197], secretion residue composition variation (technique: SALDI-MS combined with MCF) [198] – caution: the last approach requires the dusting of nano-sized MCF (See section 3.2.6 for details).

**Age determination** – Flatbed scanner combined with feature extraction was proposed to distinguish eccrine-rich and sebum-rich secretion residues as well as to estimate their age (i.e., 2-hour-old, half-a-day-old and one-day-old) [199]. The differentiated diffusion of two classes of lipids (i.e., fatty acids and triglycerides) was proposed to estimate the age of fingerprints [200]. Unfortunately, the authors realized the strong influence of the underlying substrate on the diffusion rates, making the establishment of an age determination model a task more complex than expected. The observation of ridge discontinuities [201], of ridge height modifications [202] and of ridge width modifications [203] were proposed for age determination of fingerprints. In the latest article, the authors proposed to use ridge widths measured on inked prints as an age reference (i.e., similar to fresh marks). The authors of these last three articles somewhat recommended to take their conclusions with caution.

**Water content:** Two articles dispelled the myth that fingerprints contain about 98% water at the time of their deposition [204,205] – see details below.

**Emulsion chemistry:** Synchrotron-based ATR-FTIR-FPA combined with confocal Raman microscopy was used to provide information about the spatial distribution of eccrine and sebaceous material in different kinds of secretion residues (i.e., natural, eccrine-rich and sebum-rich) [206] – see details below. The amount of squalene in fingerprints (obtained by GC-MS) was used as a marker of homogeneity when comparing uncontrolled and controlled deposition protocols [207].

**Lipid composition and aging:** The compositional changes of the lipid fraction of sebum-rich fingerprints left on paper was monitored using GC-MS [208]. By observing the relative composition of 15 lipids over 28 days, the authors confirmed previously-established trends (e.g., squalene degradation, intra- and inter-variability) and emphasized the impact of storage conditions, the dynamic of degradation, as well as the persistence of free fatty acids and wax esters over time. GC-MS combined with MSTFA derivatization was used to monitor the degradation of unsaturated fatty acids contained in sebum-rich fingerprints left on aluminium [209]. By considering an aging time up to 14 days, the authors identified decanal as the main degradation product, emphasized the role of storage conditions, and validated the use of derivatization to detect unsaturated

fatty acids and their degradation products (e.g., oxoacids as aging markers). The impact of solar irradiation was investigated by monitoring the “squalene: pentadecanoic acid” ratio over time, using GC-MS/MS [210]. LC-MS was used to monitor the degradation of unsaturated triglycerides into lipid mono-ozonides through an ozonolysis mechanism [211]. The glycerides content of fresh sebum-rich fingerprints left on filter paper was analysed by UPLC-IMS-QToF-MS following a lipidomics-based approach [212], allowing the identification of approximately 100 di- and triglycerides.

**Protein composition and aging:** Proteomics was proposed to study the impact of time on the proteins contained in secretion residue [213]. Using fingerprints left on glass and analysed after 4–16 days, the LC-MS-based method allowed recording a proteome for each fingerprint and emphasized the presence of cytokeratins (dominant species), antibiotic proteins and secreted blood proteins. The authors also investigated the impact of time over the 31 proteins that were identified in all samples and emphasized a significant effect for five of them (i.e., K2C1, K22E, K1C9, K1C10 keratins and dermcidin). The authors suggested that these proteins could be used as new aging markers.

**Secretion residue migration:** The evolution of fingerprint morphology with time was monitored by AFM [214]. Using silicon wafers and Formica, the authors illustrated how secretion residue may horizontally migrate (4 nm thick film) towards the inter-ridge area over a few microns shortly after deposition, before starting to disrupt after about one week. The impact of such migration on the detection techniques as well as on the secretion residue behaviour on other substrates are yet to be determined.

**Impact of time on detection performance:** Boudreault and Beaudoin explored how the effectiveness of conventional detection techniques may be impacted by the aging of fingerprints [215] – see details below.

**Donor profiling:** The analysis of fingerprints left on mobile phones (using a UPLC-Q-ToF MS/MS workflow) provided information about the chemicals that can be found in fingerprints and hence on donors' hands, including: cosmetics, medications, pesticides/insecticides, or diet metabolites [216]. Lifestyle inference was also proposed by combining the molecules present through a metabolomic approach. In a similar way, MALDI-MSI was proposed to identify exogenous compounds from fingerprints and hence infer information about the donor's lifestyle [217]. Fingertips were spiked by contact with various substances (e.g., bug sprays, sunscreens, cooking oils, alcohols, citrus fruits) before leaving fingerprints on glass slides. Using PCA and a targeted approach, the authors proposed to distinguish product brands from fingerprint analysis. It should be noted that these two studies require the identification of lifestyle markers (e.g., active molecules) and the creation of an exogenous compound database generated from an exhaustive range of products before being considered for casework. Amino acid profiling of 19 donors (fingerprints left on aluminium) was proposed using UPLC-ToF MS and UPLC-QqQ MS/MS [218]. From an analytical point of view, the authors determined the LOD and LOQ for all amino acids and emphasized the advantage of using an amide stationary phase to avoid a derivatization step. From a donorship point of view, the authors charted the total amounts of amino acid per fingerprint (in ng) as well as the amino acid distribution for each donor (amino acid profiling). In follow-up study, the use of hydrogels from dextran-methacrylate solutions was proposed to collect hydrophilic compounds (e.g., amino acids or DNA) from a fingerprint [219]. After removal from the surface, the hydrogels were used for extraction and

quantification (UPLC-MS) of amino acids. The authors also showed that the surface can still be processed with fingerprint detection techniques (they tested CA) to detect ridge patterns, although a slight degradation of fine details was observed.

**Reviews linked to fingerprint composition:** AFM applied to traces of forensic interest, including fingerprints [220], analytical techniques aiming at exploiting the chemical composition of fingerprints [221], extensive review covering fingerprint composition variability and alternatives in terms of artificial secretions [222].

**Acronyms used:** **AFM** (atomic force microscopy), **APCI** (atmospheric pressure chemical ionisation), **ATR** (attenuated total reflectance), **BY40** (Basic Yellow 40), **CA** (cyanoacrylate), **CV** (crystal violet), **DESI** (desorption electrospray ionisation), **EIS** (electrochemical impedance spectroscopy), **FPA** (focal plane array), **FTIR** (Fourier-transform infrared), **GC** (gas chromatography), **HPLC** (high-pressure liquid chromatography), **IMS** (ion mobility spectroscopy), **IND/Zn** (1,2-indanedione containing zinc chloride), **K1C9** (type I cytoskeletal 9), **K1C10** (type I cytoskeletal 10), **K22E** (type II cytoskeletal 2 epidermal), **K2C1** (type II cytoskeletal 1), **LC** (liquid chromatography), **LOD** (limit of detection), **LOQ** (limit of quantification), **MS** (mass spectrometry), **MS/MS** (tandem mass spectrometry), **MSI** (mass spectrometry combined with imaging), **MSTFA** (N-methyl-N-trimethylsilyltrifluoroacetamide), **NIN** (ninhydrin), **ORO** (Oil Red O), **PD** (physical developer), **QCM** (quartz crystal microbalance), **QNM** (quantitative nanomechanical mapping), **QqQ** (triple quadrupole), **QToF** (quadrupole time-of-flight), **R6G** (Rhodamine 6G), **SRS** (stimulated Raman scattering), **TPD** (temperature-programmed desorption), **UPLC** (ultra-performance liquid chromatography), **Q-ToF** (quadrupole time-of-flight)

**Water content** – The erroneous claim that the water content of a fingerprint is close to 98% is most likely due to an inference from eccrine sweat composition. In his article, Kent used published analytical data, theoretical models and common sense to build his argument [204]. Considering fingertip contamination due to daily activities (e.g., sebum, cosmetics, food), the water content of fingerprints cannot be inferred from sweat only. Moreover, there exists no evidence that a purely eccrine fingerprint would contain approximately 98% water, due to evaporation on the skin and transfer mechanisms upon contact. Based on theoretical considerations and analytical data, Kent estimated that water content of a natural fingerprint would be closer to 20% or even less. In another study, Keisar et al. addressed the question of the water content in freshly-deposited fingerprints by combining QCM and TPD-MS to measure mass loss upon drying [205]. Their methodology included a stepwise hand-washing procedure, the deposition of eccrine-rich fingerprints (sweat), and the monitoring of mass loss due to substrate heating (ca. 40°C). Using QCM, mass loss ranging from 20 to 70 wt% was observed, mostly within the first minutes after deposition. These values exceeded those predicted by Kent [204], but they confirmed that the water content of a fingerprint is far from being close to 98 wt%. The influence of the donor and the presence/absence of a hand-washing procedure were shown to have a great influence in the variability of results. Eccrine-rich and natural secretions were associated with water loss values ranging from 40 to 70 wt-% and from 20 to 60 wt%, respectively. The lowest values associated with natural fingerprints are due to the lowest content of water in the secretion residue, mostly caused by the presence of compounds other than sweat (e.g., sebum or exogenous components).

**Emulsion chemistry** – Using synchrotron-based ATR-FTIR-FPA combined with confocal Raman microscopy, Dorakumbura et al.

aimed at providing fundamental knowledge about the spatial distribution of eccrine and sebaceous material in fingerprints [206]. Nine donors were asked to leave natural, eccrine-rich, and sebum-rich fingerprints on substrates adapted to the analytical techniques (i.e., zinc selenide and calcium fluoride slides for FTIR, and glass slides for Raman). The fingerprints were readily analysed and imaged (<5 h aging time). The differentiated imaging of eccrine and sebaceous material allowed emphasizing the overall water-in-oil structure of the natural and sebum-rich emulsions, with localized areas of eccrine material embedded in a bulk of sebaceous material. Quite interestingly, the presence of (sub-)micron droplets of lipids in eccrine-rich fingerprints was emphasized, which can be associated with localized oil-in-water emulsion. The origin of lipids in eccrine-rich fingerprints is still to be confirmed, but the authors hypothesised that an incomplete hand-washing procedure may be the cause of their presence.

**Impact of time on detection performance** – To assess the impact of the passage of time on the performance of detection techniques (due to water loss or to chemical and physical modifications), Boudreault and Beaudoin set up a pseudo-operational study involving various substrates (i.e., office white paper, recycled paper envelopes, transparent and white plastic bags, aluminium foil, duct tape, thermal paper receipts from restaurants, grocery stores, or gas-stations), 200 participants leaving fingerprints without having received restrictive instructions, four aging times (i.e., 1 day, 1 week, 1 month, and 11 weeks), and eight detection processes (i.e., IND/Zn, NIN, IND/Zn  $\rightarrow$  NIN, ORO, PD, CA  $\rightarrow$  R6G, CA  $\rightarrow$  BY40, CV) [215]. It should be noted that all the samples were protected from light, environmental elements and dust during their aging, as a way to mimic their storage as evidence before being processed. The authors concluded that ORO (all porous substrates) and IND/Zn (white and thermal papers) showed a significant decrease in quality over time (See the *Note* below about IND/Zn). On recycled paper, ORO failed to detect 11-week-old marks. Overall, NIN (alone or in sequence with IND/Zn), PD, CA  $\rightarrow$  R6G/BY40, and CV showed no significant difference in fingerprint quality over time. R6G and BY40 were slightly superior to each other on black and transparent plastic bags, respectively.

Note: the use of half-marks was considered to compare two techniques. However, distinct fingerprints were used to compare two aging times (not half-marks), which may influence the conclusions regarding the evolution with time. For example, on thermal papers, the scores associated with IND/Zn (alone) showed a significant decrease of quality after 24 h, whereas other scores associated with IND/Zn (as the first step of the IND/Zn  $\rightarrow$  NIN sequence) showed no significant decrease of scores up to 11 weeks. A significant increase of quality was even observed for 1-month-old marks. Similarly, a significant increase of quality was observed with 1-month-old marks (aluminium) processed with CA  $\rightarrow$  R6G/BY40. The authors failed to provide an explanation for these discrepancies.

### 3.2. Fingerprint detection and imaging/recording

**Preliminary remarks** – For ease of reading, 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**).

Also, the articles referring to unconventional approaches or based on limited sets of fingerprints were characterized as *Preliminary/pilot studies*. Follow-up studies are required or expected, and caution should therefore be taken with some of the expressed conclusions.

#### 3.2.1. Research interests overview

From August 2016 to June 2019 (incl.), 365 articles were published in relation to fingerprint detection and imaging/recording. This represents an increase of approximately 50% compared to the 2013–2016 report. To get a better sense of the research interests represented, the published articles were classified according to their main research scope(s) (Fig. 2).

Surprisingly, the main topic of interest for these last three years has been *Powder dusting* (129 articles, which represent 35% of all the articles dealing with fingerprint detection and imaging/recording). This is especially surprising as there is no expressed need from practitioners for new dry powders that could explain such an urge in this field. When compared to the 2013–2016 report, the sudden rise of interest for *Powder dusting* appears more evidently, with an absolute difference of +94 articles (Fig. 3). More concerning: among those 129 articles, **97 articles promote nanoparticle powdering** (75% of the articles related to dry dusting and **27% of the articles related to fingerprint detection and imaging/recording**). In the 2013–2016 report, “only” 20 articles were associated with the dusting of dried nanoparticles (less than 10% of all the articles related to fingerprint detection). Such evolution goes against all ethical considerations for practitioners in terms of health and safety issues, and against any scientific strategy dedicated to fingerprint detection. Indeed, for most of those articles, fingerprint detection capability appears as a pretext for the synthesis and characterization of optically-active nanomaterials. When looking at the origin of these articles, it appears that most of them come from a very limited number of research units. One research group (comprising a single individual) is even associated with approximately 40 articles – almost half of the articles promoting the dusting of nanoparticles. This publication rate and the message it carries should raise concerns from the scientific and forensic communities. For these reasons, and similar to the 2013–2016 report, the articles promoting this practice will only be cited in Section 3.2.6 but not further described.

Besides dry powdering, the other topics of interest are *Nanoparticles in suspension* (36 articles/+11 articles compared to 2013–2016), *Dusting of micro-sized particles* (32 art./+17 art.), *Chemical imaging* (28 art./stable) and *Contaminated fingerprints* (25 art./–4 art.). Similar observations were made in the 2013–2016 report. The trends related to security matters (explosive- and drug-contaminated fingerprints) and the technical specialization linked to fingerprint detection and imaging/recording are thus confirmed. Unfortunately, this technological leap keeps suffering from the absence of follow-up studies (several “one-shot” papers), from the need for overspecialized equipment requiring specific abilities, and from a failure to account for forensic considerations such as the absence of integration into operational procedures. As opposed to these top-rated topics, the historical techniques and conventional substrates were characterized by a lower number of articles and by a mitigated evolution. Most of them receded or remained stable: *Amino acid reagents* (12 art./–1 art.), *Cyanoacrylate fuming* (10 art./–5 art.), *Powder suspension* (4 art./stable), *Lipid stains* (3 art./–2 art.), *Adhesives and tapes* (3 art./stable). Among the topics that progressed, it is possible to cite the detection of fingerprints on *Metal/cartridge cases* (17 art./+5 art.) or on *Banknotes* (6 art./+4 art.), as well as *Blood-containing fingerprints* (20 art./+5 art.) and *Physical developer* (6 art./+3 art.). One explanation could be that research efforts are now focused on other detection techniques or on other fields related to forensic science, such as digital traces. Such an evolution of the research scopes somewhat raises the question of the actual needs of the practitioners (*aka* “stakeholders”).

To try answering this question, each main research scope was further characterized according to the number of articles published

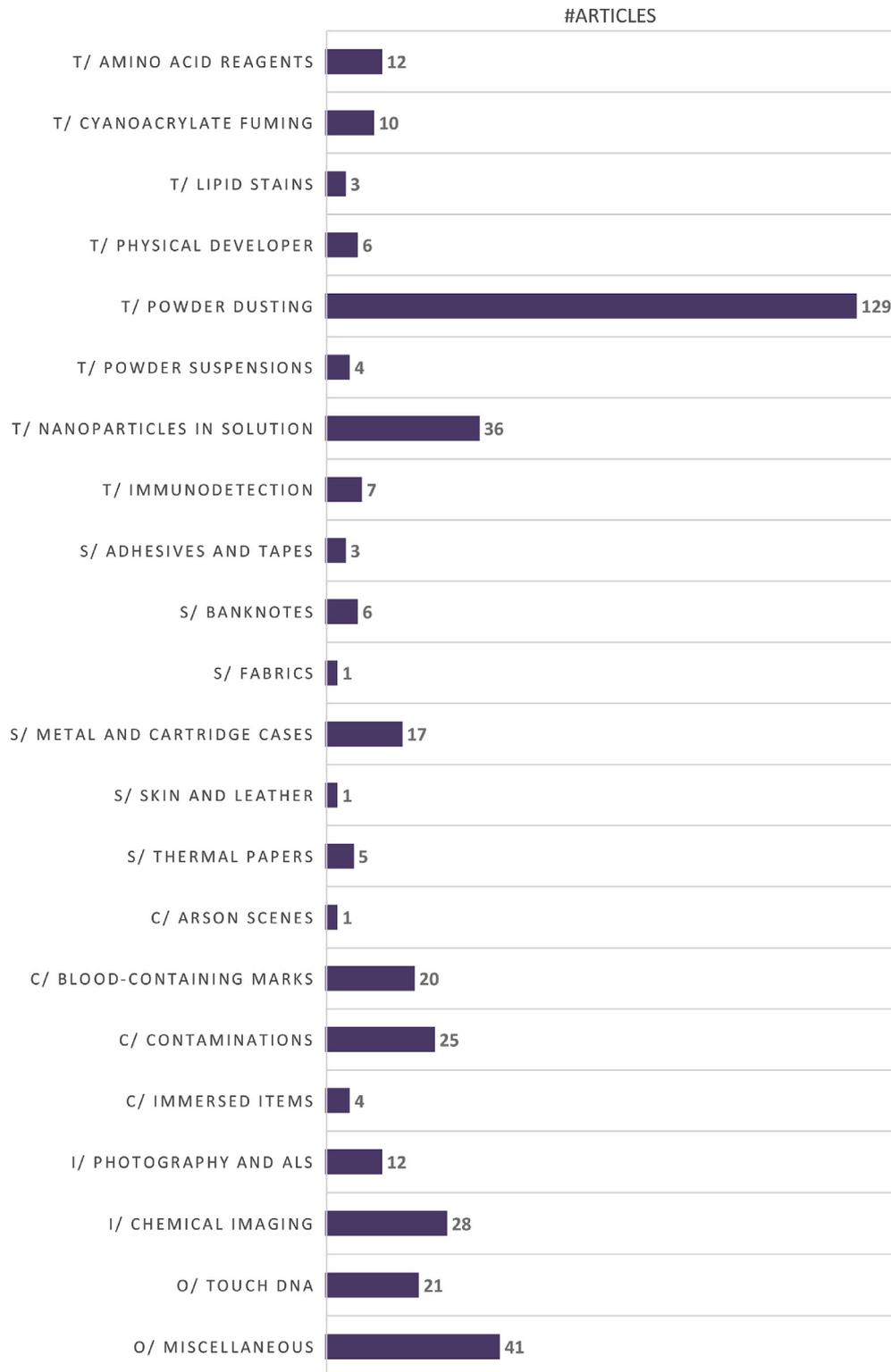


Fig. 2. Distribution of the main research scope(s). [Note: some articles may be associated with more than one scope, explaining why the sum of all the reported values exceeds 365].

in “Forensic” journals and “Non-Forensic” ones (Fig. 4). The underlying postulate is that people willing to reach the forensic practitioners/community with a topic of interest would rather publish their research in a forensic journal. When looking at Fig. 4, it is immediately apparent that most of the articles related to *Dry powdering* (93%) are published in chemistry-oriented journals

(among which are 100% of the articles promoting the dusting of nanoparticles). This tends to confirm that such a topic is not aimed at the forensic community and even less at practitioners. The other topics that are almost exclusively published in chemistry-oriented journals are: *Immunodetection* (86%), *Nanoparticles in suspension* (83%), and *Chemical imaging* (82%). On the contrary, the topics that

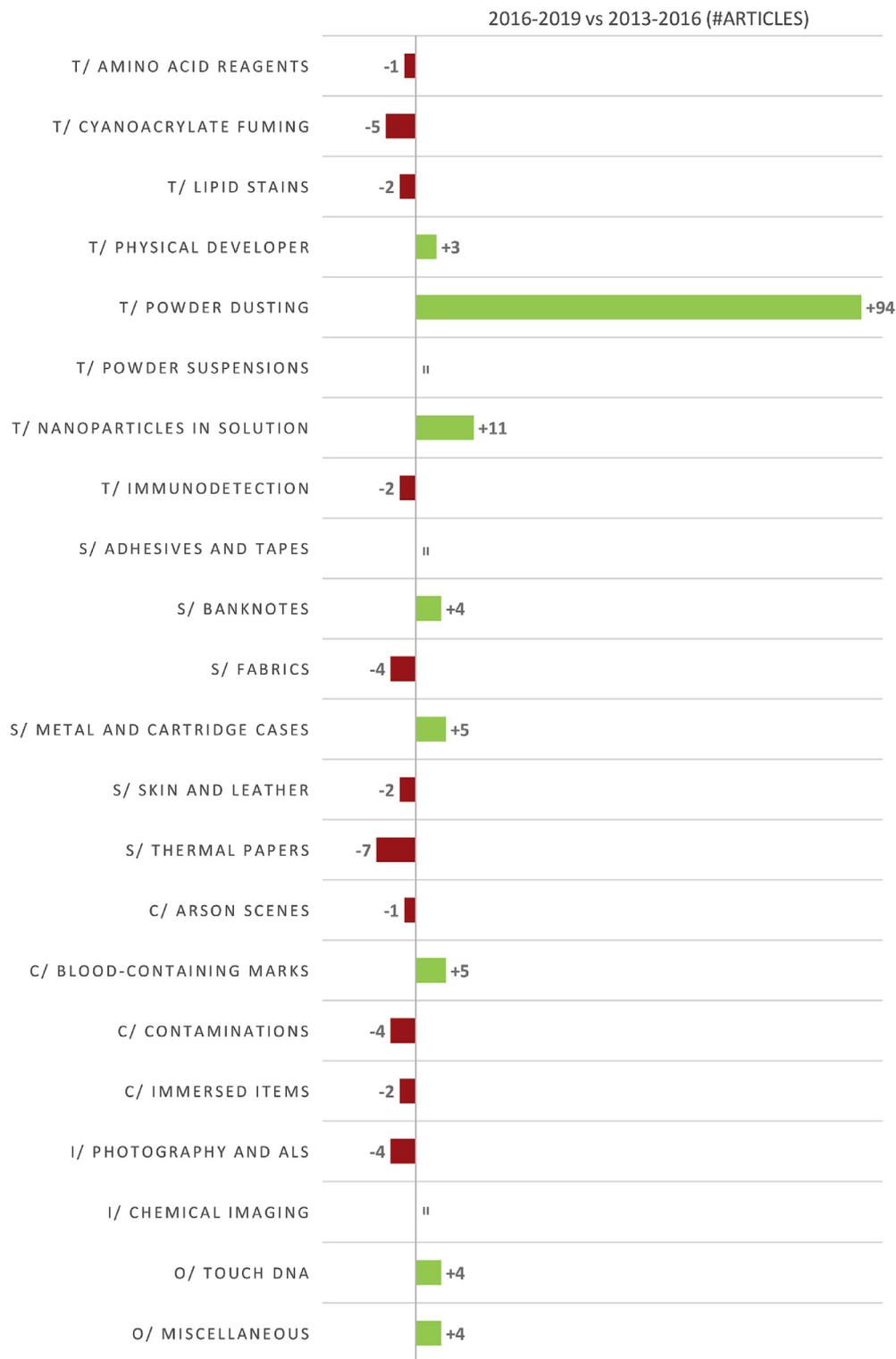
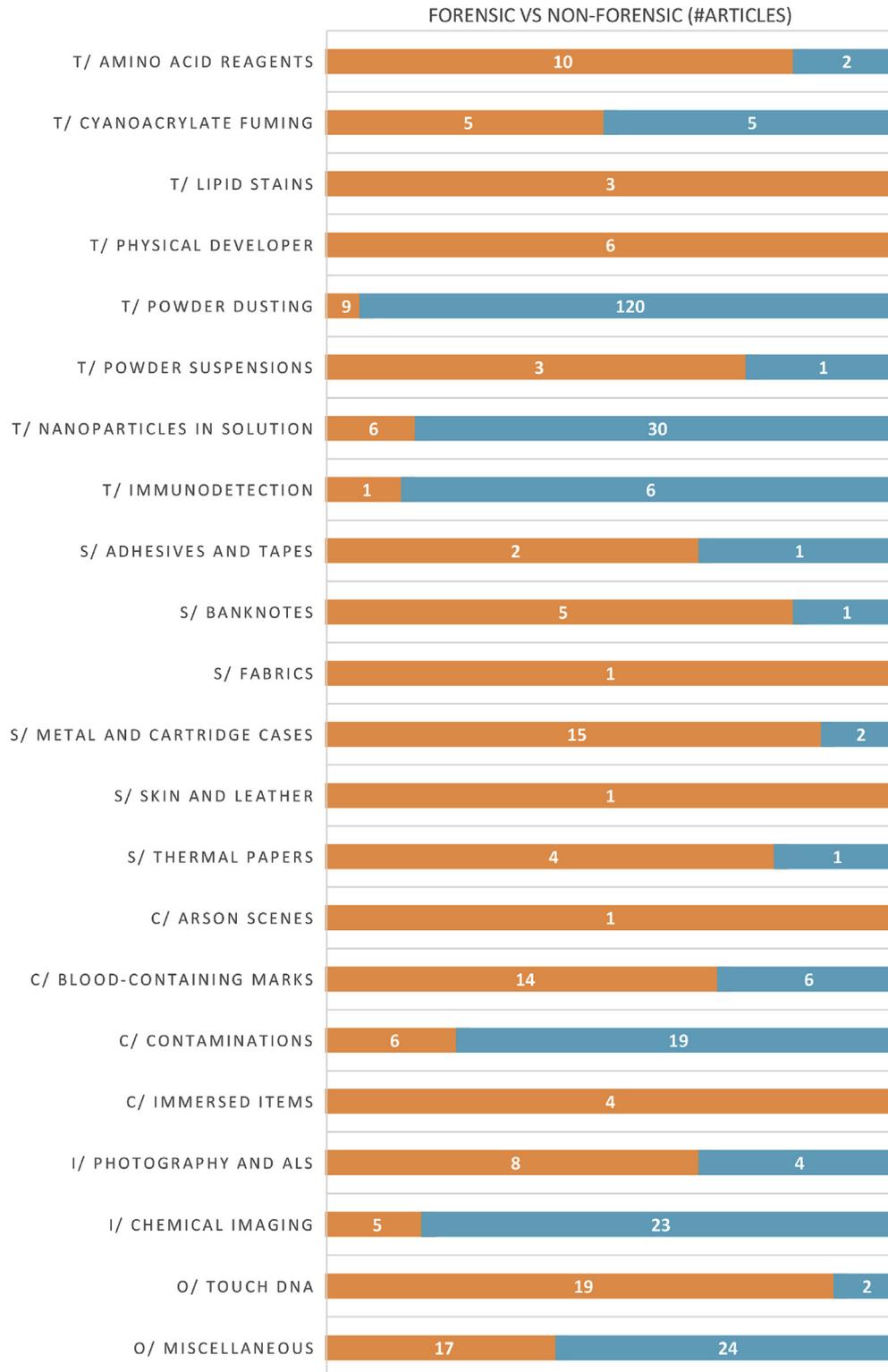


Fig. 3. Absolute differences (in terms of article numbers) associated with each category between the 2013–2016 report [6] and the current review period.

are almost exclusively published in forensic journals are: *Physical developer*, *Lipid stains* and *Immersed items* (100%), *Metal and cartridges* (88%), *Banknotes* (83%), *Amino acid reagents* (83%), and *Thermal papers* (80%). *Fabrics*, *Skin/Leather*, and *Arson scenes* being covered by one article only, they were not added to the list of 100% for clarity reasons. If these topics are not associated with the highest absolute numbers of articles, they are certainly more

representative of the current state of research dedicated to the forensic community and to practitioners in general. They are also addressing historical topics related to fingerprint detection.

**IFRG guidelines** – In the 2013–2016 report, the IFRG guidelines [223] were cited by 29 articles (which represented approximately 10% of the articles covered). In this report, the IFRG Guidelines were cited in 60 articles, which represents 15% of the articles cited in



**Fig. 4.** Proportions of articles published in “Forensic” (orange) and “Non-Forensic” (blue) journals for each main research scope. [Note: some articles may be associated with more than one scope, explaining why the sum of all the reported values exceeds 365]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Sections 3.1 and 3.2. The visibility of these guidelines has consequently doubled in three years. Their influence can be seen in the evolution of the published methodologies, with an increasing consideration of natural fingermarks instead of sebum-rich ones,

for example. If it must be acknowledged that two-thirds of the articles that cite these guidelines are from authors that are part of the IFRC, one-third is from authors who are not, which is encouraging. However, efforts should be maintained to increase the awareness of

such a document within the research community, because a lot of articles are still referring to inadequate methodology with regards to the claimed objectives (e.g., single donor or fresh sebum-rich marks) or overstating conclusions. Also, with the exception of two, none of the articles associated with the dry-dusting of nanoparticles cited these guidelines. With regards to the two that cited them, one is wrongfully citing the IFRG guidelines (referring to the uniqueness of ridge patterns) and the second one is correctly referring to a Phase 1 study.

### 3.2.2. T/Amino acid reagents

**Preliminary/Pilot studies:** Sublimation of genipin under vacuum (1 Pa, 140°C) followed by exposure to heat and humidity (80°C and 80% RH for 1 h) was proposed to detect fingermarks on porous substrates [224]; proof-of-concept results were obtained and compared to genipin and NIN in solution. NIN analogues bearing a coumarin moiety were synthesized [225]; proof-of-concept results were obtained with processed marks absorbing UV. Tropolone has been proposed as a new amino acid reagent [226]. The reaction product is said to absorb UV, resulting in dark ridges on an illuminated background. The authors also showed that post-treatment could be carried out to produce luminescent azo dyes. However, the performance of tropolone was inferior to NIN.

**Advanced/Operational studies:** An operational study was conducted on 1500 used train tickets (Israel), which were processed with either 1,2-IND/Zn, DFO, or NIN [227]. Train tickets being composed of a regular cellulose-based layer and a thermal-sensitive layer, they were left to dry overnight after treatment with IND/Zn or DFO, without the application of heat. Overall, the best performance was obtained with IND/Zn, followed by DFO and NIN.

A study was conducted to assess the ideal time between the processing of a substrate with NIN and its observation or its processing with the next technique in the sequence (e.g., physical developer) [228]. Using marks obtained under controlled conditions, the author reports no additional marks up to 7 days after the application of NIN when heat and humidity is applied (NYPD protocol: 70°C and 65% RH for 20 min). On the contrary, additional ridge details and less background staining were observed when the items were left at ambient conditions after the application of NIN.

A three-step validation study was published with regards to the use of 1,2-IND in the UK [229–231]. An optimized formulation and a detection protocol were proposed – see details below.

Solstice® Performance Fluid (Honeywell) has been tested as an alternative to HFE7100, whose supply may be at risk due to regulations aimed at hydro-fluorinated solvents [232] – see details below.

The addition of zinc chloride in a DFO formulation showed improved performance and higher luminescence intensity compared to the standard DFO formulation [233]. However, the modified formulation was still outperformed by IND/Zn and it was shown to be less stable (reduced shelf-life), especially when the temperature was below 19°C. It should be noted that the use of other metals was tested (i.e., iron, nickel, and palladium), but led to unsuccessful results compared to the conventional formulation.

Fritz et al. investigated the factors that could influence the performance of IND/Zn [234]. Considering natural secretions left by 131 donors on paper and two aging times (i.e., three days and one month), the authors confirmed their previous observations (See 2013–2016 report) regarding the influence of the donors'

gender and age, the impact of hand washing prior to deposition, and the consistency of the quality grading procedure.

**Case report:** Brazelle et al. reported the observation of the same fingermark throughout both sides of an IND/Zn-processed item [235]. Three cases were described: a spiral notebook, printed counterfeit currency, and a thermal paper receipt. Marks observed throughout a substrate appear to be less intense and sometimes more diffused than the ones observed on the deposition side, with the exception of the thermal paper for which no variance in fluorescence or diffusion was observed. Most importantly, from a dactyloscopic point of view, the ridge pattern is laterally-reversed compared to the actual pattern on the non-deposition side.

**Acronyms used:** **DFO** (1,8-diazafluoren-9-one), **IND** (1,2-indanedione), **IND/Zn** (1,2-indanedione containing zinc chloride), **NIN** (ninhydrin), **RH** (relative humidity)

**IND/Zn validation in UK** – The validation study of IND/Zn in the UK consisted of three successive publications. The first one focused on the formulation (with or without methanol and zinc ion) [229]. The recommended formulation contains both methanol and zinc ions. If the exact role of these two chemicals is still unclear, their presence resulted in the highest luminescence intensity of the detected marks. However, the authors also observed that too much methanol could result in ink running and in ridge diffusion. The second study consisted of optimizing the detection protocol [230]. Dry oven, heat press, and infrared lamp (TFD2, from Foster + Freeman) were compared. The dry oven showed better performance, followed by the heat press and the IR lamps. The authors observed that processing the substrates for 10 min at temperatures above 120°C may result in marks of lower intensity. The recommended range lies between 90°C and 120°C with no humidity. Interestingly, the authors observed that if a sample is processed at a lower temperature (50°C, 30°C, and to a lesser extent 20°C) and stored for at least one day before observing the results, the resulting luminescence can equal the one obtained at 100°C. This could be interesting for application at crime scenes or on substrates that may be damaged by excessive heat. The third study consisted of a comparative study and a pseudo-operational trial aiming at comparing the optimized IND/Zn technique and the previously recommended DFO formulation. 7500 split marks (comparative study) and approximately 860 porous items that were realistically-handled (pseudo-operational trial) were processed either with the sequence IND/Zn ⇌ NIN or with DFO ⇌ NIN. The newly optimized IND/Zn technique outperformed DFO on all sets of samples, in terms of number of recovered marks, brightness and quality. Additional marks were also observed when NIN was applied after IND/Zn or DFO (+18% for IND/Zn and +37% for DFO). To conclude, the recommended “CAST 2014” formulation is the following: 0.25 g IND +45 mL ethyl acetate +45 mL methanol +10 mL acetic acid +1 L HFE7100 + 1 mL zinc stock solution (0.1 g zinc chloride + 4 mL ethyl acetate + 1 mL acetic acid) – Shelf life: one year. The substrates were to be processed in a dry oven (0 %RH) at 100°C for 10 min. This formulation of IND/Zn outperforms the previously recommended DFO. It is also strongly recommended to use NIN in sequence as additional marks were detected (+18% for the sequence IND/Zn ⇌ NIN).

**Solstice as an alternative to HFE7100** [232] – When comparing formulations of IND/Zn and NIN prepared with both carrier solvents in a pseudo-operational trial setup, comparable performances were obtained (less than 4% difference in the number of marks detected). DFO (based on HFE7100 and HFE71DE) showed to be 20% less efficient compared to IND/Zn, confirming the superior performance of the latter. The application of NIN in sequence with IND/Zn resulted in approximately +20% marks (both for HFE7100

and Solstice), supporting the results previously obtained by the CAST. The authors also emphasized the influence of the substrate type (e.g., newspaper, envelopes, magazines) on the performance of all amino acid reagents. A shelf-life of 6 months was estimated for the Solstice-based formulations. Neither Solstice PF nor HFE7100 were shown to cause ink running on their own.

### 3.2.3. T/Cyanoacrylate fuming

**Preliminary/Pilot studies:** Three alternatives to ethyl-2-CA were tested (i.e., methyl, n-butyl and 2-octyl) to detect fingerprints on glass, aluminium and plastic [236]. Overall, it was shown that the substrate, the composition of the secretion residue and the age of the marks do influence the relative performance, that ethyl-CA and butyl-CA resulted in the best performance, and that they both led to the creation of polymer structures presenting light scattering properties. Fluorescent PPV nanoparticles in aqueous solution [237] and a fluorene-based dye taking advantage of AIE [238] were proposed as post-fuming dyes, the latest showing promising results in terms of luminescence. In all these studies, home-made fuming cabinets without humidity control were used.

The advantage of increasing the concentration of Lumicyano powder to 4% (compared to 1%) has been illustrated in terms of luminescence intensity and resistance to fading with time [239]. In another study, Lumicyano 5% was compared to CA ⇨ BY40 (non-porous substrates) and CA ⇨ black powder (semi-porous substrates) [240]. Overall, the conventional processes were preferred, with the exception of leather-like material for which Lumicyano 5% successfully detected fingerprints (as opposed to CA ⇨ black powder, which failed). However, these conclusions should be taken with caution given that only one donor and artificial secretion pads were used in the first study, and two donors providing a limited set of fresh fingerprints (not cut in half) were used in the second one.

**Mechanism:** Chemical imaging was used to image the secretion residue present under a CA polymer layer [241] – see section 3.2.19 for details. MALDI-MS was used to get a better understanding of the CA fuming process [242]. Basing their study on the detection of four compounds linked to the polymerization process, the authors proposed a reaction mechanism. Using spots of chemicals, they also suggested that fatty acids and amino acids act as catalytic nucleophiles initiating the polymerization mechanism. In both studies, home-made fuming cabinets were used.

**Dye staining:** The performance of Coumarin-480 as a post-fumigation dye was compared to R6G [243]. At the completion of a pseudo-operational trial based on 100 HDPE plastic bags (cut in two) and 200 glass bottles, Coumarin-480 showed similar performance on glass and better performance on HDPE bags, compared to R6G.

**One-step CA fuming:** A comparative study was conducted between Lumicyano and the conventional CA ⇨ BY40 sequence [244] – see details below. The impact of one-step CA fuming on subsequent DNA profiling was also assessed by Khuu et al. [245] – see section 3.2.20 for details.

**Case report:** Tapps et al. described how they applied CA fuming followed by R6G on a plastic bag linked to a thirty-year-old cold case [246]. A good quality fingerprint was detected.

**Available reviews:** one-step fluorescent CA [247], CA [248].

**Acronyms used:** **4Z** (chloro-6-ethoxy-1,2,4,5-tetrazine), **AIE** (aggregation-induced emission), **BY40** (Basic Yellow 40), **CA** (cyanoacrylate), **MALDI** (matrix assisted laser desorption ionisation), **MS** (mass spectrometry), **PPV** (poly p-phenylene vinylene), **R6G** (rhodamine 6G)

**Comparative study** – In their study, Risoluti et al. compared a two-step-process (CA ⇨ BY40) with a one-step-process (7.7% Lumicyano) [244]. Fresh (3-day-old) and aged (100-day-old) fingerprints left on plastic (bottles and sheets) were processed with both techniques. The marks were then observed using different optical methods: UV reflection (SceneScope RUVIS), white light and luminescence (Crimescope, exc. 415 nm for BY40 and 515 nm for Lumicyano). Luminescence decay with time (up to 20 days post-fuming), compatibility with a subsequent DNA analysis, and molecular interaction between the polymer and the Lumicyano dye (i.e., 4Z) were also assessed. Lumicyano performed similarly or better than the two-step-process with regards to ridge clarity and contrast. The authors also recommended combining the application of Lumicyano with a dye-staining step (BY40) to offer different optical configurations. This conclusion agrees with those of previously-published articles (See 2013–2016 report). The fluorescence decay limit was set to 6 days for eccrine marks and 20 days for sebum-rich marks. Lumicyano was shown to be compatible with a subsequent DNA analysis (extraction and amplification), but the authors obtained mixed results in terms of profile quality: uninterpretable (mostly obtained with aged marks), clean (mostly obtained with fresh marks), and mixtures. The authors also found no hint of a chemical bond between the fluorophore and the polymer.

### 3.2.4. T/Lipid stains

**New reagent/Comparative study:** PMA was proposed to detect marks on porous and non-porous substrates through staining of the water-insoluble fraction of the secretion residue, which includes sterols, lipids, fatty acids and triglycerides [249] – see details below.

**Advanced/Operational studies:** Fritz et al. investigated the factors that could influence the performance of ORO with regards to PD (Tween 20 formulation) [250]. Considering natural secretions left by 148 donors on paper and two aging times (i.e., three days and one month), the authors confirmed previously-published trends [1]: overall poor performance of ORO [2], better performance of the sequence ORO ⇨ PD compared to ORO alone, and [3] decreasing performance of ORO with older marks. Their study also emphasized the fact that ORO and PD target two different fractions of the secretion residues (i.e., lipid fraction for ORO and non-water-soluble fraction encompassing eccrine components for PD).

**Available reviews:** ORO [251].

**Acronyms used:** **ORO** (Oil Red O), **PD** (physical developer), **PMA** (phosphomolybdic acid)

**Comparative study** – PMA is not a lipid stain *per se*, but it was compared to ORO because the targeted fraction is similar (i.e., water-insoluble fraction vs lipids) [249]. PMA showed performance similar to ORO with fresh marks left on porous substrates, and superior performance with marks older than one week. On the contrary, ORO outperformed PMA on porous items that have been wetted (1-h-long immersion). Performance was somewhat higher on non-porous substrates, such as aluminium, stainless steel, and acetate sheets. According to the authors, this observation is consistent with an absorption of the secretion constituents by the porous substrates. In such cases, amino acid reagents would be preferred. Unwanted background staining was observed on all items, with varying degrees of occurrence. It should be noted that the PMA reaction protocol requires exposing the samples to long-wave UV for 15 min. Further studies are consequently required.

### 3.2.5. T/Physical developer

**Silver nitrate:** With the goal of cost reduction, Coppes et al. compared the efficiency of three PD solutions prepared with different grades of silver nitrate [252]. Overall, silver nitrate of USP grade (99.8%) or of technical grade (purity not specified, cheapest) can replace ACS grade (99.0–99.97%, most expensive) with negligible impact on PD performance.

**Tween 20 formulation:** The longevity of the Tween 20-based PD working solution was assessed [253] – see details below. De la Hunty et al. published a communication article about the use of the newest PD formulation (based on Tween 20 in place of Synperonic N), providing practical recommendations to introduce the technique in a laboratory [254].

**PGME formulation:** an alternative to the Tween 20 formulation was proposed in a paper addressing the processing of several-year-old documents by amino acids reagents and PD [255] – see details below.

**PD vs SMD:** Moret et al. compared SMD II and PD in their ability to detect marks as standalone techniques, in sequence with IND/Zn and NIN, and on wetted documents [256]. Overall, PD resulted in higher performance compared to SMD II, the latter suffering from inconsistencies with regards to the range of porous substrates and from a lower contrast. Moreover, the application of amino acid reagents beforehand negatively impacted SMD II performance, in contrast to PD. Additional observations were made with regards to the fingermark ages, depletion series, donor variability, and substrate types.

**PD vs ORO:** Fritz et al. investigated the factors that could influence the performance of PD (Tween 20 formulation) with regards to ORO [250] – see section 3.2.4 for details.

**Acronyms used:** **ACS** (American Chemical Society), **DFO** (1,8-diazafuoren-9-one), **DGME** (decaethylene glycol monododecyl ether), **IND/Zn** (1,2-indanedione containing zinc chloride), **NIN** (ninhydrin), **ORO** (Oil Red O), **PD** (physical developer), **RH** (relative humidity), **USP** (U.S. Pharmacopeia), **SMD** (single metal deposition)

**PD (Tween 20)** – The longevity of the Tween 20-based PD working solution was assessed by processing items with one-week-old to 16-week-old solutions [253]. Up to 9 weeks, the PD working solution was effective enough to detect fingermarks\*<sup>1</sup> within 20 min of immersion. Older solutions were more prone to staining plastic-based labware (PP and PVC) or to presenting silver metallic beads. In terms of appearance in the storage bottle, freshly prepared PD working solutions were clear and started to develop cloudiness after one day up to two weeks, with presence of a precipitate in solutions older than 2 weeks. As a conclusion, the authors recommended a shelf-life of 2 months for the Tween 20-based PD working solution.

[\*<sup>1</sup>] It should be noted that artificial secretions (Latent Print Standards Pad, Sirchie) freshly deposited on white copy paper were used in this study.

**PD (DGME)** – Bleay et al. assessed the performance of the recommended “porous substrates” sequence (*i.e.*, DFO or IND/Zn  $\rightarrow$  NIN  $\rightarrow$  PD) when applied on several-year-old documents [255]. The four-step study spanned from 2013 to 2018. The authors investigated the effectiveness of (1) the 2013 sequence (incl. DFO and Synperonic N PD) on 11- to 17-year-old cheques and about 60-year-old documents, (2) the 2013 sequence and an alternate PD formulation (DGME-based) on about 90-year-old documents, (3) the current sequence (incl. IND/Zn and Synperonic N-based PD) and the alternate PD (DGME-based) on about 16- to 22-year-old cheques, and (4) the blue toning post-process on 90-year-old documents. Overall, the conclusions were the following:

- the amino acid reagents successfully developed marks on about 30-year-old documents but were found to be less effective on older ones (environmental exposure unknown);
- PD was shown to be highly effective at detecting marks on old documents, up to 90-years-old, and should consequently be considered in the sequence as it does not appear to be affected by the application of amino acid reagents beforehand;
- no noticeable difference in performance between the Synperonic N-based PD formulations and the proposed alternative based on DGME;
- blue toning (see below) resulted in +10% to +25% marks compared to PD only;
- additional marks could appear on documents processed with amino acid reagents several days after the treatment.

**Blue toning** – Blue toning is a new PD post-processing proposed by Bleay et al. [255]. Using Fotospeed BT20 Blue Toner, blue toning consists of chemically replacing silver by a ferric ferrocyanide complex (Prussian blue). The sample must be first wetted before being immersed in the blue toning solution for 3 min. The sample is then rinsed in water for 5 min and then in a print washer for 10 min. As a result, fingermarks detected by PD appear blue, which makes them more visible and helps enhance the contrast in some cases. Overall, the authors observed +10% to +25% marks compared to PD only.

### 3.2.6. T/Powder dusting

**Preliminary remark regarding nanoparticles:** As emphasized in the “Research interests overview” section above, there has been a dramatic increase in publications recommending the dry dusting of nanoparticles and of so-called “nanophosphors”, which are composed of aggregated nanoparticles. The most commonly encountered justification is that “the smaller the particle size, the better the ridge details”, which has been proven false (See 2013–2016 report). Moreover, in several cases, fingermark detection capability appears as a pretext for the synthesis and characterization of optically-active nanomaterials. For ethical and health and safety reasons, this practice should be avoided at all costs. Similarly to the 2013–2016 report, we have taken the decision to cite these publications without describing their content further [257–353]. All the articles below refer to the application of micron-sized particles.

**Preliminary/Pilot studies:** Several kinds of compounds were proposed to detect fingermarks on non-porous substrates through powder dusting: ABC dry fire extinguisher powder [354], Fuller’s earth [355], anthracene and naphthalene [356], BaTiO<sub>3</sub>:Dy<sup>3+</sup> microspheres [357], chitosan-tripolyphosphate microparticles [358], La<sub>2</sub>(MoO<sub>4</sub>)<sub>3</sub>:Eu<sup>3+</sup> microcrystals [359], phenyl-doped graphitic carbon nitride [360], bis(salicylidene) cyclohexyl-1,2-diamino zinc(II) complex [361], functionalized montmorillonite [362,363], carbon dot@TiO<sub>2</sub> core-shell composite [288], household powders such as gram flour, coriander, cumin or black pepper [364], Ca<sub>4</sub>(PO<sub>4</sub>)<sub>2</sub>O:Eu<sup>2+</sup> phosphors [365], Eu(Phen)<sub>2</sub> complex intercalated clay hybrids [366], AIE-based magnetic powder doped with tetraphenylethene derivatives [367], pyrene [368], silica microparticles coated with phenanthroimidazole derivative [369], europium (III) coordination polymer [370], Yb<sup>3+</sup>/Er<sup>3+</sup> co-doped ceramics presenting upconversion properties [371], cationic dye – diatomite composite [372], benzazole-doped silica microparticles [373], Yb<sup>3+</sup>/Er<sup>3+</sup>/Tm<sup>3+</sup> doped fluorides presenting upconversion properties [374], porous graphitic carbon nitride [375], YBO<sub>3</sub>:Eu<sup>3+</sup>/Tb<sup>3+</sup> phosphors [376], Mg<sub>2</sub>TiO<sub>4</sub>:Mn<sup>4+</sup> phosphor [377], carbazolyl-based phosphor [378], and AIE-based imidazole derivatives

[379]. It should be emphasized that several of these studies are conducted with an extremely low number of donors leaving fresh, sebum-rich fingermarks. Similar to the dusting of nanoparticles, fingermark detection capability often appears as a pretext for the synthesis and characterization of optically-active materials. The expressed conclusions are consequently to be taken with extreme caution.

**NIR luminescent powder:** Spirulina platensis, cuprorivaite, and chromium-doped zinc gallogermanate were proposed as dusting powders for their ability to emit in the NIR range [380]. Among the advantages of an NIR-emitting powder, the suppression of some background illustrations and of reflective interference from shiny surfaces appears to be the most relevant in the context of detection.

**Wildlife:** Following a previous study, McMorris et al. studied how environmental exposure could accelerate the deterioration of fingermarks left on bird of prey feathers [381]. Green magnetic fluorescent powder successfully detected marks on feathers stored indoors for up to 60 days, while this limit drops to 14 days for feathers exposed to outdoor conditions (however occasional successes were obtained for an exposure time of 21 days).

**Impact on DNA profiling:** The effects of fpNatural 1™ [382] and of black and magnetic powders (Sirchie®) on the recovery of DNA were covered, as well as cross-contamination issues [383] – see section 3.2.20 for details.

**Available reviews:** overview [384] and critical review [385] regarding the application of nanoparticles to fingermarks.

**Acronyms used:** **AIE** (aggregation-induced emission), **NIR** (near-infrared)

### 3.2.7. T/Powder suspensions

**Preliminary/Pilot studies:** A cost-effective WPS formulation composed of zinc carbonate, natural dye (curcumin and anthocyanin) and a liquid detergent was proposed to detect fingermarks on wetted non-porous substrates [386].

**Advanced/Operational studies:** Three papers coming from the CAST (UK) were dedicated to the iron-based BPS, with regards to its composition, performance and shelf-life [387–389] – see details below.

**Available reviews:** SPR [390].

**Acronyms used:** **ABS** (acrylonitrile butadiene styrene), **BPS** (black powder suspension), **C-IOPS-09** (2009 CAST iron (II/III) black powder suspension), **HDPE** (high density polyethylene), **PE** (polyethylene), **SPR** (small particle reagent), **T100-EG** (Triton X-100 + ethylene glycol), **uPVC** (unplasticized polyvinyl chloride), **WPS** (white powder suspension)

**Black Powder Suspension (UK)** – Iron-based BPS can be used to detect fingermarks on non-porous substrates, especially if they have been wetted. The performance of a BPS is however closely linked to its formulation and the reagents used. In their first paper, Downham et al. focused on the 2009 CAST iron (II/III) BPS formulation (aka C-IOPS-09), especially with regards to the providers of the iron (II/III) oxide chemical, the performance of T100-EG as an alternative to Kodak Photo-Flo, and the shelf-life of the formulation [387]. A four-step study was consequently carried out using seven donors, natural marks, depletion series, and a variety of non-porous substrates (i.e., painted steel, PE board, HDPE carrier bag, glass, uPVC, and Formica laminate). In the second paper, the authors focused on the detergent formulation, especially with regards to the molecular mechanisms taking place in solution and the possibility of diluting the detergent solution [388]. To reach that goal, a

three-step study was carried out with a methodology similar to the first one (considered substrates: painted steel, PE board, plastic board, HDPE carrier bag, uPVC, Formica laminate, and pale wood-effect laminate). In their third paper, the authors focused on the iron (II/III) oxide providers and on the possibility of using Tween 20 to replace Triton-X100, which could become hard to purchase due to EU regulations [389]. In that case, a four-step study similar to the previous ones was carried out (considered substrates: painted steel, ABS board, plastic board, HDPE carrier bag, uPVC, Formica laminate, ceramic tile, glass, and pale wood-effect laminate).

The conclusions of all three studies were the following:

(Note: references are added to ensure a temporal continuity between the observations).

- iron (II/III) oxide chemical [387] – Not all chemicals presenting the CAS # 1317-61-9 are suitable for BPS. Indeed, it appears that the chemicals provided by Fisher Scientific, Sigma Aldrich, and Bayferrox were equivalent in appearance (i.e., black paint) and performance. On the contrary, the chemicals provided by Mistral Chemicals and Scientific Laboratory Supplies resulted in poor detection performance, chemicals that are difficult to keep mixed and blue-tinted black solutions. With regards to particle size, it appears that the first three providers present particles ranging from 50 nm to 1000 nm, which differs from Mistral Chemicals (<75 µm) and Scientific Laboratory Supplies (no data);
- iron (II/III) oxide chemical [389] – Different batches from the recommended Fischer Scientific iron (ii/III) oxide (product #: I/1100/53) could lead to major differences in performance with the C-IOPS-09 (e.g., background staining, performance loss up to approximately –20%, incompatibility with diluted surfactant formulations). The size distribution of the particles seems to be the key parameter (in the case of Fischer Scientific, a modification in the size distribution was observed between the 2008 and 2015 batches). Therefore, the authors recommended the use of 50–100 nm iron (II/III) oxide nanopowder from Sigma Aldrich (product #: 637106).
- detergent [387] – Kodak Photo-Flo can be replaced by T100-EG as both formulations lead to similar performance;
- detergent [387] – No loss of effectiveness was observed when using a fresh or a 2-year-old detergent solution;
- detergent [387] – The Triton-X100 concentration in the T100-EG mix could be reduced 10 times without noticeably impacting the performance of the BPS. However, the concentration must ensure the formation of micelles, critical in the prevention of unwanted iron oxide deposition;
- detergent [388] – Ethylene glycol plays no major role in the detection process but helps Triton-X100 to dissolve and makes post-application rinsing easier;
- detergent [388] – The T100-EG solution could be diluted by a factor of 10 (equivalent performance) up to a factor of 100 (marginal decline of performance). However, in their next study, the authors emphasized that this conclusion may be closely related to the properties of the iron (II/III) batch that was used [389];
- detergent [389] – A 4% or a 40% Tween 20 surfactant solution could be used in replacement of Triton-X100
- BPS shelf-life [387] – C-IOPS-09 remains effective up to 100 days after being prepared, even if a freshly-prepared one recovered slightly more marks (results to be confirmed);

Overall, CAST do not recommend modifying the C-IOPS-09 formulation based on these observations, for extended trials including an extended range of substrates are still required. Nevertheless, a 10% Tween 20 surfactant solution combined with

iron (II/III) oxide nanopowder (50–100 nm) from Sigma Aldrich showed promising performance. As emphasized by the authors, control measures should be taken during the preparation of the BPS formulation.

### 3.2.8. T/Nanoparticles in solution

**Preliminary/Pilot studies:** Several kinds of NPs in solution were proposed to detect fingerprints on various substrates (e.g., non-porous, semi-porous, adhesive side of tapes): C-dots ⇨ FITC-functionalized [391], green-emitting [392], white-emitting [393], orange-emitting [394], nitrogen- and sulfur-doped [321], suspended in poly (vinyl alcohol) [395], sprayed in hydrochloride solution [396]; gold NPs ⇨ functionalized with lysozyme-targeting aptamers [397], functionalized with antibodies (i.e., anti-lysozyme, anti-human IgG and anti-cotinine) to serve as tags for SERS chemical imaging [398]; nanophosphors ⇨ NIR-emitting lanthanide-based [399], EDC/NHS-functionalized [400]; nanorods ⇨ carboxyl-functionalized [401], antibody-functionalized [402]; QDs ⇨ functionalized cadmium-based [403]; silica NPs ⇨ methylene blue-doped [283], carbon-doped [326], FITC-doped silica NPs [404], fluorophores-doped [286], Nile red-doped [405]; silver NPs ⇨ in-situ generated [406]; other NPs ⇨ NIR-emitting polymer dots doped with NIN [407], AIE-based nanomaterials built on diphenylpyrimidinone and salicylideneamine [408], poly [p-phenylenevinylene] [409], functionalized carbon nanotubes [410]; Co<sub>2</sub>TiO<sub>4</sub> NPs [310], rare-earth-doped upconversion NPs [411], AIE-based heteroleptic iridium complexes [412,413], coronenediimide nanostructures [337], and antibody-functionalized polystyrene NPs applied to drug-spiked fingerprints [414]. It should be emphasized that several of these studies were conducted with an extremely low number of donors leaving fresh, sebum-rich fingerprints, or were carried out by dropping few microliters of solution directly on the fingerprints. The expressed conclusions are consequently to be taken with extreme caution.

**Silica nanoparticles (extended studies):** Lee et al. carried out an optimization study with regards to the use of RuBpy-doped carboxyl-functionalized silica NPs in aqueous solution to detect fingerprints on non-porous substrates [415]. Several parameters were considered (i.e., NPs concentration, ionic strength, pH, immersion time and temperature of the working solution) and the optimized protocol compared with a previously-published one. Overall, detection performance was improved by decreasing the NPs concentration and heating the working solution to 40°C. The authors emphasized the need for further optimization with regards to batch-to-batch consistency and relative performance with conventional methods. The efficiency of NR-doped mesoporous silica NPs in aqueous solution was compared to ORO and to NR [405]. The underlying idea is that NR molecules contained in the silica NPs would be progressively released and would target the lipid fraction of the secretion residue. The authors optimized the detection protocol involving the NPs (i.e., NPs concentration, immersion time and temperature of the working solution) and compared its performance with ORO and NR solutions on dry and wetted thermal papers (wetting times: from 2 h to 2 weeks). Similarly to Lee et al.'s study, heating the working solution gave better results although a temperature of 25°C was considered to prevent any damage to the text. Overall, the performance of the optimized NPs solution was assessed to be the highest, followed by NR and ORO.

**Gold nanoparticles:** MMD showed promising results on the new €5 and €10 banknotes [416] – see section 3.2.10 for details. Considering 23 different substrates (non-porous, porous, semi-

porous), Newland et al. explored the mechanism behind the SMD II process and confirmed some protocol aspects such as the size of the nanoparticles, the solution temperature or the bath shaking, as well as a dependency of the performance to the substrate nature [417]. Another study aimed at determining if some paper characteristics (e.g., roughness, porosity, surface pH) could be correlated to SMD II performance [418]. No correlation was noted, but the authors emphasized the role that the surface coating (silica or calcium carbonate) may play in the detection process. Moret et al. compared SMD II and PD in different scenarios [256] – see section 3.2.5 for details. Antibody-functionalized gold NPs were considered for a multi-target immunogenic approach to detect natural and blood-contaminated fingerprints [419] – see section 3.2.16 for details. **Black powder suspension:** As fully described in the previous section, a 50–100 nm iron (II/III) oxide nanopowder from Sigma Aldrich (product #: 637106) is currently recommended to prepare the C-IOPS-09, resulting in nanoparticles in suspension [387–389].

**Available reviews:** MMD [420], overview [384] and critical review [385] regarding the application of nanoparticles to fingerprints.

**Acronyms used:** **AIE** (aggregation-induced emission), **C-dots** (carbon dots), **C-IOPS-09** (2009 CAST iron [II/III] black powder suspension), **EDC/NHS** (1-ethyl-3-(3-dimethylaminopropyl)-carbodiimide mixed with N-hydroxysuccinimide), **FITC** (fluorescein isothiocyanate), **IgG** (immunoglobulin G), **MMD** (multimetal deposition), **NIN** (ninhydrin), **NIR** (near-infrared), **NPs** (nanoparticles), **NR** (Nile red), **ORO** (Oil Red O), **PD** (physical developer), **QDs** (quantum dots), **RuBpy** (tris [2,20-bipyridyl] dichlororuthenium [II] hexahydrate), **SERS** (surface-enhanced Raman spectroscopy), **SMD** (single metal deposition)

### 3.2.9. S/Adhesives and tapes

**Preliminary/Pilot studies:** OCT is proposed to image fingerprints beneath adhesives without requiring their removal from the surface they adhere to Ref. [421,422]. From the preliminary results that were published, the cross-section imaging ability of the technique allowed the imaging of fingerprints lying beneath opaque tapes. However, further research is required.

**Adhesive removal:** A small-scale study addressed the question of the impact of using un-du® on the subsequent cyanoacrylate fuming ⇨ dye sequence [423]. Despite the risk of tearing and gumming observed with duct tape, no detrimental impact was observed, even when using an excess of un-du®.

**Acronyms used:** **OCT** (optical coherence tomography)

### 3.2.10. S/Banknotes

**Preliminary/Pilot studies:** VMD<sub>Cu</sub> combined with NIR observation represents a promising way to detect fingerprints on (UK) polymer banknotes [424], the advantage of using NIR rather than naked eye observation being clearly demonstrated. Additionally, the authors investigated the use of a gelatin lifter applied on the processed banknote and subsequently treated with rubeanic acid to obtain green fingerprints.

**GBP polymer banknotes:** Released in 2016 and 2017, the new €5 and €10 polymer banknotes required updating the existing detection sequences that were developed for paper-based currencies. Two studies specifically addressed these questions [425,426] – see details below. MALDI-MSI was also applied to €5 polymer banknotes as a proof of its ability to image ridge details

without background interference [427] – see section 3.2.19 for details.

**Euro banknotes:** With regards to the new €5 and €10 banknotes, released in 2013 and 2014 respectively, MMD showed promising results compared to the techniques usually recommended for euro banknotes (e.g., IND/Zn, NIN, PD, VMD<sub>Au/Zn</sub>) [416]. The performance of MMD is most likely due to the presence of a protective layer on these two banknotes. According to the authors, the new €20 and €50 banknotes don't have this protective layer, resulting in better performance of the traditional techniques.

**Case report:** In the context of a drug trafficking investigation, a vacuum seal bag was processed with CA, resulting in the detection of a friction ridge-like pattern [428]. After further examination, the “friction ridges” were linked to the face of Queen Elizabeth II present on the CAD 20 polymer banknotes. This case raises the question of a transfer between the banknote and the plastic substrate. The most likely explanation is an embossment process caused by the intaglio printings contaminated with secretions present on the banknote surface.

**Acronyms used:** **BPS** (black powder suspension), **BR14** (Basic Red 14), **BY40** (Basic Yellow 40), **C-IOPS-09** (2009 CAST iron [II/III] black powder suspension), **CA** (cyanoacrylate fuming), **IND/Zn** (1,2-indanedione containing zinc chloride), **IR** (infrared), **LWUV** (Long-wave ultraviolet), **MALDI** (matrix assisted laser desorption ionisation), **MMD** (multimetal deposition), **MSI** (mass spectrometry combined with imaging), **NIN** (ninhydrin), **NIR** (near-infrared), **PD** (physical developer), **SPR** (small particle reagent), **VMD** (vacuum metal deposition), **VMD<sub>Ag</sub>** (silver-based monometallic VMD), **VMD<sub>Ag/Zn</sub>** (silver-zinc VMD), **VMD<sub>Au/Zn</sub>** (traditional gold-zinc VMD), **VMD<sub>Cu</sub>** (copper-based monometallic VMD), **WPS** (white powder suspension)

**Polymer-based GBP banknotes** – In their first study, Downham et al. assessed 14 detection techniques/processes for their ability to detect fingermarks on the new £5 banknotes [425]. The following techniques were considered: anti-Stokes powder, NIN, black magnetic powder, *fpNatural*® 1 and 2 powders (emitting in NIR), C-IOPS-09, CA, VMD<sub>Au/Zn</sub>, VMD<sub>Ag/Zn</sub>, MMD, gel lifter, Lumicyano, PolyCyano UV, SPR, Wet Powder™ BPS and WPS. All of the processes, with the exception of NIN, were able to detect fingermarks on the £5 banknote. In terms of performance, MMD, C-IOPS-09 and Wet Powder™ WPS were the three techniques that recovered marks with the highest quality overall, followed by *fpNatural*® 2 (NIR-NIR fluorescence), Wet Powder™ BPS and black magnetic powder. Quite surprisingly, the three cyanoacrylate techniques (CA, Lumicyano and PolyCyano UV) and the two VMD processes (VMD<sub>Au/Zn</sub> and VMD<sub>Ag/Zn</sub>) resulted in lower performances, although they are usually considered as the most-suited techniques for non-porous and semi-porous substrates. Regarding all the techniques, the authors observed that the primary observation conditions (white light or luminescence) were somewhat limited with regards to the complexity of the background. They emphasized the added benefit of observing the detection results with IR reflection or of applying a gel lifter subsequently, as it led to +6% up to +44% recovered marks. NIR-emitting powders (such as *fpNatural*® 1 and 2 powders) could overcome background issues. The authors also emphasized that ethanol-based and aqueous-based BY40 and BR14 dye staining were unsuitable for this substrate, due to dye absorption by the banknote and strong background emission afterwards. In their second study, Downham et al. assessed 5 detection sequences for their ability to detect fingermarks on the new £10 banknotes [426]. The following sequences were considered:

- (S.1) CA ⇨ VMD<sub>Au/Zn</sub> ⇨ BY40 (whole banknote) ⇨ VMD<sub>Ag</sub> ⇨ gel lifting
- (S.2) CA ⇨ *fpNatural*® 2 powder ⇨ black magnetic powder ⇨ gel lifting
- (S.3) black magnetic powder ⇨ C-IOPS-09 ⇨ gel lifting
- (S.4) *fpNatural*® 2 powder ⇨ *fpNatural*® 2 suspension ⇨ gel lifting
- (S.5) MMD ⇨ blue toning (BT20 Blue toner kit, Fotospeed) ⇨ gel lifting

In addition to conventional observation means (white light and luminescence), the authors also observed the results under IR reflection (RG780 nm long pass filter) and LWUV reflection (330–385 nm band-pass filter). Overall, Sequence 4 resulted in the highest performance, closely followed by Sequence 3. Black magnetic powder imaged in the NIR was found to be the best single process, as it detected almost all the marks visualized upon application of Sequence 3. The use of *fpNatural*® 2 was more challenging compared to black magnetic powder, mostly due to its pale blue colour which makes the progress of the mark enhancement process difficult to assess. Finally, similar to the first study, observation under IR or LWUV reflection and gel lifting (using black gel lifter, BVDA) provided an efficient means to increase the number of detected marks. LWUV reflection could also be used on unprocessed banknotes to observe latent marks in the early stage of a sequence. Given that both these studies were performed on un-circulated banknotes, further studies are expected to be conducted on worn ones.

### 3.2.11. S/Metal and cartridge cases

**Preliminary/Pilot studies:** Various approaches were proposed to detect fingermarks on cartridge cases or other metallic surfaces: molecular complexes composed of L-alanine and conventional amino acid reagents (i.e., DFO, IND, MTN, and NIN) applied to brass and fired cartridges [429]; co-electrodeposition of silver and copper particles in deep eutectic solvent (mix of choline chloride and ethylene glycol), the secretion residue acting as a mask [430]; gel-based electrolytes (calcium chloride and potassium permanganate) applied to stainless steel and aluminium [431]; deposition of potassium birnessite (oxide mineral of manganese) on unfired cartridges and flat metal surfaces, the secretion residue acting as a mask [432]; electrostatic adsorption using a modified portable high-low voltage self-discharge device, applied to unfired brass cartridges [433]; nanoconjugate composed of carbon nanotubes, *Candida rugosa* lipase and safranin, applied in suspension on immersed stainless steel knives [410]; SKP and SEM/EPMA imaging of latent and VMD-processed fingermarks on metallic surfaces [434]; ToF-SIMS applied to flat metallic surfaces [435]; modified VMD process involving the deposition of gold followed by zinc, ZnS, or ZnO on aluminium [436]; and in situ reduction of aryldiazonium gold (III) salts induced by human sweat, applied to flat metallic surfaces [437].

**Metal corrosion:** Cooper-Dunn et al. proposed to investigate further how the composition of secretion residue could influence brass corrosion and fingermark detection [438]. Using different kinds of secretions (i.e., eccrine-rich, sebum-rich, mix, and artificial secretions), the authors emphasized that stearic acid (free fatty acid) can induce corrosion and that winter/summer periods impact the ability of eccrine fingermarks to corrode brass.

**Comparative studies:** The performance of GB on brass cartridge cases was compared to the conventional approaches (e.g., CA ⇨ BY40 or palladium deposition) [439–441] – see details below.

An extensive study focused on the mechanisms behind the recovery of fingerprints from brass cartridge cases [442] – see details below. The way exposure to outdoor environment may impact the recovery of fingerprints on metallic items was investigated [443] – see details below.

**Fingerprint vs touch DNA:** A study addressed the decision of the Raleigh Police Department (Raleigh, NC, USA) to send their cartridge cases for touch DNA instead of fingerprint detection, due to more success with DNA profiling than with ridge details [444]. Using unfired and fired brass cases, deliberate and chance fingerprint deposition (using up to 5 donors), and two kinds of firearms (i.e., semi-automatic and revolver), the authors processed five hundred cases with the conventional sequence CA ⇄ R6G. With the exception of deliberately handled cartridge cases, which led to minimal ridge details (assessed as “no value for identification”), no ridge detail was observed on the fired cases that were naturally handled.

**Available reviews:** extensive review about optical, physical, chemical, and physicochemical techniques to detect fingerprints on metallic surfaces [445].

**Acronyms used:** CA (cyanoacrylate), BY40 (Basic Yellow 40), C-BPS (carbon-based powder suspension), DFO (1,8-diazafluoren-9-one), EPMA (electron probe micro-analyser), EDX (energy dispersive X-ray spectroscopy), GB (gun blue), IND (1,2-indanedione), MTN (5-methylthioninhydrin), NIN (ninhydrin), R6G (Rhodamine 6G), SEM (scanning electron microscope), SKP (scanning Kelvin probe), ToF-SIMS (time of flight secondary ion mass spectroscopy), VMD (vacuum metal deposition), XRD (X-ray diffraction)

**Gun blue** – Dove proposed the application of GB through electrodeposition, as an alternative to the “passive” application of the reagent [439,440]. The underlying idea is that an electrically stimulated metallic surface would attract the chromic agents and hence promote the reaction of GB with the metal, reducing in the same way the risk of overdevelopment through unwanted reaction with the secretion residue. The first study was conducted with fresh sebum-rich fingerprints left by one donor on spent brass and nickel-plated brass cases, considering dilution series of GB (Super Blue Liquid Gun Blue from Birchwood Casey) [439]. The electrodeposition of GB was carried out by immersing a cartridge case in a diluted GB solution while an electric current provided by a 1.5V battery ran through it. The optimized GB concentration was determined to be 15% of the commercial solution (15 mL GB + 85 mL water), which resulted in a homogenous deposition across the substrate in approximately 18 s. The optimized GB electrodeposition protocol was compared with two conventional approaches (i.e., “passive” application of GB through immersion and the CA ⇄ BY40 sequence), using separate brass cases. In terms of performance, GB electrodeposition gave the highest scores, followed by CA ⇄ BY40 and passive GB. The protocol seems somewhat optimized for brass only, given the mixed results obtained on nickel-plated brass cases. In his second study, Dove compared the performance of GB electrodeposition to palladium deposition using fired cartridge cases [440]. The study was conducted with fresh sebum-rich fingerprints from one donor and brass cases that were left unfired or that were loaded in a weapon magazine then fired. On unfired cartridge cases, GB electrodeposition resulted in a significantly higher quality compared to palladium deposition. On fired cases, the quality scores tended to be similar between the two reagents, with a majority of fingerprints not detected. Also, the author observed that ridge details were mostly visible near the base of the cartridge case, as opposed to the mouth where secretion residues were damaged by the firing process. This observation is consistent with former conclusions in the

field. The author proposed different explanations for this phenomenon (e.g., hardened structure near the rim, friction upon ejection, temperature across the surface of the case and blowback of heated gases). It has to be noted that Girelli et al. also investigated the deterioration mechanism upon firing [442] – see below. Morrissey et al. proposed to compare the performance of three processes aiming at detecting fingerprints on unfired and fired brass cartridge cases: (1) GB (Birchwood Casey Perma Blue, diluted by two), (2) CA ⇄ BY40, (3) CA ⇄ GB [441]. Similar to Dove, fresh fingerprints left by one donor were considered, but these were eccrine in their composition. On unfired and fired cases, the performance of GB alone was higher than CA ⇄ BY40 and CA ⇄ GB. Given the limited and ideal nature of the fingerprint sets used in these three studies (i.e., fresh sebum-rich/eccrine marks left by one donor), caution should be taken regarding the conclusions, further studies being required.

**Recovery on brass** – In a thorough and extensive study, Girelli et al. aimed at offering a better understanding of the mechanisms underlying the recovery of fingerprints from brass [442]. To reach that goal, the authors considered eccrine fingerprints left by two donors, three brass substrates (i.e.,  $\alpha$ -brass plates, unfired and fired cases), three post-firing aging times (i.e., 1, 7 and 14 days), four detection sequences: (1) CA ⇄ GB ⇄ BY40, (2) CA ⇄ GB ⇄ Ardrex, (3) Prussian blue, and (4) aqueous electrolytes, and various surface characterization techniques (i.e., EDX, SEM, XRD and metallographic examination). On brass plates bearing fingerprints, the firing process was mimicked by heating them at 63°C and 200°C. Overall, the sequence CA ⇄ GB ⇄ dye (BY40 or Ardrex) outperformed the application of Prussian blue and aqueous electrolytes. GB seemed particularly efficient on aged marks that were exposed to high temperatures, whereas CA was more apt to detect fresh marks exposed to lower temperatures. The authors observed that cartridge cases from the same batch could differ in their surface morphology and oxidation level, supporting the fact that a combination of detection techniques are required compared to a single process. With regards to the detrimental impact of the firing process on secretion residue, the authors concluded that it was mostly caused by the blowback of hot gases through the loosened interstices between the cartridge case and the firearm chamber wall (and to a lesser extent by the friction between these two elements). This statement was supported by the high-speed recording of firing events, by the ellipsoidal shape of the degradation pattern, and by the higher resistance of sebum-rich secretions (increased viscosity).

**Process efficiency** – Pitera et al. studied how outdoor environment may impact the effectiveness of fingerprint recovery on metal substrates [443]. To reach that goal, the authors considered three metallic substrates (i.e., bronze, brass, and stainless steel), two environmental conditions (i.e., inside storage and outdoor for 2 years), three types of secretions provided by a unique donor (i.e., natural, sebum-rich and eccrine), three aging times (i.e., 1 day, 1 week and 2 weeks), and four detection processes (i.e., CA ⇄ BY40, Lumicyano 1%, GB, and C-BPS). It has to be noted that the fingerprints were left on the weathered metals after these were exposed to outdoor conditions for two years (not before). GB solution was purchased (Perma Blue® Liquid Gun Blue, Birchwood-Casey) and diluted with water before used (1:1 and 1:32.3 v/v). The C-BPS was also purchased (Wet Powder™ Black, Kjell Carlsson Innovation) and applied with a brush. The impact of outdoor environment on the metallic surfaces was observed with SEM: bronze appeared duller with rougher and pitted surface; brass appeared duller but without coarser pitting, the microstructure seeming less affected than bronze; stainless steel was the least affected of the three metals, which is not surprising. With regards to the effectiveness of detection techniques, not a single one appeared optimal for all the

metals (see below). Overall, weathered metals were able to be processed by at least one technique (*i.e.*, weathered bronze and brass by CA ⇨ BY40 or Lumicyano, weathered stainless steel by C-BPS). It appears important to emphasize again that the fingerprints were left on the weathered substrates but were not exposed to outdoor environment. Also, given the fact that only one donor was considered in this study, further studies are awaited to confirm these observations. With regards to the process effectiveness, the following observations were made:

- CA, CA ⇨ BY40, and Lumicyano: effective on all the metals, especially unexposed bronze and brass, with a slight decrease of efficiency with weathered metals;
- GB: especially effective on unexposed bronze, slightly effective on unexposed brass, but ineffective on stainless steel and all the weathered metals;
- C-BPS: especially effective on stainless steel (new and weathered), slightly effective on new bronze and brass.

### 3.2.12. S/Skin and leather

**Preliminary/Pilot studies:** The simultaneous fuming of cyanoacrylate and iodine on leather is proposed to detect marks on leather [446]. To reach that goal, a home-made fuming cabinet equipped with two hot air guns (one for cyanoacrylate and the other for iodine) was built. Positive results were obtained on light-colored leather, however poor contrast negatively impacted the results obtained on dark-colored leather.

### 3.2.13. S/Thermal papers

**Preliminary/Pilot studies:** Proof-of-concept studies to detect fingerprints on thermal papers involved the vacuum sublimation of Lawsone [447] or a brief immersion in hot water [448]. The latter was shown to be destructive for the sample and secretions, with low performance on aged marks. Exposure to hydrochloric fumes was compared to ThermoNin [449]. Using fresh to 21-day-old artificial eccrine secretions and sebum-rich fingerprints, the authors showed that ThermoNin was consistent in its ability to detect fingerprints on thermal papers. On the contrary, the performance of hydrochloric fumes was inconsistent and mostly successful with fresh sebum-rich marks. It should also be noted that artificial sebaceous secretions (Lightning Powder pad) were discarded because they reacted with the thermal paper upon contact and resulted in the immediate development of dark ridge details.

**Advanced/Operational studies:** A new optimized detection sequence dedicated to thermal papers has been proposed [450] – see details below. Similar to what has been concluded for DFO (See 2013–2016 report), the addition of PVP to IND/Zn makes it suitable to detect fingerprints on thermal papers while avoiding any risk of background darkening [450]. A solution of IND/Zn: 8 wt% PVP (1 : 0.4 v/v) and a development under heat press (10 s at 150°C) were set as the optimal formulation.

**Acronyms used:** **DABCO** (1,4-diazabicyclo [2.2.2]octane), **IND/Zn** (1,2-indanedione containing zinc chloride), **LCA** (Lumicyano), **NIN** (ninhydrin), **PE** (petroleum ether), **PVP** (polyvinylpyrrolidone)

**Optimized detection sequence** – In a two-step study, Hallez et al. assessed the performance of five detection techniques (*i.e.*, black magnetic powder, LCA 6%, thermal development, NIN, and IND/Zn) and four whitening agents (*i.e.*, acetone, ethyl acetate, bleach 3.8% and DABCO) combined in three sequences that were further compared with the sequence currently recommended by

the French gendarmerie [450]. It has to be noted that the IND/Zn and NIN formulations were conventional ones (PE as carrier), IND/Zn samples were processed through heat press (10 s at 165°C), and NIN samples were left in a ventilated closet for 48 h. The first step consisted of comparing the performance of each technique using two types of thermal papers, five donors, depletion series, and 1-day-old to 2-month-old fingerprints. As a result, three sequences were proposed in addition to the one recommended by the French gendarmerie (*i.e.*, Sequence 1):

- (S.1) black magnetic powder ⇨ IND/Zn ⇨ NIN
- (S.2) LCA ⇨ IND/Zn ⇨ whitening (ethyl acetate) ⇨ NIN
- (S.3) LCA ⇨ whitening (ethyl acetate) ⇨ IND/Zn ⇨ NIN
- (S.4) heat (25sec, 54°C) ⇨ heat (3min, 85°C) ⇨ LCA ⇨ whitening (ethyl acetate) ⇨ IND/Zn ⇨ NIN

The pseudo-operational trial consisted of processing 200 receipts by each of the four sequences. Sequence 4 outperformed the others and led to +16% good quality marks compared to Sequence 1. The authors also emphasized the fact that LCA should be observed through its two excitation domains (*i.e.*, 325 nm and 515 nm) to maximise the recovery of marks.

### 3.2.14. C/Arson scenes

**Practice-oriented or case-related studies:** Recovering fingerprints on petrol bombs (Molotov cocktails) is usually considered after the search for flammable liquids, which requires heating the items for a specified time (*e.g.*, 15 min to 24 h at 50–130°C) [451]. In this context, the authors simulated such a procedure by considering three flammable liquids (*i.e.*, gasoline, kerosene and diesel) and black magnetic powder to detect fingerprints. Among the three flammable liquids, gasoline was shown to be the most detrimental (loss of approximately half the marks after exposure to fumes at 130°C for 15 min), followed by kerosene (–20%) and diesel (no observed deterioration). Lowering the heating temperature reduced the detrimental effect of gasoline (–16% at 60°C for 15 min).

### 3.2.15. C/Blood-containing fingerprints

**Preliminary/Pilot studies:** Various approaches were proposed to detect blood-containing fingerprints: nanoparticles in suspension [310,399,402], water-soluble benzalole dyes [452], ortho-phenylenediamine and Zar-Pro™ strips [453], or aggregation-induced emission involving serum albumin and tetraphenylethene maleimide [454]. Steam thermography has been investigated further in its ability to image ridge details from blood fingerprints left on fabrics [455], with positive results for hydrophobic fabrics such as acrylic and polyester. Extracts of Glycine max root nodules were proposed to be used as artificial blood for their content in leghaemoglobin, structurally similar to human haemoglobin and which seems to react with heme-sensitive reagents and protein stains [456]. MALDI-ToF-MS was considered to determine the origin of a blood stain (*i.e.*, “human” as opposed to “non-human”) following an in-situ bottom-up approach combined with proteomic differentiation of haemoglobin [457]. Visible wavelength reflectance HSI was proposed to estimate the age of blood-containing fingerprints [458]. Finally, with regards to blood pattern analysis, Shiri et al. emphasized the fact that secretion residue on a substrate can alter the size and shape of bloodstains [459].

**Practice-oriented or case-related studies:** NIN being sprayed at crime scenes (*i.e.*, plaster walls), the question was raised

whether it was possible to distinguish non-blood marks from blood-containing marks, both appearing purplish after reaction with NIN [460]. The authors showed that slight differences exist in the UV–vis spectra and proposed the use of a colour spectrophotometer with a  $L^*a^*b$  scale to successfully differentiate both kinds of marks.

In another study, the question was raised whether it was possible that a wet finger in contact with dry blood could leave a blood-containing fingermarks afterwards (substrate: paint metal from a car) [461]. Considering natural and induced fingertip wetting, the authors observed that it was possible when the fingertip was wetted with cold tap water shortly before contact with blood, but that a negative mark was always left on the original dry stain (due to removal of blood from it). In the same context, the mechanisms leading to the deposition of a blood-containing fingermark were thoroughly investigated by two teams [462,463] – see details below.

The impact of the sequential application of detection techniques was addressed with regards to the detection of blood marks on dark substrates [464], the efficiency of protein stains post-CA [465] and the efficiency of presumptive/confirmatory tests for blood [466] – see details below.

In a study aimed at determining the possibilities for detecting concealed blood marks at a crime scene, Lupica considered two scenarios: (1) 24-h-old blood marks on walls subsequently concealed with paint (up to three layers), and (2) 24-h-old blood marks left by a shoe sole on a carpet subsequently cleaned using a Rug Doctor carpet cleaning machine [467]. On the wall, a powerful alternate light source (e.g. Mini-Crimescope, exc. 420–430 nm) and Bluestar Forensic gave positive results in terms of location and shape of the blood stains (no ridge details though, possibly due to the use of bovine blood). On the carpet, only Bluestar Forensic yielded positive results. In the same context, another study focused on Bluestar Forensic's ability to retrieve blood marks concealed by paint [468]. Considering three different substrates (i.e., brick, flakeboard and drywall) and three different paints, the authors showed that Bluestar Forensic successfully enhanced blood marks through three to four layers of paint (no ridge details though), with the performance mostly influenced by the type and colour of paint.

**Acronyms used:** **5-SSA** (5-sulfosalicylic acid), **AV17** (Acid Violet 17), **AY7** (Acid Yellow 7), **BY40** (Basic Yellow 40), **CA** (cyanoacrylate), **DFO** (1,8-diazafluoren-9-one), **hHb** (human haemoglobin), **HSI** (hyperspectral imaging), **IFM** (infinite focus microscopy), **IO-BPS** (iron-oxide black powder suspension), **IR** (infrared), **KM** (Kastle-Meyer), **MALDI** (matrix assisted laser desorption ionisation), **MS** (mass spectrometry), **NIN** (ninhydrin), **PP** (polypropylene), **SPR-W** (white-colored small particle reagent), **ToF** (time of flight), **VMD<sub>Au/Zn</sub>** (traditional gold-zinc vacuum metal deposition), **WEAA** (mix of water, ethanol, and acetic acid in a 14:5:1 v/v/v ratio)

**Formation mechanisms** – Blood-containing fingermarks can be obtained from three different mechanisms: (1) blood-contaminated fingertip in contact with a surface, (2) clean fingertip in contact with a blood-contaminated surface, and (3) latent fingermark subsequently contaminated with blood (aka “faux blood mark”). Providing a method able to distinguish these three scenarios in casework could bring valuable information with regards to the reconstruction of events. Deiningering et al. proposed to use IFM and its ability to characterize the topology of a sample to distinguish among these three mechanisms [462]. The underlying idea was that 3D measurements would emphasize distinctive differences in ridge and valley heights between samples generated by the three scenarios. Unfortunately, it was shown that the

deposition substrate strongly affects the way blood behaves (e.g., spreading before drying) and hence the height measurements. This observation makes the height measurement by IFM unsuitable for an application to crime-scene cases due to the high variety of substrates and conditions that could be encountered. Also, IFM was shown to be incompatible with glass and gel lifters due to the absence of light reflectance apart from the ridges. Geller et al. addressed the same question by considering an optical approach [463]. The underlying idea was that the colour of the ridges with regards to valley would emphasize distinctive differences between samples generated by the three scenarios. Unfortunately, the results showed that the ridge colour approach cannot assuredly distinguish the three mechanisms, mainly due to the observation of colour inversion cases (i.e., cases for which the colour of ridge/valleys is the opposite of the predicted models). Also, the approach requires identifying ridges, which requires the presence of pores or obtaining of reference fingerprints. About tonal reversal (i.e., presence of blood in the valleys), it should be noted this phenomenon can be observed for each of the three mechanisms, as reiterated by Deiningering et al. [462].

**Sequences/Impact of techniques** – As a follow-up of their study dedicated to the detection of blood marks on dark plastic, Bouwmeester et al. investigated how CA ⇔ BY40 and SPR-W could be used in sequence and their impact on DNA recovery [464]. Using black PP plates and blood marks aged from 1 day to 6 months, the authors showed that a better contrast was obtained when the blood marks were first fixed with ethanol then processed with SPR-W. The application of CA (⇔ BY40) was seen to be detrimental for the recovery of blood marks. On the contrary, a higher amount of DNA was recovered subsequently to SPR-W if CA (⇔ BY40) was applied first, emphasizing the importance of prioritizing evidence according to the case. Mutter et al. proposed to investigate the impact that CA (atmospheric and vacuum) may have on the subsequent application of protein stains (methanol-based and WEAA) [465]. Considering three non-porous substrates (i.e., white plastic bags, white ceramic tiles and aluminium sheets), depletion series, lysed horse blood, and four aging times (from 1 day to 28 days), the authors compared the sequence CA ⇔ AV17 with the sole application of AV17 [note: using different sets of fingermarks]. Before protein staining, the blood marks were fixed with an aqueous solution of 5-SSA. The results showed a pronounced detrimental effect of CA (atmospheric and vacuum) on AV17 (WEAA), with approximately –90% marks detected. However, no detrimental effect was observed when AV17 (methanol) was used subsequently to CA, supposedly due to the ability of methanol to soften the polymer and provide access to the underlying blood. However, as emphasized by the authors, the use of a methanol-based formulation may have a detrimental impact on the subsequent presumptive/confirmatory tests, as opposed to the WEAA formulation [note: this is confirmed by the study of Stewart et al., described just below]. Finally, VMD<sub>Au/Zn</sub> showed no detrimental effect on the subsequent application of AV17. With regards to presumptive/confirmatory tests, Stewart et al. investigated how their efficiency may be impacted by the application of detection techniques [466]. The authors considered four substrates (i.e., white paper, brown envelope, white tile, and blue linoleum), depletion series, three aging times (from 1 day to 28 days), eight detection techniques (i.e., black magnetic powder, IO-BPS, CA, AV17, AY7, NIN, DFO, and Bluestar Forensic Magnum), one presumptive test (i.e., KM), and two confirmatory tests (i.e., Takayama and RSID-Blood). Please note that both methanol and WEAA formulations were tested for AV17 and AY7. Overall, KM and RSID-blood were largely unaffected by the detection techniques:

- KM: the methanol formulations of AV17 and AY7 inhibited the KM reaction for >50% of the marks or delayed its reaction by

1 min, on the contrary to the WEAA formulations which has no or little impact;

- Takayama: poor sensitivity on un-processed blood marks and easily affected by many detection techniques (e.g., IO-BPS, AV17, AY7, DFO);
- RSID-Blood: the most robust of the three blood tests as it remained largely unaffected, with the exception of NIN.

Besides these results, the authors also emphasized (1) the high sensitivity of KM and RSID-Blood (somewhat inferior to KM) as opposed to the Takayama test, (2) the high performance of AV17 (both formulations), AY7 (both formulations) and Bluestar to detect blood marks, and (3) no detrimental impact of the detection techniques on DNA recovery and profiling.

### 3.2.16. C/Contaminations (other than blood)

**Preliminary/Pilot studies:** The following articles aim at detecting contaminants in fingermarks without ridge pattern imaging. For this reason, they are not extensively described: hyperspectral SRS to image exogenous compounds in spiked fingermarks (i.e., gun powder and benzoic acid) [193], PS-MS to detect illicit drugs in spiked fingermarks [469] or from fingermarks left by drug users [470], IR laser ablation coupled to vacuum capture and MALDI-MS to detect caffeine and condom lubricant in spiked fingermarks [471], silver sputtering combined with MALDI-MSI to detect flunitrazepam-spiked fingermarks [472], PDMS-PDA-Ag sandwich applied to fingermarks and further removed to allow Raman imaging of artificially-contaminated fingermarks (i.e., R6G and 4-ATP) [473], a combination of fingermark lifting (directly from the fingertip or from a surface, using an adhesive enriched with gold NPs) and SERS imaging of artificially-contaminated fingermarks (i.e., R6G and cotinine) [474], SERS applied to artificially-contaminated fingermarks (i.e., TNT, RDX, PETN) [475], MALDI-ToF-MSI applied to fingermarks contaminated with plant-derived psychoactive biomarkers [476], MALDI-MSI applied to spiked fingermarks (i.e., solubilized TNT and medical drug powder) post CA-fuming [241], an LC-MS method aiming at establishing a cut-off between drug users and environmental contamination [477], degradation with time of explosives and illicit drugs in fingermarks using FTIR spectroscopy imaging [478], use of LA-ICP-MSI to image gunshot-related metals in fingermarks [479], NALDI-MSI, MALDI-MSI and DESI-MSI were compared to image spiked fingermarks (i.e., methamphetamine, cocaine and heroin in solution) [480], SERS imaging of sebum-rich fingermarks spiked with methamphetamine (i.e., contact with solution) and left on an agarose gel [481], FT-ICR-MS to identify TATP in secretion residue after explosive handling [482], electrochemiluminescence to image fingermarks spiked with nicotine and TNT (i.e. solution dropped onto existing latent fingermarks) [483], SRXRF applied to sunscreen-contaminated fingermarks [484], antibody-functionalized polystyrene NPs applied to drug-spiked fingermarks (i.e., ketamine and amphetamine in solution on the fingertip) [414], and MALDI-MSI to image cocaine-contaminated fingermarks left on £5 polymer banknotes [427] – see section 3.2.19 for details.

**Body fluid contamination:** The use of antibodies and aptamers to detect fingermarks contaminated with body fluids (i.e., blood, semen, saliva) was investigated [419] – see details below. MALDI-ToF-MSI was applied to image fingermarks left by blood- and vaginal-fluid-contaminated fingertips<sup>[\*]</sup> [485]. AgLDI-MSI was applied to drug- and blood-contaminated fingermarks after their processing with conventional detection techniques [486] – see details below. The use of proteomics to study the

impact of time on the proteins contained in secretion residue was shown to be efficient on body-fluid-contaminated fingermarks (i.e., saliva, vaginal fluid and urine) [213] – see section 3.1 for details.

<sup>[\*]</sup> Note: inferences about activity are expressed with regards to the presence or absence of contaminant proteins on ridges or valleys. Caution should be taken regarding such conclusions as it has been shown that the reality is much more complex, as described in section 3.2.15.

**Practice-oriented or case-related studies:** The processing of glassine stamp bags containing heroin was investigated [487] – see details below.

**Acronyms used:** **4-ATP** (4-aminothiophenol), **6-MAM** (6-monoacetylmorphine), **AB** (Amido Black), **Ab-AuNPs** (antibody linked to gold NPs), **AgLDI-MSI** (silver-assisted MALDI-MSI), **AY7** (Acid Yellow 7), **CA** (cyanoacrylate), **DESI** (desorption electrospray ionisation), **DFO** (1,8-diazafluoren-9-one), **FT-ICR** (Fourier-transform ion cyclotron resonance), **FTIR** (Fourier-transform infrared), **IND/Zn** (1,2-indanedione containing zinc chloride), **LA-ICP** (laser ablation inductively coupled plasma), **LC** (liquid chromatography), **LCV** (Leuco Crystal Violet), **MALDI** (matrix assisted laser desorption ionisation), **MS** (mass spectrometry), **MSI** (mass spectrometry combined with imaging), **NALDI** (nanostructure-assisted laser desorption ionisation), **NPs** (nanoparticles), **PD** (physical developer), **PDA** (polydopamine), **PDMS** (polydimethylsiloxane), **PETN** (pentaerythritol tetranitrate), **PS** (paper spray), **R6G** (Rhodamine 6G), **RDX** (1,3,5-trinitro-1,3,5-triazinane), **SERS** (surface-enhanced Raman spectroscopy), **SRS** (stimulated Raman scattering), **SRXRF** (synchrotron radiation X-ray fluorescence), **TATP** (triacetone triperoxide), **TNT** (trinitrotoluene), **ToF** (time-of-flight)

**Body fluid contamination** – The potential for operational use of Ab-AuNPs and aptamers to detect fingermarks has been assessed by Lam et al. [419]. The authors considered four donors, natural secretions and blood-contaminated fingermarks, depletion series (5), six non-porous (i.e., plastic Ziplock bags, gray shopping and black garbage bags, cling film, beverage can and plastic water bottle) and two semi-porous substrates (i.e., glossy magazine and cardboard), various aging times (up to 4.5 months), an extensive range of antibodies (17) and aptamers (7) fluorescently-tagged and combined to form a multiplex solution, and four conventional detection processes (i.e., CA ⇄ R6G or IND/Zn or AY7 or AB ⇄ PD) for comparison purposes. The multiplex approach was first optimized then compared to conventional methods. Overall, the conventional methods outperformed the multi-target immunogenic reagents, whose performance seemed to be more influenced by donor variability. The authors also assessed the possibility of introducing the multiplex solution in the conventional detection sequence (i.e., before CA, after CA and in replacement of PD). The multiplex solution was either detrimental to the subsequent techniques or offered no additional benefit. Overall, the authors concluded that immunogenic reagents cannot be considered a suitable alternative to conventional detection techniques. Lauzon and Chaurand evaluated the potential of AgLDI-MSI to detect the presence of exogenous compounds in contaminated fingermarks and its compatibility with conventional detection techniques [486]. The authors considered sebum-rich fingermarks spiked with drugs and blood-contaminated marks. As expected, the imaging of exogenous compounds was successful. The authors also assessed the performance of AgLDI-MSI after the application of conventional reagents (i.e., IND/Zn, AB, and LCV) on blood-contaminated marks. AB-processed and LCV-processed marks were successfully imaged through the heme and crystal violet groups, respectively. IND/Zn-processed marks were imaged by focusing on an un-identified

group, but a lack of sensitivity was noted.

**Processing of heroin-contaminated bags** – Barnes et al. looked for an alternative procedure to process glassine stamp bags containing heroin or fentanyl for fingerprints, the current one involving cutting the bags open and transferring the drug before processing them with NIN [487]. The authors looked for a process in which the drug is kept inside the bags during fingerprint detection. To reach that goal, they considered five donors (2 good, 2 moderate and 1 poor, based on NIN), freshly-deposited natural marks and four detection processes (i.e., magnetic powder, DFO, sprayed NIN, DFO ⇌ NIN). The questions considered by this study were threefold: (1) impact of the humidity chamber on the heroin contained in the bags, (2) performance of the techniques, and (3) mass added to the bags by the fingerprint detection techniques. A slight degradation of heroin into 6-MAM was observed, caused by exposure to humidity during the NIN process. This seems not to be an issue for the authors because Pennsylvania laws state that a powder containing any amount of heroin is illegal, but this could be an issue in other states or countries. Also, the powder was more difficult to manipulate as it stuck to the spatula. With regards to performance, magnetic powder was the most efficient technique. They somewhat observed that the fingerprints processed with magnetic powder were detected with a tonal reversal (i.e., light ridges on darker background), which was not explained. The authors argued that further studies may be required with regards to the best way to process the items, involving older marks and a clarified quality metrics. Finally, NIN and DFO ⇌ NIN resulted in a measurable mass increase of 0.0017 g and 0.0013 g, respectively (magnetic powder and DFO: 0.0004 g and 0.0003 g). However, these values were less than the tolerance level for uncertainty of the authors' laboratory (0.002 g).

### 3.2.17. C/Immersed items

Preliminary/Pilot studies: Functionalized carbon nanotubes applied to immersed stainless steel [410].

Practice-oriented or case-related studies: Using aluminium soda cans bearing fresh sebum-rich fingerprints and three different scenarios (i.e., sheltered and unsheltered winter environments, and forced insertion into snow), McCook et al. investigated the effect of snow on the recovery of fingerprints [488]. The following sequence was applied one month after the cans were recovered from snow: CA ⇌ dye staining (Ardrox or RAM) ⇌ black powder. Even if the contact with snow was shown to be detrimental to ridge details, encouraging results were obtained for all scenarios and contact times, with the forced insertion into snow being the most detrimental event.

Considering three non-porous substrates (i.e., glass, compact discs, knife blades) bearing fresh sebum-rich fingerprints and immersed in fresh and sea water for 1–10 days, Madkour et al. investigated the impact of immersion on fingerprint detection [489]. Once retrieved from water the items were dried before being processed (i.e., black powder, CA, or SPR). Promising results were obtained with all three techniques, with sea water and prolonged immersion time being the most detrimental conditions.

An empirical study investigated how water immersion could impact the detection of fingerprints by VMD [490]. Three non-porous substrates (i.e., white PE bags, transparent PVC, and glass) and immersion times from 1 h to one day were considered, the items being dried for 24 h before being processed. When compared to unwetted substrates, the immersion induced a modification of colour shades for VMD<sub>Ag</sub> and VMD<sub>St-Ag</sub> (all substrates) and of contrast for VMD<sub>Au/Zn</sub> on PE. Overall, the results were in accordance with previously-published

studies related to VMD applied to polymers and other non-porous substrates.

Available reviews: detection of fingerprints on wetted items [491].

Acronyms used: CA (cyanoacrylate), PE (polyethylene), PVC (polyvinylchloride), RAM (mix of Rhodamine 6G, Ardrox, and 7-[p-methoxybenzylamino]4-nitrobenzene-2-oxa-1,3-diazole), SPR (small particle reagent), VMD (vacuum metal deposition), VMD<sub>Ag</sub> (silver-based monometallic VMD), VMD<sub>Au/Zn</sub> (traditional gold-zinc VMD), VMD<sub>St-Ag</sub> (sterling silver-based monometallic VMD)

### 3.2.18. I/Photography and forensic light sources

Preliminary/Pilot studies: Geometrical compensation to flatten fingerprints captured on a curved surface [492], long-wave UV fluorescence of eccrine secretions induced by a 266 nm short-wave UV laser [493], autofluorescence of fingerprints induced by a shortwave UV-pulsed laser (exc. 230 or 280 nm) and imaged through time-resolved spectroscopy (obs. 280–530 nm, max. 440 nm for aged marks) [494], deflectometry combined with windowed Fourier transform analysis to image latent marks on specular surface [495], optical setup involving two light sources and a mirror to capture fingerprints left on curved items [496], polarization imaging detection of fingerprints by using active polarized light and multi-angle recording [497], short-wave UV fluorescence of sebum-rich secretions induced by a two-dimensional laser scanning system [498].

Lenses and filters: the combined use of a narrow bandpass filter centred at 560 nm (FF-1.0, Arrowhead Forensics) and a standard orange barrier filter (Coherent 1153747) allowed the observation in luminescence of unprocessed fingerprints when excited with a 532 nm laser (TracER, Coherent Inc.) or a Flare Plus 2505 nm LED (Rofin Forensic) [499]. This optical configuration also perfectly suits the observation of chemically-processed fingerprints (e.g., IND/Zn, R6G). In another study, the performance of the FF-1.0 filter (Arrowhead Forensics) and its combination with orange filters has been investigated using (un)processed marks left on various substrates [500]. Good performance was obtained overall with the use of 532 nm laser (TracER, Coherent Inc.) and with the combined use of FF-1.0 with the orange Exposed Curved Barrier (Arrowhead Forensics). Rimmasch emphasized the importance of using a focus test chart with a pattern (dot or grid) to ensure the absence of distortion with each used combination of camera and lens [501].

Long-wave UV reflection: the use of long-wave UV reflection (i.e., 315–400 nm) was reported as an optical method perfectly suiting the observation of items processed with CA, prior to the application of fluorescent dyes [502]. Compared to short-wave UV reflection (such as RUVIS), long-wave UV reflection is claimed to be safer for the practitioner and for touch DNA.

Dark adaptation: the importance for practitioners of adapting their eyes to the dark before looking for processed marks in luminescence has been emphasized by McMurchie et al. [503]. Using amino acid solutions, various characters, shapes and icons were printed on white paper which were then processed with DFO. On average, +16% additional patterns were recovered by the participants when darkness adaptation was carried out using a commercial device (i.e. Crime-lite Eye™, Foster + Freeman Ltd). It should be noted that participants required about 10 min to adapt their eyes to the dark, based on the device.

Available review: thorough review of ALS in the form of a landscape study covering optical phenomena, filters, trace observation and specificities of commercially-available ALS

[504], recent trends linked to the observation of fingermarks in luminescence or in the NIR domain [505,506].

**Acronyms used:** **ALS** (alternate light sources), **CA** (cyanoacrylate), **IND/Zn** ((1,2-indanedione containing zinc chloride), **NIR** (near-infrared), **R6G** (Rhodamine 6G), **RUVIS** (reflective ultraviolet imaging system), **UV** (ultraviolet)

### 3.2.19. I/Chemical imaging

**Note:** due to a strong overlap between the two topics, the articles dealing with chemical imaging applied to spiked fingermarks (e.g., drug or explosives) for contamination imaging purposes only are cited in section 3.2.16.

**Preliminary/Pilot studies:** SALDI-MSI ⇨ imaging of deprotonated fatty acids contained in sebum-rich fingermarks pre-processed by gold-silver sputtering [507]; Raman ⇨ SERS to image artificially-enriched fingermarks left on functionalized glass and further processed with antibody-functionalized gold NPs (i.e., anti-lysozyme, anti-human IgG and anti-cotinine) [398], hyperspectral SRS to image endogenous fatty acids contained in sebum-rich fingermarks and exogenous compounds (i.e., gun powder and benzoic acid) contained in spiked fingermarks [193]; other ⇨ LIBS to image ridge patterns and distinguish overlapping fingermarks through their content in ions [508], through their difference of ages [509] or by using a chemometric approach [510], DESI-MSI to image ridge pattern and provide information about the donor of the fingermark (e.g., such as gender, ethnicity, and age) through lipid profiling [185], HSI to estimate the age of blood-containing fingermarks through the absorption of haemoglobin in the visible range and the presence of the Soret peak at 415 nm [458], SKP and SEM/EPMA to image latent and VMD-processed fingermarks on metallic surfaces [434], synchrotron-based ATR-FTIR-FPA combined with confocal Raman microscopy to characterize secretion residue through spatial distribution of eccrine and sebaceous material [206], ToF-SIMS applied to flat metallic surfaces [435].

**MALDI-MSI:** The compatibility of AgLDI-MSI with conventional detection techniques was investigated [511] as well as the application of MALDI-MSI to case-related fingermarks [512] or to the new GBP polymer banknotes [427] – see details below. Silver sputtering combined with MALDI-MSI was applied to sebum-rich and flunitrazepam-spiked fingermarks [472]. MALDI-MSI was used to infer donor's lifestyle through the identification and imaging of exogenous compounds [217], to estimate the age of fingermarks through the differentiated diffusion of two classes of lipids [200], and to confirm LC-MS results about the degradation of lipids with time [211] – see section 3.1 for details. The ability of MALDI-MSI to analyse the composition of CA fuming marks was assessed [241]. Using compound intensities normalized to those of latent marks, the authors proved the compatibility of the techniques. With regards to the impact of the fuming process on fingermark composition, the authors found no evidence that the endogenous secretion compounds were chemically altered by the fuming process, but the detection of some exogenous compounds (cosmetics, explosives, drugs) was closely linked to their ionic nature and the matrix choice. MALDI-MSI was also used to get a better understanding of the CA fuming process [242] – see section 3.2.3 for details. Electrospray of TiO<sub>2</sub> NPs was proposed to optically detect sebum-rich and hand-cream-spiked fingermarks, and to be used as a matrix for MALDI-MSI analysis [513]. **Available reviews:** overview of the MALDI-MS(I) protocols applied to fingermark profiling and imaging, including fingermark preparation and matrix application [514]. Review of

spectroscopic-based imaging techniques (e.g., FTIR, SERS) applied to traces of forensic interest, including fingermarks [515]. Extensive review of the use of MALDI MS(I) for fingermark analysis, including: detection and mapping of endogenous or exogenous compounds, compatibility with fingermark detection techniques, and operational capabilities [516]. Advances [517] and critical review [385] regarding the application of chemical imaging to fingermarks.

**Acronyms used:** **AgLDI-MSI** (silver-assisted MALDI-MSI), **ATR** (attenuated total reflectance), **BPS** (black powder suspension), **BY40** (Basic Yellow 40), **CA** (cyanoacrylate), **DESI** (desorption electrospray ionisation), **EPMA** (electron probe micro-analyser), **FPA** (focal plane array), **FTIR** (Fourier-transform infrared), **HSI** (hyperspectral imaging), **IgG** (immunoglobulin G), **IND/Zn** (1,2-indanedione containing zinc chloride), **LC** (liquid chromatography), **LIBS** (laser-induced breakdown spectroscopy), **MALDI** (matrix assisted laser desorption ionisation), **MS** (mass spectrometry), **MSI** (mass spectrometry combined with imaging), **NIN** (ninhydrin), **NPs** (nanoparticles), **ORO** (Oil Red O), **PD** (physical developer), **R6G** (Rhodamine 6G), **SALDI** (surface assisted laser desorption ionisation), **SEM** (scanning electron microscope), **SERS** (surface-enhanced Raman spectroscopy), **SIMS** (secondary ion mass spectroscopy), **SKP** (scanning Kelvin probe), **SRS** (stimulated Raman scattering), **ToF** (time-of-flight), **VMD** (vacuum metal deposition)

**MALDI-MSI** – The compatibility of AgLDI-MSI with conventional detection techniques (i.e., IND/Zn, NIN, ORO, PD, CA, dry powders) was investigated [511]. Using sebum-rich fingermarks left on various substrates (e.g., paper, cigarette cardboard, plastic bags, adhesive lifter post-dusting), the authors tested the application of AgLDI-MSI after each single detection technique and in the following sequences: IND/Zn ⇨ NIN ⇨ ORO ⇨ AgLDI-MSI, and CA ⇨ R6G ⇨ AgLDI-MSI. Most of the techniques showed good compatibility with AgLDI-MSI, with the detection of several compounds such as (unsaturated) fatty acids, cholesterol, squalene, and cosmetics, to cite a few. It was still possible to detect compounds when AgLDI-MSI was applied last in a sequence (e.g., fatty acids, squalene, wax esters and some triglycerides for paper). The authors also made the following observations: (1) many lipids were removed by the application of ORO, (2) on paper, it seems that the application of the detection techniques (with the exception of PD) cause a slight diffusion of the lipids which induces a degradation of the imaging quality. The opportunity to apply MALDI-MSI to fingermarks linked to four cases has allowed assessing the feasibility of the approach and identifying the benefits and limitations of the technique [512]. The four marks were linked to three different cases (i.e., cannabis farm, murder and harassment), and were processed by conventional detection techniques beforehand (i.e., dry powders or CA ⇨ BY40). One was analysed on the item (i.e., plastic bag processed with CA ⇨ BY40) whereas the three others were analysed after being dusted and tape lifted. In the cannabis farm case, ridge patterns were imaged using non-forensically relevant compounds such as antibacterial agents (toilette products). In terms of ridge details, MALDI-MSI performance was inferior compared to the conventional detection techniques applied beforehand. However, cocaine traces were identified in the fingermarks linked to the harassment case and to the cannabis farm, which could constitute forensically relevant information to provide. Regarding the latter, no THC was identified (most certainly due to a low ionisation capability). MALDI-MSI was also applied to the new £5 polymer banknotes [427]. In their study, Scotcher and Bradshaw described the sample preparation, the imaging capabilities with regards to the illustrated background, the performance with regards to depletion series (8) or to contaminants (i.e., fingertips spiked with

cocaine in solution), and the compatibility with conventional detection techniques (i.e., CA and Wetwop™ BPS). Overall, MALDI-MSI behaves as expected from previous studies. In terms of contrast, MALDI-MSI was unaffected by the banknote security features except for the Blenheim Maze illustration and the “£5” characters, which resulted in signal suppression for the fingerprint. The study should however be considered as a proof-of-concept given that only one donor was used and that the banknote samples were cut for analysis (without explanation).

### 3.2.20. O/Touch DNA

**Note:** The aim of this report is not to extensively cover the question of genetic material contained in secretion residue (i.e., touch DNA). Only the studies involving fingerprint detection techniques in addition to touch DNA were cited below.

**Fingerprint detection ⇔ DNA recovery:** The effect of fpNatural 1™ on the recovery of DNA was assessed [382]. No significant effect on the quantity or quality of DNA was noted with fingerprints left on glass slides. Similarly, the impact of black and magnetic powders (Sirchie®) was studied with fingerprints left on glass slides [518]. Magnetic powder resulted in the least alleles recovered (35%) compared to black powder (66%). Brush cross-contamination issues and decontamination procedures were addressed through different scenarios involving dried saliva and touch DNA [383]. DNA transfer was demonstrated, confirming the need to either decontaminate used brushes (squirrel hair) using either Virkon 5% or sodium hypochlorite 1%, or dispose of used brushes (fiberglass, which suffered from the decontamination procedure). The recovery of residual touch DNA on a surface after tape-lifting (post-dusting) is recommended as it can contain a substantial amount of genetic material [519,520]. The impact of fingerprint detection techniques (i.e., CA, CA ⇔ dye, CA ⇔ powder, IND/Zn, NIN, PD, aluminium and magnetic powders) on the recovery of DNA was addressed [521]. Briefly, CA, IND/Zn, NIN, and aluminium powder showed no or limited impact to DNA recovery, while magnetic powder hindered DNA recovery and PD led to poor quality profiles. Considering four dusting powders (i.e., black, aluminium, and magnetic black and white) and three lifting processes (i.e., tape, gel lifter and a silicon-based casting compound), Subhani et al. showed that most “dusting powder/lifter” combinations lead to the successful recovery of DNA [522].

The impact of one-step CA fuming has been addressed in two articles [244,245] – see details below.

**DNA recovery ⇔ fingerprint detection:** A study aimed at assessing the impact of DNA recovery prior to fingerprint detection was carried out [523]. The following were considered: three kinds of secretions (i.e., natural, sebum-rich and eccrine-rich), three donors, five substrates (i.e., white office paper, glass slides, aluminium, black textured PP, and varnished wood), four aging times (i.e., 4 h, 2 days, 1 and 4 weeks), five DNA recovery methods (i.e., dry cotton swab, wet cotton swab, flocked swab with nylon tip, gel lift, and tape lift), and three fingerprint detection processes (i.e., aluminium powder, NIN, CA ⇔ BY40). Unsurprisingly, all the DNA recovery methods had a detrimental impact on ridge pattern quality, especially on glass. The least damaging methods were dry swabbing, flocked swabs and gel lifting applied to textured PP, varnished wood and paper. The most damaging methods were wet swabbing and tape lifting. In another study, the use of hydrogels from dextran-methacrylate solutions was proposed to collect hydrophilic compounds (e.g., amino acids or DNA) from a fingerprint [219]. The DNA recovery yield ranged from 20 to 60% (quantity) compared to cotton swabs. The authors also showed that the surface can still be

processed with fingerprint detection techniques (CA) to detect ridge patterns, although with slight degradation of fine details. **DNA recovery processes:** The localized application of solvent (i.e., un-du or chloroform) followed by swabbing (COPAN 4N6FLOQSwabs) was proposed to collect DNA from BPS-processed fingerprints (Wetwop™) on the adhesive side of duct tapes [524]. The successive application of acetone and water followed by swabbing (cotton swabs) was proposed to recover DNA on the adhesive side of electrical tapes [525]. Mini-tape lifting (Scenesafe FAST™) and scraping methods were recommended, over dry and wet swabbing, to recover DNA from clothing [526]. The use of direct PCR swabs (omitting the extraction step) was proposed to generate the genetic profiling of fingerprints that were swabbed post-dusting [527]. An optimized workflow was proposed to process fingerprints that were previously dusted, tape-lifted and left for four weeks before DNA recovery [528], direct cutting and double swabbing were shown to be the best process.

**DNA vs ridge pattern detection:** In an attempt to promote informed decisions between fingerprint detection and touch DNA collection, Kartasinska and Tomaszewski reviewed 122 caseworks recorded between 2010 and 2013 in Poland [529], which represented 514 exhibits processed for fingerprints and/or for touch DNA. In their paper, the authors discussed the effectiveness of each process (positive results for fingerprint detection and touch DNA analysis in 27% and 60% cases, respectively), the sequence of collection, the possibility of obtaining successful DNA profiling after fingerprint detection (positive “identification” in 40% of such cases), the usefulness of conducting touch DNA collection after an inconclusive fingerprint detection process, and the impact of the type and nature of the item to be processed (specific cases). The autofluorescence of fingerprints (excited at 365 nm) was proposed to estimate the quantity/quality of genetic material available [530]. Given that no correlation was found, this approach cannot be used as a selection tool for DNA profiling. Considering three substrates (i.e., glass, PE bags and white office paper), the impact of deposition pressure on fingerprint morphological characteristics (ridge pattern quality and area) and on the quantity/quality of DNA was assessed [531]. Overall, fingerprint quality followed a curve that decreased at higher pressure values, while DNA quantity/quality increased with the pressure. The compatibility between genetic material enhancement (using a nucleic acid dye) and ridge pattern detection was assessed [532]. The question of recovering fingerprints or touch DNA from cartridge cases has been addressed [444] – see section 3.2.11 for details. Using Diamond™ Nucleic Acid Dye (Promega) and six conventional detection techniques (i.e., five dusting powders and CA, using a home-made fuming cabinet without humidity control), the authors assessed the impact of DNA staining on the subsequent ridge pattern detection, and vice versa. They concluded that DNA staining should be carried out first, followed by fingerprint detection. [Note: given that the application of the nucleic acid dye was performed locally, using 10 µl of solution directly applied on fingerprints whose positions were already known, this approach is currently not viable and complementary studies are consequently required.]

**Acronyms used:** **BPS** (black powder suspension), **BY40** (Basic Yellow 40), **CA** (cyanoacrylate), **IND/Zn** (1,2-indanedione containing zinc chloride), **NIN** (ninhydrin), **PD** (physical developer), **PE** (polyethylene), **PP** (polypropylene), **R6G** (Rhodamine 6G)

**Impact of one-step luminescent CA** – The development of one-step CA fuming processes has raised the question of their impact on subsequent DNA profiling compared to conventional CA [245]. To

answer this question, Khuu et al. considered four donors, natural and DNA-spiked (saliva) fingerprints left on glass and aged for two weeks, two one-step CA processes (i.e., PolyCyano UV from Foster + Freeman and Lumicyano™ from Crime Science Technology) and a conventional fuming process (i.e., CA ⇨ R6G). Overall, all CA processes caused DNA degradation. Nevertheless, Lumicyano™ was shown to have the same impact on DNA as the conventional CA process. In contrast, the impact of PolyCyano UV was shown to be more significant in terms of allele drop outs. Confirmatory studies are required, including an increased pool of donors and various substrates or aging times. Risoluti et al. also assessed the effect of a one-step CA process (7.7% Lumicyano) on subsequent DNA analysis [244]. They concluded that Lumicyano is compatible with DNA analysis (extraction and amplification), but the authors obtained mixed results in terms of profile quality: uninterpretable (mostly obtained with aged marks), clean (mostly obtained with fresh marks), and mixtures.

### 3.2.21. O/Miscellaneous detection techniques and research topics

Numerous detection techniques or research topics don't fit in the previous categories and are consequently cited in this section.

#### Preliminary/Pilot studies:

- sputtering of gold-silver alloys to optically detect sebum-rich fingerprints [507]. It has to be noted that the technique appears similar to VMD (e.g., colour tones, degradation with time of ridge details with silver) and is further combined with chemical imaging (fatty acids mapping).
- self-triggered alarm system using a triboelectric nanosensor and nitrocellulose membrane as substrate for fingerprints upon contact [533];
- sulfonated poly (diphenylacetylene) polymer in solution interacting with sweat components and exhibiting a “turn-on” emission mode [534];
- PDMS support covered by a PDA thin film then applied on a fingerprint: transfer of PDA into sweat and ridge pattern visualization through PDA-catalysed electroless silver deposition (positive image on the substrate, negative image on the PDMS support) [535];
- follow-up of the above study: PDMS support covered by a PDA thin film and a silver layer then applied on a fingerprint to allow optical detection and Raman chemical imaging [473];
- CTF-developed fingerprints combined with transmission-/reflection-mode multiwavelength digital holography [536];
- use of an AIE-based tetraphenylethene-based dye [537], conjugated polyelectrolyte [538], diphenylpyrimidinone derivatives [539] or acridinediones [540] to detect sebum-rich marks on various substrates;
- p-C1-PDPA film taking advantage of swelling-induced emission enhancement to detect sebum-rich marks on non-porous substrates [541];
- two-step detection of sebum-rich fingerprints involving the lifting of secretion residue by a hydrophilic cellulose membrane followed by dye staining of the membrane (the sebum-rich secretions acting as a mask) [542] (note: this study has been further reported by Ref. [543]);
- use of paraffin candle soot to detect sebum-rich fingerprints on various substrates [544];
- two-step detection of sebum-rich fingerprints involving the lipophilic adsorption of nitric oxide (NO) followed by the application of 1,2-diaminoanthraquinone [545];
- sublimation of lanthanide complexes to detect fingerprints on non-porous substrates [546];
- use of lysozyme-binding aptamers combined with a lanthanide-based carboxymethyl nanocellulose hydrogel

[547] or embedded in two DNA strands with a G-quadruplex/NMM complex [548] to detect (fresh sebum-rich) fingerprints on various substrates;

- use of electrolytes in aqueous solutions to detect marks on various substrates [549];
- two-step detection of fingerprints involving the transfer of secretion residue to a nanofibrillated cellulose membrane doped with fluorescent C-dots, followed by CA fuming and dye-staining (using super-paramagnetic iron oxide NPs; application mode not specified) [550];
- metal-free room-temperature phosphorescent materials synthesized from modified ureidopyrimidinone units [551].

**Various (challenging) substrates:** The recovery of fingerprints on latex gloves (inner side) can be carried out by using NIN [HFE] (protocol: 10 s immersion followed by 1–2 h drying in a fume hood) [552]. The performance of NIN [HFE] was higher than the non-porous alternatives: BPS or CA ⇨ CV. Black carbon paper can be processed using the following sequence: CA ⇨ IND/Zn (or NIN) ⇨ RAM [553]. As emphasized by the authors, black carbon paper is sometimes used as controlled substance packaging because of its alleged ability to interfere with older X-ray scanners. Also, only one brand of carbon paper (i.e., Staples) was used in this preliminary study and further studies are required to assess the efficiency of the sequence on other brands. Fingerprints can be recovered from chalk sticks by activated charcoal dusting (up to 1-h-old marks), charcoal powder-based BPS (up to 3-day-old marks) or iodine fuming (up to 1-h-old marks) [554].

**MOF:** the use of MOF to detect fingerprints on various substrates was further investigated by two teams [555,556] – see details below.

**Secretion residue – substrate interactions:** Some interesting non-forensic studies addressed the interactions between secretion residue and the underlying surface. These could be of interest to get a better understanding of the behaviour of fingerprints left on some surfaces (e.g., anti-fingerprint surfaces, touch screens) and are consequently included in this report. Using artificial sebum and a standard deposition protocol (e.g., quantity of matter, pressure, surface area, deposition time), Stoehr et al. emphasized the resistance to cleaning of secretion residue, which undergoes a shear banding phenomenon [557]. In a study aiming at proposing an anti-fingerprint coating for stainless steel, Kesmez et al. investigated the impact of surface roughness on the ability for a fingertip (contaminated with a mix of artificial sebum and sweat) to transfer material [558]. The combination of a mesoporous layer embedding enzymatic molecules (e.g., lipases) was proposed as a new anti-fingerprint coating [559]. Using stamped artificial sebum, the authors illustrated how the secretion residues were actually degraded in a couple of days. Forchelet and Bécue investigated the impact of anti-fingerprint coatings (i.e., liquid, plastic films and glass) on the visibility of latent fingerprints and on conventional detection techniques (i.e., CA, SPR, VMD<sub>Au/Zn</sub>) [560]. The main conclusions were that anti-fingerprint coatings do not prevent the deposition of secretion residue, that they could improve the preservation and observation of latent fingerprints (better ridge details), and that they do not hinder the application of detection techniques (limited impact). In fact, most of the anti-fingerprint coatings seem to optimize the “easy-to-clean” properties instead of hindering secretion residue deposition or visibility. Luda et al. proposed an artificial emulsion (mix of 95% w/w sweat-related and 5% w/w sebum-related chemicals) stamped on anti-fingerprint surfaces to investigate the surface properties that are correlated with the visibility of the secretion residue [561]. While secretion residue was shown to impact the

transmittance and wettability of the underlying substrate, its visibility was correlated to five surface properties: roughness profile, variation of gloss, haze, luminance, and diffuse reflectance.

**Practice-oriented or case-related studies:** Jabbel et al. addressed the question of the feasibility of a secondary transfer (of secretion residue) from a non-porous substrate (glass) to a porous one (office paper) [562]. Considering natural and sebum-rich fingermarks, different transfer conditions (i.e., contact pressure and time, fingermark age) and various detection techniques (i.e., IND/Zn, NIN, ORO, aqueous Nile blue, SMD II), they showed that secondary transfer is possible only when the secretion residue comes in contact with the second surface shortly after its deposition (no transfer was detected for 1-day-old fingermarks and older). The clarity of the transferred fingermarks increased with the pressure (starting from 500g) and the contact time (starting from 2 h). They also hypothesised that the transfer involves water-soluble compounds as poor results were obtained with techniques other than IND/Zn and NIN.

**Collaborative exercises:** The output of the latest EFP-WG collaborative testing were published, covering the following years: from 2006 to 2014 [81], 2015 [79] and 2016 [80]. The EFP-WG collaborative testing covers three main fields: detection, imaging, and comparison (identification). In their articles, the authors provided an overview of the tests, discussed the issues that were identified (e.g., test design and delivery, incomplete reported results, performance evaluation, administrative limitations) and the lessons that were learned (e.g., need for clear instructions, reproducibility of the samples, need for specific workshops).

**Miscellaneous:**

- the impact of using hand sanitizers on the recovery of fingermarks has been investigated by Chadwick et al. [563]. Non-alcoholic hand sanitizers (e.g. Deb® or EcoHydra®) were shown to significantly improve the quality of fingermarks detected by IND/Zn or NIN, and marginally magnetic powder. The authors explained these results by the presence of benzalkonium chloride (active ingredient) and by the increased moisture content on the ridges readily after the application of the product;
- the creation of a publicly available collection of high-resolution fingermarks of different ages (i.e., 1-day-old to 3-year-old) was described [564];
- an extensive study focused on the factors that influence fingermark deposition and detection [565] – see details below;
- a group of young postdoctoral researchers published their shared interest about forensic science challenges, including fingermark detection [566];
- in an attempt to help determine the directionality of a fingertip upon contact with a surface, the impact of lateral movement on paper and porcelain has been studied [146] – see details below;
- the use of black gelatin lifters (Gellifters, BVDA, NL) was proposed to discreetly recover fingermarks from items (e.g., covert operation) [567]. Episcopic coaxial illumination was recommended to observe the lifted marks. Unsurprisingly, the best results were obtained with fresh (<24 h) sebum-rich fingermarks (as opposed to natural marks) left on smooth non-porous surfaces (as opposed to semi-porous ones). Poor results were obtained with porous substrates. The authors also noted that the protective film should not be placed back on the lift as it decreases the quality of the observed ridges (if it is placed back, the observation should be performed in less than 1 h).

- the impact of ionizing radiation on various traces of forensic interest, including fingermarks, was proposed as a literature review combined with post-irradiation experiments [568]. Regarding fingermarks, sebum-rich and eccrine secretions were left on aluminium and office paper before being exposed to increasing doses of radiation and processed with CA⇨RAM (aluminium samples) or DFO⇨NIN (paper samples). The authors showed that fingermarks on paper and metal were impacted by the four kinds of radiation, even at low doses (i.e., 0.0005 kGy beta, 0.002 kGy neutron, 0.12 kGy alpha, and 0.5 kGy gamma), but that many fingermarks remain of value for comparison purposes. Further experiments are required to confirm some observations and fill some methodological gaps.

**Available reviews:** recovery of fingermarks from post-blast debris [569].

**Acronyms used:** **AIE** (aggregation-induced emission), **AY7** (Acid Yellow 7), **BPS** (black powder suspension), **BSA** (bovine serum albumin), **BY40** (Basic Yellow 40), **C-dots** (carbon dots), **CA** (cyanoacrylate), **CTF** (columnar thin film), **CV** (crystal violet), **DAQ** (1,2-diaminoanthraquinone), **EFP-WG** (ENFSI Fingerprint Working Group), **ENFSI** (European Network of Forensic Science Institutes), **GV** (Gentian Violet), **IND/Zn** (1,2-indanedione containing zinc chloride), **MBD** (7-[p-methoxybenzylamino]-4-nitrobenz-2-oxa-1,3-diazole), **MOF** (metal organic framework), **NIN** (ninhydrin), **NIN[HFE]** (ninhydrin formulation prepared with HFE-7100 as solvent carrier), **NMM** (N-methyl mesoporphyrin IX), **NPs** (nanoparticles), **ORO** (Oil Red O), **p-C1-PDPA** (poly [1-phenyl-2-p-[trimethylsilyl]phenylacetylene]), **PDA** (polydopamine), **PDMS** (polydimethylsiloxane), **PE** (polyethylene), **PP** (polypropylene), **R6G** (Rhodamine 6G), **RAM** (mix of R6G, Ardrex and MBD), **SMD** (single metal deposition), **SPR** (small particle reagent), **VMD<sub>Ag</sub>** (silver-based monometallic VMD), **VMD<sub>Au/Zn</sub>** (traditional gold-zinc vacuum metal deposition)

**Factors influencing fingermark deposition** – In an extensive study involving 14'000 fingermarks, Chadwick et al. provided information about the mechanisms involved in fingermark deposition (e.g., substrate, secretion residue composition, donor, aging time, depletion series) and their impact on fingermark quality [565]. To reach that goal, the authors considered four substrates (i.e., office premium paper, office recycled paper, PP sleeves and PE Ziplock bags), natural fingermarks left by five donors, depletions series of four marks, two aging times (i.e., 3 and 7 days), and two detection processes (i.e., CA ⇨ R6G and IND/Zn). With regards to the influence of the substrate, the fingermarks left on porous substrates were more likely to be detected and were of better quality compared to those left on non-porous substrates. This could be explained by the interactions between the secretion residue and the substrate or by the higher efficiency of IND/Zn compared to CA ⇨ R6G. They also assessed the ratio of non-detected marks to be of 11.7% overall, with the highest score on non-porous substrates (i.e., 20%) compared to porous substrates (3%). It should however be kept in mind that fresh marks (3- and 7-day-old) were considered in this study. With regards to the donor, they confirmed the fact that the quality of marks decreases along a depletion series. Donor intra- and inter-variabilities were emphasized, with the authors expressing the need for more fingermarks from the same donor when designing a study focused on detection. Variations of detection performance were observed between the fingers (the thumb leading to highest quality marks as opposed to the little one) and between left and right hands (at the finger level). Further studies are required to confirm these trends.

**Fingertip movement upon contact** – Tate et al. conducted a study aimed at providing information related to the movement of a

fingertip upon contact with a surface [146]. In their introduction, the authors provided useful information about the structure of paper matrices and coatings. They also discussed the way practitioners usually characterize a substrate in terms of porosity (linked to air flow) and not absorbency (linked to liquid uptake). In terms of methodology, the authors considered two substrates (*i.e.*, white office paper and white ceramic tile), two kinds of secretion residue (*i.e.*, sebum-rich marks and artificial secretions using the Sirchie Latent Print Standards pad), and one linear movement upon contact (4 cm proximal translation) with varying contact times before and after the movement (*i.e.*, from 1 s to 4 s). Porous and non-porous samples were processed by NIN and black powder dusting, respectively. On the porous substrate, the movement resulted in two fingermarks separated by a drag smear without ridge details. The “starting” fingermarks appeared darker and provided good ridge details. The “finishing” fingermarks appeared lighter, provided ridge details and were surrounded by a corona, that was darker and more defined than the core area. On the non-porous substrate, the movement resulted in one fingermark (the “finishing” one) and a drag smear without ridge details. The “starting” fingermarks were obliterated by the movement and contained no ridge details, at the exception of an incomplete corona. The “finishing” fingermarks were of overall good quality, with slight ridge details compression and expansion above and below the core, respectively.

**MOF** – The use of MOF to detect fingermarks on non-porous substrates (which has resulted in a mediatic buzz during Fall 2015) has been challenged by two teams [555,556]. As a reminder, MOF are lanthanide-based fluorescent crystals that could self-assemble in an aqueous environment, especially in the presence of inducing agents such as proteins and amino acids. In their study, Moret et al. considered seven types of secretion residue (*i.e.*, natural, sebum-rich, BSA- and lysozyme-enriched, blood and semen-contaminated), eight donors, depletion series (3), five substrates (*i.e.*, glass, sandwich and black garbage bags, aluminium, silver duct tape), three application protocols, and three detection processes for comparison (*i.e.*, CA ⇔ R6G, AY7 and luminescent SPR). Overall, Terbium-based MOF were less efficient than conventional detection techniques. The authors nevertheless emphasized the possibility of using MOF as a luminescent SPR if further optimization studies are carried out. In their study, de Jong et al. considered using Terbium- and Europium-based MOF as a stand-alone technique and as post-CA dye. To reach that goal, they considered six donors, depletion series (10), three substrates (*i.e.*, glass, aluminium, transparent tape) and two detection processes for comparison (*i.e.*, CA ⇔ BY40 and GV). Overall, the performance of MOF to detect fingermarks on non-porous substrates was poor. The authors nevertheless emphasized the possibility of using MOF as post-CA stains or as an alternative to GV to process the adhesive side of tapes. However, further studies are required, encompassing optimization of the protocol.

#### 4. Other body marks

Cheiloscopy in forensic science still finds some space in the forensic literature [570–572]. The development of automatic techniques for pattern matching [573–575] are promising. This is the type of systematic research that is required to go beyond the sole and simplistic argument of “uniqueness” and meet the requirements expected by PCAST.

Similarly, the anatomy of the external ear received some attention by researchers [576–579], some insisting on the uniqueness of the organ [579] but the most promising lines of inquiry are linked to the use of the ear as a biometric system [580–584], including age and gender estimation [577]. We are not

aware of research on earprints or earmarks, the most recent efforts being focused on the external organ.

Research in barefoot impressions concentrated on the variability observed in the dimensions of marks when jumping on surfaces [585,586], and differences between dynamic bare and sock-clad footprints [587]. An overview of the different methods and indices that are being used to evaluate footprints for comparison and identification purposes is due to Mukhra et al. [588]. We are far from the type of extensive research that would be needed to satisfy the strong requirements set by PCAST.

#### 5. Miscellaneous and case reports

The importance of recording and assessing the location of marks and their relationship with the alleged activities cannot be overstated. Bunter [589] presented a case where the location of the marks on a gate allowed confirmation of the allegations of the defendant who left these marks innocently 10 years before the incident under investigation. Kowalski [590] gives very informative case examples (involving firearms or knives) where the location of marks is decisive to understanding past actions.

The interpretation of marks in blood has always been challenging when the issue is to assess whether the mark was made by bloody friction ridge skin on a clean surface or by a clean area of friction ridge skin on a surface contaminated with blood. Geller et al. gave, using three models, very useful advice on how to document and analyse such marks on non-porous surfaces [463].

Bunter [591] in a short communication to magistrates presented a few cases of exaggerated fingerprint identification evidence and misinterpretation.

The postmortem recovery of fingerprints from corpses has always been a challenge. Novel or adapted techniques from mummified remains have been proposed [592,593].

Eldridge reported on a case of close-non match between siblings [594]. Sellenraad reported on a case of fingerprint forgeries observed on checks [595]. A simple superimposition allowed the detection of the patent prints.

Cases of laterally reversed prints are well-known to practitioners when adhesive surfaces or stamps are involved. These transfers of residue can also occur without an identified primary source such as a piece of tape [235]. Secondary transferred marks can be very clear to the point of not being distinguishable from the primary source based on their fingerprint features, apart from being mirror images [562].

It is worth reiterating that the quality of known impressions remains a decisive factor in the ability to exploit fingermarks [596] and the risk posed by livescan systems to generate artefacts in the form of features that could be confused with scars, creases [102] or additional ridges [597].

We note a case involving a palm mark detected on a notoriously difficult surface: a cartridge [598].

And finally, a case of a fingerprint pattern with three deltas due to an abnormal development of the right thumb of the subject [599].

#### Disclaimer

This is a republication in journal form of a conference proceeding that was produced for the 19th Interpol Forensic Science Managers Symposium in 2019 and was originally published online at the Interpol website: <https://www.interpol.int/content/download/14458/file/Interpol%20Review%20Papers%202019.pdf>. The publication process of this was coordinated for the Symposium by the Interpol Organizing Committee and the proceeding was not individually commissioned or externally reviewed by the journal.

The article provides a summation of published literature from the previous 3 years (2016–2019) in the field of fingerprints and other body impressions and does not contain any original, experimental data. Any opinions expressed are those solely of the authors and do not necessarily represent those of their agencies, institutions, governments, Interpol, or the journal.

### Declaration of Competing Interests

The authors have no competing interests to declare.

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