

FINGERMARKS AND OTHER IMPRESSIONS – A REVIEW

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Nicole Egli, PhD, Sébastien Moret, MSc, Andy Bécue, PhD and Christophe Champod, PhD



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Corresponding author:
Prof. Christophe Champod
Ecole de Sciences Criminelles / Institut de Police Scientifique
Faculté de Droit et des Sciences Criminelles
Quartier Sorge / Batochime
Université de Lausanne
CH-1015 Lausanne-Dorigny, Switzerland
e-mail: Christophe.Champod@unil.ch
<http://www.unil.ch/esc>

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1 Introduction

The purpose of this paper is to provide an overview of the papers dealing with fingerprints and other impressions that have been published between August 2010 and June 2013. We tried to offer an extensive coverage of the published sources (mainly in English), but remain conscious that exhaustiveness is not possible. The reader will realise that the area is very active and counts with more than 470 publications. We cover here both matters in relation to the detection of marks (mainly fingermarks) and matters associated with the forensic identification process.

In introduction, we would like to highlight some important books and reports that bring an important contribution to the field and can be used as key references:

- The Scientific Working Group on Friction Ridge Analysis, Study and Technology (SWGFAST) has published in 2011 a fingerprint sourcebook available online¹ (1). This book covers many subjects in the fingerprint individualisation process. There are chapters on history; anatomy and physiology; morphogenesis; the recording of exemplars; classification systems; automated identification systems; fingerprint detection, preservation, examination methodology; documentation; equipment; quality assurance; the interaction with the law; research on individualization; abilities and vulnerabilities in this area.
- The Home Office Centre for Applied Science and Technology (CAST) published in 2012 a comprehensive source book dealing with the full range of fingermark detection techniques (2). It is an essential reference for any laboratory, especially with the prospect of the generalised introduction of accreditation under ISO 17025. It is also available online².
- The *Advances in Fingerprint Technology* is now in its third edition (3). Ramotowski edited a rich and exhaustive volume covering the most advanced topics from the full spectrum of detection techniques to statistical modelling and digital imaging.
- A extensive report has been written by a group of experts convened under the NIST (National Institute of Standard and Technology) and addresses human factors in latent print analysis (4). The report is also available online³. It not only analyses current practices and their contribution to errors, but also investigates how to reduce error and how to implement these solutions practically. Report writing and documentation is as much part of this report as pre-trial communications, working conditions (lighting, workstations, for example), education and training, and finally the role of management.
- After years of dispute in Scotland, the report of the Fingerprint Inquiry – a judicial inquiry devoted to the mis-identification of both Shirley McKie and David Asbury – has been published (5), also available online⁴. The report gives a set of

¹ <http://nij.gov/pubs-sum/225320.htm>

² <https://www.gov.uk/government/publications/fingerprint-source-book>

³ http://www.nist.gov/manuscript-publication-search.cfm?pub_id=910745

⁴ <http://www.thefingerprintinquiryscotland.org.uk/inquiry/21.html>

recommendations (86 in total) that should be considered carefully by all laboratories. Among them, we note selectively the following:

Recommendation 1: Fingerprint evidence should be recognised as opinion evidence, not fact, and those involved in the criminal justice system need to assess it as such on its merits.

Recommendation 3: Examiners should discontinue reporting conclusions on identification or exclusion with a claim to 100% certainty or on any other basis suggesting that fingerprint evidence is infallible.

Recommendation 9: Features on which examiners rely should be demonstrable to a lay person with normal eye sight as observable in the mark.

Recommendation 53: Subject to any requirement under ISO 17025 and recommendations 50 and 51, note-taking as to the detail found on analysis and the process of comparison, though not mandatory, should become the general practice for all fingerprint comparison work.

Recommendation 66: Before a finding of 'unable to exclude' is led in evidence, careful consideration will require to be given to (a) the types of mark for which such a finding is meaningful and (b) the proper interpretation of the finding. An examiner led in evidence to support such a finding will require to give a careful explanation of its limitations.

In our last report (6), we reported on the increased scrutiny both by the courts and by commentators or scholars on the way fingerprint evidence was admitted and presented. During this reviewing period, we noticed a decrease of the number of challenges in court (e.g. *Daubert* or *Frye* hearings). However, a few cases raised important issues both in England and Wales and in the United States of America.

In *R. v. Smith* (7), the court of Appeal of England and Wales quashed a conviction and made general observations regarding the provision of fingerprint evidence. The court was astonished by the absence of contemporaneous notes taken during the examination process, stating that “No competent forensic scientist in other areas of forensic science these days would conduct an examination without keeping detailed notes of his examination and the reasons for his conclusions.” In relation to the reports produced, the court’s decision stressed that: “The quality of the reports provided by the Nottinghamshire Fingerprint Bureau for the trial reflected standards that existed in other areas of forensic science some years ago and not the vastly improved standards expected in contemporary forensic science.” This case is echoing the issues raised in the Fingerprint Inquiry in Scotland and has triggered the UK forensic science regulator to re-think quality standard in this area (8).

In *State of Minnesota v. Terrell Matthew Dixon* (9), the court after hearing highly recognized experts from both parties ruled that the State met its burden of demonstrating that the ACE-V method of friction-ridge-print analysis is widely accepted as reliable by experts in the field and that a fingerprint expert may be allowed to testify that she framed her identification opinion “to a reasonable scientific certainty.”

In *United States of America v. Clacy Watson Herrera* (10), the court went as far as suggesting that fingerprint expert offering opinion regarding sources is akin to an art expert or similar to eyewitness testimony. The court went on saying “Matching evidence of the kinds that we’ve just described, including fingerprint evidence, is less rigorous than the kind of scientific

matching involved in DNA evidence;” and recognized that “evidence doesn’t have to be infallible to be probative” and hence declared to be admissible.

In *United States of America v. John Charles McCluskey* (11), the court concluded “that the fingerprint identification testimony, while perhaps not “scientific,” is sufficiently reliable to be admitted into evidence at trial”, but the expert “will not be permitted to testify that any individual is the source of a particular print “to the exclusion of all others,” or that she is “100% certain” about an identification, or any variant thereof. There simply is no evidence in the record to support such a conclusion. To the contrary, the National Research Council, the FBI, and SWGFAST have all recognized the lack of scientific basis for such testimony and have advised against permitting examiners to express opinions to this level of certainty. Such a conclusion lacks a reliable scientific basis.”

These few US cases testify to the developing attitude from courts to refrain from accepting fingerprint evidence as facts that could be expressed with 100% certainty or suggesting that the evidence alone is enabling the exclusion of all others in the world except the concerned individual. Legal scholars also rightly call for more humble conclusions in the area (12-14).

The 2009 report of the US National Academy of Sciences triggered some additional comments during our reviewing period. One paper attempted to find an appropriate consensus and foster a culture of research in all identification areas, including fingerprints (15). That paper was well received and judged very balanced by Bono (16). As noted by Margot in response, the research culture has to start with an appropriate academic culture with forensic science being recognized as an academic discipline (17).

Finally, we would like to draw attention to two articles that address several debates that also concern the impressions domain (18, 19). Also in a larger forensic perspective is the presentation of a project to be carried out in the Australian justice system, aiming at assessing the effectiveness of forensic science (20).

2 Fingermarks

2.1 Friction ridge skin individualization process

Biedermann and colleagues (21) make a strong case for the use of probabilistic statements in the forensic identification disciplines, rather than stating blunt certainties. They rightly insisted on the probabilistic nature of the endeavour. The inferential and probabilistic principles involved in matters of individualisation have been explained and updated in a short entry of the Encyclopedia of Forensic Sciences (22), stressing on the decision theory that underpins that type of conclusion. An updated overview of the standards of proofs used in various countries has also been published (23). Broadly speaking the practice divides between countries applying a numerical standard (a fixed number of minutiae in agreement are required to declare an identification) and countries applying a holistic approach (the assessment is left to the examiner’s judgement based on the whole range of available features). A call to move from the numerical standard approach towards a holistic approach when the case allows has been made in Czech Republic (24).

Two short articles covering history, current practice and the usefulness of probability models (25, 26) also highlight the usefulness of such models.

Cole (27) discusses individualization, and data supporting it, in a reaction to the report of the National Academy of Sciences (28). A reaction of the European Network of Forensic Science Institutes has also been published (29), stating the different initiatives of the EFPWG (European Fingerprint Working Group) as well as Interpol that address some of the points made in the report. Kaye discusses uniqueness, and why even a large number of pairwise comparisons do not allow proving it (14). He then suggests alternative ways of expressing conclusions, rather than individualisation. A discussion of the arguments supporting uniqueness, and of the reasons why uniqueness is unproven but also irrelevant not only to forensic identification sciences but also to the legal system has been published (30). Cole exposes the difficulties associated with new ways of testifying that experts have recently explored (31), but what is clear from the recent literature is that the days where invoking “uniqueness” as the main (if not the only) supporting argument for an individualization conclusions are over.

We cannot overemphasize the need for training and continuous education in the area, especially in the light of the recent changes that occurred and to come. The paper by Mustonen and Himberg (32) describing a novel approach developed in Finland to educate fingerprint experts is inspirational.

2.1.1 Fingerprint features

Irmak (33) suggests a link between friction ridges and Merkel cells. In a study by Kücken and Champod (34), the formation of friction ridges is modelled, linking the distribution of Merkel cells to the ridge pattern on the surface of the skin. In particular, a very small perturbation of the Merkel cell arrangement (one cell) leads to differences at the level of minutiae. This study therefore provides an explanation of the variability of friction ridge patterns due to morphological events.

A study carried out on identical twins using two matching algorithms showed that both can distinguish fingerprints from identical twins. Accuracy is lowered however when including twins rather than random non-corresponding sources. Furthermore, the probability of observing the same pattern on the prints from the analogous fingers from twins does not vary between the four fingers studied (left and right index and middle fingers) (35).

A MSc thesis analysed distortion in fingerprints (36). In particular, an objective measure of the difference between elements extracted from marks from the same source is given; this is also contrasted with observations carried out on impressions from different sources. A different type of within-finger variability, variation due to growth, has also been analysed statistically (37). First, it is shown that fingerprints grow isotropically, and second, a model, linking fingerprint growth to overall body growth is developed. The difference observed between the modelled fingerprint and the fingerprint at the oldest recorded age are of the same order of magnitude as between a control and a rolled impression taken at the same age. Schneider reported also on the algorithmic modification required to enable matching fingerprints taken at different periods of growth (38).

Level 1 features

General pattern frequencies in the palm have been studied (39) on the basis of 499 individuals' palmprints. The frequencies with which loops, whorls, deltas and vestiges are observed are shown for the different areas of both palms. A comparison between palms of men and women didn't show any significant differences.

Several studies have been carried out on the subject of ridge density, in different populations and different papillary areas (fingers, palms), always showing that women have higher ridge density than men in a given population (40-46). Jowaheer and colleagues use different analytical methods to analyse these gender differences (47); a correct prediction rate of 90% or above is obtained depending on the analytical method used. Eshak and colleagues show in addition that women have a smaller finger width and surface, and a larger ridge count. Additional information concerning the distribution of ridge densities over the palm is given by Gutiérrez-Redomero and Alonso-Rodríguez (43). Other studies use ridge density measures to distinguish samples of males originating from different populations (40, 44, 48).

Sangam (49) has studied the distribution of general patterns on the fingers in the general population. Also, a comparison between male and female individuals has been carried out, and statistically significant differences in ridge counts, where males had higher counts than females, have been found. Saleem and colleagues (50) published general pattern frequencies in a population from Pakistan.

General patterns in polydactyly and syndactyly cases have been described and illustrated (51). In several cases, no general pattern was observed on the supernumerary finger in cases of incomplete radial polysyndactyly (as well as on the main finger in one case). In complete radial polydactyly, many radial loops were observed on the supernumerary fingers (4 out of 7 fingers, from 2 out of 4 subjects).

The frequencies of general patterns in 100 fingerprints of poliomyelitis patients have been reported (52). The frequencies of general patterns have been analysed for the different blood groups, and significant differences in these frequencies obtained (53). A person displaying absence of papillary ridges has been described (54), as well as an overview of possible causes of such an absence. The study of the family of the initial patient with this disorder shows that a skin-specific isoform of the SMARCAD1 gene is implicated in the regulation of dermatoglyph development (55).

An anthropomorphic discussion of pattern force and its potential role in the formation of particular characteristics in the centre of whorls is proposed by Viellieux and Thornton (56).

Level 2 features

Based on a sample of 2000 fingerprints from 200 individuals from Spain, Gutiérrez-Redomero and colleagues analyse frequency data on minutiae (57). Twenty different minutia types are analysed on different areas of the finger surface, and interesting relationships between minutia type and placement on the finger, general pattern and finger number are presented. In a further study, a similar analysis is presented for two Argentinian samples, and a difference in minutiae frequencies between the Argentinian samples and the Spanish sample are demonstrated (58); differences persist when conditioning the comparison by pattern type. Therefore the difference between the populations is a difference in minutiae type frequency, not only one of pattern type. Interestingly, no differences in minutiae type frequencies were

observed between the sexes in the Spanish sample, whereas such a difference was significant in the Argentinian samples (58). Taylor and colleagues reported on the spatial analysis of minutiae taken from more 1200 fingerprints (59). Using GIS (Geographic Information Systems)-based spatial characterisation, they computed density of minutiae, ratio between ridge endings and bifurcations and confirmed higher minutiae densities associated with pattern with higher degrees of line curvature and around focal points (core and deltas).

Level 3 features

Pore area reproducibility has been studied on microscopic images of fingerprints as well as on 500 dpi livescan images (60). While pore area was reproducible when several images were taken within one hour, this reproducibility was not observed between images taken on different days. Furthermore, on images taken at 500dpi, the pore area could not be measured, due to lack of resolution (60). Anthonioz and colleagues have also studied reproducibility of level 3 features, noting that the most reproducible features are pore position along the ridge and particular shapes of minutiae, while ridge edge shapes are subject to artefacts from the development methods (61). The stability over a long time (up to 48 years in the sample studied) of pores and ridge edge features, as well as the value they can add to a comparison, are discussed by Oklevski (62, 63). The relationship between minutiae and pores as well as their joint use is described in more detail in a following study (64). This second study also investigates the persistence of characteristics of the flexion creases of the palm. The finer details of the distal transverse crease are not stable over many years (but over up to 5, most characteristics did reproduce), while greater stability is observed in the proximal transverse crease and thenar crease (64).

Sex differences in the frequency as well as type and shape of pores in a South Indian population have been investigated (65). As would be expected from research on ridge density, women have a higher frequency of pores than men, while there was no significant sex difference in type and shape of pores. Similar results are presented in (66); pore type, position across the ridge, and size show no difference between genders, while pore frequency is higher for females. In the same study (66), an increase in overall size of pores with age was observed.

2.1.2 Probability models

Neumann and colleagues present a very advanced model to compute likelihood ratios (weight of evidence associated with a comparison between a mark and a print), integrating variations in annotations and due to distortion (67). Data concerning the validation of that model is also presented. To date, this work presents the most extensive validation exercise towards a probabilistic system that can be implemented in casework. Then, two extensions to the model have been proposed: the comparison to the complete ten fingerprints of a suspect (68) as well as the possibility of taking into account general pattern (69).

Taylor and colleagues computed false-match probabilities using Monte Carlo simulations (59) on a dataset of 1200 fingerprints. As expected the probability of a false match decreases as fingerprint attributes (e.g., minutiae number, minutiae type) were added to the model and also depends on the location on the fingerprint (core, delta or periphery). Srihari and Su (70, 71) reported on the use of graphical models to represent the spatial distributions of minutiae and their dependencies. The model is used to compute the probability of random correspondence and then compute a likelihood ratio. The model has been applied to the marks and prints associated to the Brandon Mayfield case and the NIST27 dataset. Murch and colleagues (72)

developed a hierarchical representation of relations among minutia and friction ridges, allowing searching rare features. They also developed a model to synthesize fingerprints.

A different model to compute likelihood ratios, based on morphometric and spatial analyses of minutiae configurations has been presented (73, 74). This model is based on the differentiation between impressions from a common source and between close non-matches, such as the impressions in a candidate list from an AFIS.

The operational use of probability models has also been studied (75) using the model developed by Neumann and colleagues (67). The marks considered were those not recovered initially (due to low quality), recovered but considered of insufficient quality for identification in the analysis stage, or marks that were compared to a fingerprint and where the conclusion was inconclusive, all in the normal course of casework. A few additional associations were found by examining a large amount of marks. While a generalized application of the model to all marks does not seem cost-efficient, some contexts where the use of such marks with a probability model is cost-efficient are highlighted. An analytical approach to the selection of marks to evaluate using a decision-theoretic framework is the subject of another study (76).

A very interesting paper details how to measure the validity and reliability of likelihood ratios, and why it is important to measure these elements (77).

2.1.3 ACE-V methodology, bias and performance

A PhD thesis (78) investigates the ACE-V methodology through different experiments, step by step, as well as addressing critical decision points of experts and testing tools to aid expert endeavour in fingerprint analysis. The mere possibility of validating the ACE-V methodology is questioned by Speckels (79).

Doak (80) stresses the importance of carrying out an analysis on the reference print as well as the mark, with particular insistence on the specific problematic of livescans. This analysis of the reference is recommended at least in the area in common with the mark.

Consistency in the analysis between- and within experts has been tested, paying also attention to the role played by the presence of a comparison print (81). The presence of the true source comparison print significantly decreases the number of characteristics annotated. Furthermore, both within- and between experts, differences (which were sometimes large) in the numbers of minutiae that were annotated have been observed. Hicklin and colleagues (82) study the process of analysis including local and overall assessment of clarity. This endeavour is based on a survey of examiners, where the participants were asked to assess the local quality of 70 marks (83). The standardisation of the process is proposed (82), and an interface presented. This interface allows to annotate local quality manually or automatically. Colour-coding different quality areas (84) or features (85) during analysis (and comparison) has been proposed. Such standardised annotations can be used operationally but also facilitate communication. The final decision in analysis, which is whether the mark is suitable for comparison or for identification, has been studied concerning biasing influences (86, 87). The presence of a matching or a non-matching comparison print influences the suitability decision. Also, the (biasing) knowledge of a previous determination may, in some situations, influence the decision made. In particular, more decisions of “unsuitable” were made when the cue given was “unsuitable”. The effects were weaker for examiners with IAI certification. Also, how these value judgements are carried out has been studied (the so-called “white-box” study)

using the quantity of features annotated as well as the quality map and how these elements link to the value judgement (88). Results show that minutiae count is the best predictor of final value judgement.

A discussion of confirmation biases, first from a psychological perspective, and then in the forensic domain has been proposed by Kassin and colleagues; reforms to counter these biases are also proposed (89). This paper is followed by a rich series of discussions. The whole bundle provides a very up-to-date synthesis of the current debate on bias in forensic science and in fingerprint comparison in particular (90-100).

The impact of the candidate list proposed by AFIS, and in particular the rank of the true source, on the examiner has been studied (101). While false exclusions and false decisions of 'inconclusive' are not linked to the true source rank in the list, false identifications were carried out with candidates at the top of the list; in these cases, the true source was lower in the list than the erroneously identified candidate.

The eye movements of novices and experts when comparing a mark to a print have been tracked, and the similarities within each group as well as the differences between the groups highlighted (102, 103). The initial results showed larger variation in the locations visited by the eyes for experts; however, once time was constrained to 20 seconds for experts and novices, the expert group showed higher consistency.

During the analysis and the evaluation stages, tools could inform the judgment of the specialists, for example an automated quality tool, a likelihood ratio, as well as an expert consensus (104). The expert consensus and quality information positively influenced decisions, while this wasn't the case for the likelihood ratio.

The subject of verification of all decisions is discussed by Black (105), based on the answers of different agencies to a questionnaire. Most responding agencies carried out verification for all identification decisions. However, a non-negligible percentage of agencies also carried out verifications of exclusions (55%), inconclusive decisions (52%), as well as 'no value' decisions (36%), and some carry out reviews of the original evidence items.

The accuracy, reliability, repeatability and reproducibility of fingerprint decisions have been tested in the so-called "black box" study (106, 107). Very interesting results on the accuracy and reliability of decisions are reported in (106), detailing the percentages of decisions of identification and exclusion that indeed correspond to ground truth. A very low rate of false positives was observed (0.1%). Among the marks determined as of value for ID, examiners are unanimous on 48% of mated pairs and on 33% of non-mated pairs (on average, a pair was examined by 23 examiners). This demonstrates a certain lack of consensus. This (lack of) reproducibility was then compared to the repeatability (intra-examiner). Here, 89.1% of individualization decisions and 90.1% of exclusion decisions were repeated. Most changes of opinion were towards inconclusive decisions. Interestingly, none of the four false positive errors included in this study were repeated (107). The observed lack of repeatability and reproducibility increases with the difficulty (as judged by the examiners). Another research group in Australia (108-110) demonstrated (if that was ever questioned) that fingerprint experts outperformed novice participants in comparison tasks and in the reliability of their associated conclusions. They showed the benefits of training and experience in the area.

2.1.4 Automated fingerprint identification systems

Our review is here restricted to a small part of the papers dealing with fingerprint/palm biometric systems with an emphasis on the forensic considerations. A methodology of comparing AFIS algorithms, as well as some factors impacting AFIS performances are presented by DeJongh & Rodriguez (111). Manual minutiae annotation is superior to automated annotation, but the difference is small for fingerprints (as opposed to marks). When comparing the regions above and below the core, better performance is obtained above the core. As distortion increases, performance decreases, and while a difference in orientation of 15° (with respect to the optimum) does not degrade performance, a difference in orientation of 45° does, and with a difference of 90° the true donor was no longer in a candidate list of 50. Puertas and colleagues (112) have obtained similar results; manual minutiae extraction yielded better results than automatic for marks; however, the manual annotation for only 12 minutiae (rather than all visible minutiae) yielded results that were inferior to automatic extraction. A case study explains the reason for a miss (no hit declared when the donor was in fact on the database) on an AFIS search (113).

Different articles address the need for image databases for research. A method for creating large numbers of simulated marks in order to test AFIS or to develop probabilistic models has thus been described (114). Park and colleagues propose a digital library for testing algorithms, including services to experiment and analyse, in particular, fingerprint matching algorithms (115). An open-source biometric repository, containing, amongst others, simulated crime-scene fingermarks with a known source is described by Tear and colleagues (116).

A case report on the possibility to increase the size of fingerprints by 20% for an automated search of a child's fingerprints against a database has been published (117). In the case report, a hit has been obtained in this way between the increased marks and a set of inked prints taken at adulthood. Another case report concerns the AFIS search of charred prints (118). On a charred body, prints of the funnel area of the palms and the right thumb could be recovered, casted with Mikrosil and submitted for searching in an automated system. No hit was obtained. After identification through other means, it was observed that the prints obtained from the corpse were much smaller (2/3 of the size) than the previously obtained reference material due to tissue shrinking.

Dominick and colleagues (119) investigated distortion due to heating of the substrate (uPVC) and its impact on AFIS searches. Searches were carried out before and after heating and the ratio between the first (true source) and the second score in the list was used for evaluation. Out of 50 unheated fingermarks, the true source was the first candidate in the list for 32. Out of the remaining 18, none matched its donor in first rank in the subsequent, post-heating search; no improvement was therefore obtained with the heating. After heating, when the original score ratio (between the first and second candidate on the list) was above 2.2, the post-heat mark almost always hit in the first rank as well, which was not the case otherwise. Horizontal distortion was more deleterious to AFIS results than vertical distortion. Overall the heat-distorted fingermarks were matched by the automated system correctly in 64% of cases. Improvement of matching by the use of simultaneous impressions through the integration of the match score of the individual marks of a simultaneous impression into a single result has been tested successfully (120).

Matcher improvement is also the subject of a study on overlapping impressions (121). Additional information from an examiner concerning one of the overlapping impression, in particular the region of interest, singular points as well as pointers ("orientation cues") help to

construct an orientation field for the mark in question. This in turn aids better separation, leading to improved matching performance.

Jain (122) treats the integration of an extended feature set into a search algorithm. The recommendations include to integrate level 1 and 2 extended features (ridge quality map, ridge flow map at level 1, and ridge skeleton at level 2), and to improve the quality of reference prints in order to be able to use level 3 extended features. An improvement of search results when including manually marked features in addition to the image has been reported by NIST (National Institute of Standards and Technology) (123, 124). Improvements were also reported when integrating pore features into final match scores (125); however, at least in some conditions, this improvement is very small (126). Different matchers are assessed concerning their usefulness for mark-to print matching, for example on a crime-scene, and the NFIQ algorithm is tested in order to verify whether it can be used for marks. While the matchers were fast enough for use on a crime scene, error rates were too high. Also, the NFIQ algorithm is not appropriate for the assessment of latent mark quality (127).

Several articles specifically address the question of the matching of latent fingermarks (128, 129) and of latent palm-marks (130, 131). Matching under distortion conditions (132) and matching when bad quality areas are present (133) have also been presented. Ground truth labelling, in particular of the matched minutiae, has been carried out in the NIST SD27 database, and performances of two matchers on this database are reported (134). One study investigating the performances obtained when matching latent to latent marks shows that there is room for improvement in this task (135).

2.1.5 Quality assurance and integration into the legal process

Bertram and colleagues (136) establish a link between form blindness and underperformance in fingerprint comparison, even after controlling for various other variables. This link indicates that form blindness testing could be used in the recruitment of future fingerprint trainees. Concerning the education of forensic scientists, Houck and Boyle carry out a content analysis of 9 books on fingerprints. This allows defining a certain number of subjects as well as the order in which they are treated, and use these recurring subjects as a basis for the establishment of a fingerprint curriculum (137). A standard for fingerprint individualisation (without a numerical standard) has been advocated, and one such standard has been articulated (138).

The question about how well non-specialists, e.g. the jurors in the U.S. legal systems, would understand testimony on fingerprints given in a probabilistic form has been addressed (139). A mock trial has been set up at a meeting of the International Association for Identification, and mock jurors (not fingerprint specialists, but people from the general public) assisted to testimony given on a fingerprint comparison where the result was expressed in the form of a likelihood ratio. Jurors understood the testimony rather well, and integrated the result into their reasoning. Some jurors, however, felt they did not understand the testimony, or that it was particularly useful. Also on the subject of testimony, Eldridge (140) discusses a Daubert hearing where issues mentioned in the NAS report (28) were cited by the defence. She details questions asked as well as the answers given.

Daubert challenges of forensic identification evidence types have been counted and classified; 176 such challenges (almost a third of all challenges to forensic identification evidence) concerned fingerprints, and in 12 of those cases the evidence was excluded or limited (141). The reasons behind exclusions or limitation of forensic identification evidence are exposed in

a second contribution (142). These reasons are reliability issues (57% in fingerprint testimony) which include the lack of a demonstration of reliability in the case at hand, insufficient documentation, existence of observer bias, unrealistic proficiency testing and implausible error rates. Swofford (143) exposes the legal validity of fingerprint individualisations, as well as their scientific reliability in a rebuttal to some challenges.

On the issue of communication, Found and Edmund detail the contents of a report in the pattern evidence domains (144). The different parts such a report should contain are stated, and an indication of the expected content is given. How to report inconclusive results is discussed by Maceo (145), in particular the fact that the reason for the inconclusive result should be clearly stated.

The CSI effect has been investigated with two surveys; this effect, as commonly understood, is not found in the results, although in certain cases, jurors expected and put weight on forensic evidence (146).

2.1.6 Fingerprint forgery and alteration

Several publications about fingerprint fabrication but also on fabricated marks are available for this period of review. Their focus ranges from spoofing using ‘gummy fingers’ of biometric readers to printed marks (using amino acids) left on crime scenes.

The scores obtained from an automated system using a livescan device and fake fingers as well as genuine ones from 12 subjects show that there is a lower score obtained for fake fingerprints. However, these lower scores are detected most easily in relation with the score obtained for the real finger. Finally, fakes obtained by a cast moulded directly on the finger (rather than from a latent mark) yield higher scores (147). Factors explaining successful spoofing attacks on a multispectral livescan sensor providing liveness detection have also been evaluated (148). Fakes created from direct moulds yielded higher success rates; other factors were the impostor, in particular the correspondence in general pattern between the impostor and the genuine finger; also, the number of uses of the mould was limited, and the ridge thickness of the genuine subject also played a role in the number of successful spoofing attempts. The direct impression of amino acids using an inkjet printer, and how to automatically detect false fingermarks created in this way as well as describing their visual properties, is the subject of several studies (149-152). For quality assurance purposes, the reproducibility of such printed impressions is also assessed (153).

Whether marks can actually be transferred is explored in a series of two articles (154, 155). Marks are powdered, lifted, the lift is applied to a clean surface and the mark powdered again. Transfer is possible, however, marks show halos due to the residue of the lifter surrounding the transferred mark. The transferred marks nevertheless show a large amount of features.

The detection of whether a finger presented to a livescan device is alive or not has been carried out using spectral analysis between 400 and 1650nm (156). Differences in spectra are observed between a) live and dead fingers, in particular dynamically as the live fingers show a ‘blanching’ effect when pressure is applied, and b) live and fake fingers. The fake finger surfaces used were not transparent.

The deliberate alteration of a subject’s fingerprints, so as not to be detected, is the subject of a study by Yoon and colleagues (157). Case studies are carried out and a classification of alterations into 3 classes is proposed. Then, the impacts on matchers of these alterations as

well as a way to detect them, going further than a standard quality detection algorithm, are described. A case study on the question of alteration has also been published (158), where a person had altered fingerprints, due to the application of a chemical; this chemical remains unknown, however.

2.2 Composition, aging and persistence of fingermarks

Two reviews about the topic of composition and aging of fingermarks were recently published (159, 160). Some studies specifically focused on the composition of fingermarks in terms of amino acids (161), lipid composition (162-164), age of the donor (165), as well as exposition to environmental conditions (166) or vacuum (167). A fingermark sampler (168, 169) and a modified dispensing device (170) were proposed to reproducibly leave marks or spot tests on substrates. Many studies tried to link the composition of the secretions (and their evolution with time) with the efficiency of some detection techniques (163, 165, 171-174), or with the age of the marks (175-182). All other articles using the composition of fingermarks for imaging purposes (or to detect contaminants) are described in "2.4.2. Chemical Imaging".

Used acronyms: CE-MS (capillary electrophoresis – mass spectrometry), CWL (chromatic white light), GC-MS (gas chromatography – mass spectrometry), NIN (ninhydrin), PVC (polyvinyl chloride), UV (ultraviolet), VMD (vacuum metal deposition).

Reviews - An updated overview of the composition of the secretion residue encompassed qualitative and quantitative data about fresh and aged marks, as well as the influence of numerous factors (i.e. donor, deposition conditions, substrate, environmental exposition and detection techniques) (159). In another publication, a thorough intercomparison between numerous analytical techniques illustrated how such techniques could be used to analyse and characterize secretion residue (160).

Amino acids - The identification and quantification of amino acids in latent fingermarks has been performed using CE-MS analysis (161). Twelve amino acids were identified in the analysed secretion residue, among which nine were quantified; the resulting relative abundances being consistent with previous studies in the field (e.g. serine and glycine as the most abundant amino acids). The authors also discussed the advantages and limitations of CE-MS compared with GC-MS. The relationship between palmar moisture and "quality" of the donor in terms of latent fingermarks has been explored (172). If most of the donors are considered as "average", there are always people being known as "poor" donors or "excellent" ones, when considering a detection technique (NIN in this case). Almog *et al.* showed that the palmar moisture level was not the main factor influencing the donorship for amino acid reagents (for example, some "excellent" donors had dry hands whereas some "weak" donors had moist palms). The authors hypothesized that the main factors influencing the quality of the marks were most likely the amino acid concentration in sweat, the density of pores, and the contact pressure. When studying the origin of the auto-fluorescence of fingermarks, which may be observed under UV for some marks, Lambrechts *et al.* concluded that tryptophan (if included in a protein sequence), its metabolites (e.g. kynurenine), and pheophorbide A (a decomposition product of chlorophyll) could play a role in the phenomenon (171). Further research is recommended, though.

Lipids - The "surface lipids" present in the external layer of fingermarks were analysed, especially the triacylglycerols, to study the influence of the gender or the use of cosmetics (162). No gender specificity has been emphasized, as confirmed by another study which concluded that the lipid composition does not vary significantly as a function of the age or gender of the donor (163). In the same study, Fritz *et al.* observed that the greatest loss of material appears during the first 3 months after deposition. Beyond this point, no significant variation in lipid composition was detected over a 9-month period. This could constitute an element in favour of the detection techniques targeting lipids. In a third study, it has been shown that the ratios of several fatty acids (and their corresponding methyl esters) were found to vary significantly between individuals of different race and gender (164). However, the authors recognized the limits of their study, especially too small a sample size.

Children - A study aimed at determining the fingermark composition of children (ranging from 2-year-old to 11-year-old) and showed that the carboxylic acid salts fraction was more stable than the esters one (165). The study also confirmed that the composition of children's marks differs from adult's ones in terms of relative ratio between the main components (i.e. carboxylic acid salts, esters, and proteins). The authors recommend adapting the detection techniques to target such components, so that children marks could be efficiently detected.

Aging - A study focused on the determination of specific patterns of degradations over time when marks are exposed to various environmental conditions (e.g. temperature, relative humidity, air currents, composition of secretions, exposure to daylight, type of substrate) (166). Titanium dioxide powdering was chosen to assess the quality of the "altered" marks. Some conclusions were expected, such as a better preservation on glass rather than on plastic, as well as a greater resistance of sebum-rich marks compared to sweaty ones. The authors somewhat observed that marks exposed to direct sunlight (indoors) degrade similarly to those kept in the dark, where environmental conditions are more constant. This last observation is contrary to the commonly accepted evolution pattern. The exposition of fingermarks to vacuum conditions was also studied (167). It was shown that fingermarks lose ca. 26% of their mass when exposed for one hour to vacuum conditions, which is equivalent to ca. 5 weeks of ageing under ambient conditions. If the exposition to vacuum persists, a significant loss of lipids (e.g. tetradecanoic and pentadecanoic acids) is observed, which is not the case when ageing occurs under ambient conditions. This could have an influence on the efficiency of detection techniques relying on the lipidic fraction of the secretions, especially if applied after a technique requiring vacuum conditions (e.g. VMD).

Age determination - In an attempt to assess the age of a mark based on its composition, Koenig *et al.* analysed the wax esters contained in secretion residue and identified seven for being present in most of the studied samples (175). The authors defined ratios (including wax esters, squalene and cholesterol) to try limiting the variability for a same individual and introducing a new method to determine the age of a fingermark (176). Another strategy aiming at determining the age of a fingermark is based on the use of a contactless CWL sensor (177-182). The main factors of influence were determined to be: sweat composition, temperature, humidity, wind, UV-radiation, surface type, contamination of the fingertip with water-containing substances, scan resolution and measured area size. Contact time, contact pressure and smearing of the mark were determined to be of minor importance.

Persistence - Sebaceous marks left on PVC shutters and white-painted aluminium frames were shown to resist smearing and scraping attempts, and could still be further detected by

powdering (173). The authors hypothesized that the coating process (e.g. hydrophobization agents or plasticizers) could be the reason of this enhanced resistance of the marks. The same authors also showed that such marks could survive the use of cleaning agents, since only 2 brands (among 6 tested) were able to remove the marks in presence (174).

Sampling - A fingerprint "sampler" has been proposed as a way to maximize the deposition of comparable marks, by controlling the applied force, the angle and area of contact, as well as the time of contact (168, 169). This way of doing may improve the reproducibility and consistency of marks left when developing a new detection technique or comparing its efficiency with existing methods. Another strategy consists in using a micro-dispensing device to "print" viscous material (such as artificial sebum) and prepare artificial fingerprints (or spot tests) on various substrates in a repeatable manner (170). It should be noted that the artificial sebum used in this study encompasses 10 components, among which olive oil (20%), jojoba oil (15%), coconut oil (15%), as well as oleic acid, paraffin wax, and palmitic acid (10% each) for the major constituents.

2.3 Fingerprint detection using chemical or physico-chemical processes

This chapter is structured according to the reagents, the nature of the substrates, or the scenario (e.g. arson scenes). In addition to the detailed sections, the following contributions constitute recent reviews that can serve as good starting points for readers not accustomed to the range of methods available in the field of latent fingerprint detection (183) and forensic science in general (184). Recent developments in fingerprint detection received also attention in China (185).

When reading the contributions of these last three years, we observed a great versatility in the fingerprint sampling protocols, if described by the authors. These information should encompass the nature of the secretions that were used (i.e. natural/non-enriched, eccrine-rich, or sebum-rich), the number of donors as well as of fingerprints per donor, the time between the deposition of the fingerprints and their processing, and if depletion series were considered. Some protocols were pretty close to realistic conditions (not necessarily casework-like), while other considered far-off realistic conditions. For example, most of the chemical imaging studies are based on highly-enriched marks obtained by touching pure contaminant powder with sebum-rich fingertips before leaving a mark. This observation emphasizes the fact that the sampling protocol is a key-element to be smartly designed since it eventually influences the possibility to compare experimental results between researchers and to operationally implement a new technique. In this context, the publication of Sears *et al.* (186) constitutes a useful guide to help standardizing the sampling protocols. However, for easiness of reading, we decided not to put the focus on the nature of the samples in this report, but rather on the conclusions made by the different authors. The readers must keep in mind that some conclusions may be obtained from idealized or non-realistic samples. Another topic of research deals with the objective assessment of fingerprint quality (post-detection). In this context, a relative contrast index model is proposed, using measures carried out with a microspectrophotometer on the ridges and in the valleys (187). The integration of quality levels implemented in ULW (universal latent workstation) before and after the detection of the fingerprints represents another assessment method (188).

2.3.1 Amino acid reagents

Ninhydrin has been used as a tool to evaluate fingerprint donorship (172). On a more practical aspect, acetone applied on ink to improve ninhydrin-developed mark is described by Coughlan (189). The combination of 5-methylthioninhydrin and zinc chloride is evaluated (190). Only one occurrence of DFO has been found, regarding a transfer of DFO-treated fingerprint (191). Reaction mechanisms of 1,2-indanedione with amino acid are studied in details (192, 193), as well as the effect of zinc and europium chloride on the luminescence (194). 1,2-indanedione is also used to evaluate the effect of postal distribution process on the recovery of fingerprints (195). Other amino acid reagents like naphthoquinones (196) and p-dimethylaminobenzaldehyde (197) are studied. Finally, comparison studies between various amino acids are proposed, mostly to evaluate 1,2-indanedione or 1,2-indanedione/zinc chloride performances (198-200). Its application on thermal papers is described in “2.3.7. Substrate – Thermal papers”.

Used acronyms: DFO (1,8-diaza-9-fluorenone), DMAB (p-dimethylaminobenzaldehyde), IND (1,2-indanedione), IND/Zn (1,2-indanedione/zinc chloride), 5MTN (5-methylthioninhydrin), NIN (ninhydrin).

Ninhydrin and analogues - NIN has been used to assess the relation between palmar moisture and fingerprint donorship (172). NIN application itself is not the core of this paper, but is only used as a tool. The results are detailed in section “2.2. Composition, aging and persistence of fingerprints”. NIN-developed marks, obscured by pen ink, can be rendered more visible by immersing the samples into laboratory-grade acetone to fade the ink (189). This treatment is not immediately detrimental to NIN-developed fingerprints, however a fading of NIN has been observed after several months. 5MTN is evaluated by Porpiglia *et al.* (190). 5MTN can be considered as a dual reagent, leading to coloured and luminescent results. Combined with zinc chloride, this molecule was shown to effectively detect fingerprints, but an evaluation against alternative amino acid reagents shows that the performances were significantly lower than DFO or IND/Zn.

DFO - DFO-treated fingerprints have been transferred from banknotes to white paper sheets (191). The transfer occurs during the treatment under the heat press. Reversed marks initially present on the banknotes were detected on the paper. This technique can help suppressing the background of the substrate.

1,2-Indanedione - Spindler *et al.* (193) present a fundamental study about the effects of several parameters on the reaction of IND. Among other parameters, ambient humidity is shown to have a strong effect on the reaction. The increase of luminescence by the addition of catalytic amounts of zinc chloride is also studied and confirmed. Mechanism studies of the IND-amino acid reaction are further detailed in her PhD thesis (192). Effect of zinc and europium chloride on the luminescence of an IND solution has been studied (194). Post-treatment with zinc chloride proved to enhance the luminescence whereas europium chloride did not lead to significant improvement. Finally, IND/Zn was used to investigate the effect of the postal distribution process (195). Test envelopes were used to assess the possibility to recover fingerprints. It has been shown that a great amount of marks can still be detected with sufficient quality. A relatively small number of deposits were affected by the handling of envelopes during the distribution process.

Other reagents - The use of substituted naphthoquinones is described in a preliminary study (196). Naphthoquinones successfully produced purple-brown fingermarks with red luminescence. The intensity of colour and luminescence depends on the naphthoquinone type. Further studies are required to determine the actual efficiency in comparison to current benchmark reagents. Another preliminary work shows that DMAB successfully reacts with amino acids and can be used to detect fingermarks on porous substrates (197). The obtained results are both coloured and luminescent.

Comparison studies - Envelopes aged from 1 to 21 years were used to compare the efficiency of NIN, DFO and IND (198). This study shows that the age of the fingermarks does not influence the ability of the methods. The authors conclude that IND gives superior results in comparison to NIN and DFO, even on old marks. The performances of DFO and IND/Zn were also compared in an extensive study (199). It was found that IND/Zn developed more fingermarks, with a brighter luminescence. These results led to a nationwide field trial, which is still underway. A comparison is also performed by Berdejo *et al.* (200) between four amino acid reagents (DFO, 5MTN, lawsone and IND/Zn). IND proved to be superior to all other reagents, but contrary to previous studies, it is stated that zinc chloride does not improve the fluorescence. The processing of thermal paper with IND/Zn has been described in combination with heat or destaining solutions (201), or by the use of a dry method (202). These studies are detailed in section "2.3.7. Substrate - Thermal papers".

2.3.2 Cyanoacrylate fuming

A lot of publications dealt with the well-known cyanoacrylate fuming process. Some authors focused on the role played by humidity (203) and temperature (204-207) on the polymerization mechanism. Others proposed different ways of rejuvenating old marks to improve the detection quality (208-210). New dyes were proposed (211, 212) as well as a proposition to subsequently apply VMD after cyanoacrylate fuming (213). A recent trend consists in a one-step procedure allowing to obtain marks readily fluorescent, without the need for stain post-processing (206, 214, 215). Miscellaneously, two quality control tests were proposed (216, 217), the possibility for DNA transfer was proved (218), the effect of fuming on the composition of some plastic bags studied (219), and the use of a commercial fuming device evaluated (220). All the articles dealing with the use of chemically-modified cyanoacrylate monomers for imaging purposes (221, 222) are described in section "2.4.2. Chemical Imaging".

Used acronyms: CA (cyanoacrylate or cyanoacrylate fuming), BY40 (basic yellow 40), DMAB (p-dimethylaminobenzaldehyde), GC-MS (gas chromatography – mass spectrometry), HCN (hydrogen cyanide), LDPE (low-density polyethylene), NIR (near infrared), R6G (rhodamine 6G), RAM (rhodamine – ardrox – methylene blue), RH (relative humidity), SPME (solid phase microextraction), TWA (time-weighted average), UV (ultraviolet), VMD (vacuum metal deposition).

Fumigation / Polymerization mechanism - Lowering the temperature during the CA process may influence the polymerization mechanism, in terms of initiation sites, quality of the polymer chains, as well as their morphology. A decrease in temperature could consequently promote a larger coverage of dense polymer chains over the mark (204), especially with aged

fingermarks. A similar conclusion has been reached by other researchers, who recommend forcing the condensation on items before the fuming processing (205, 206). Ideally, the temperature of the items should be decreased by ca. -4.5°C to ca. -11°C (i.e. -5°F to -20°F) relative to the ambient temperature, before processing them in the fuming cabinet. The authors also observed an increase of adherence of the CA dye (i.e. R6G) which is subsequently applied. In another study, it was confirmed that 80% RH constitutes the optimum level of relative humidity for the development of the most high quality marks (203). Overheating the CA monomers could generate HCN, even if none of the tested superglues generated detectable amounts of HCN when heated for 30 min at 180°C (207). Nevertheless, quantifiable amounts of HCN were generated from the thermal decomposition of CA monomers and polymers when heating at 200°C and above. Even if the released quantities are below the TWA concentration limit for workplace exposure, it is recommended to limit the heating temperatures of home-made fuming systems to values below 240°C . The relative humidity during long-term storage of items (e.g. months) before their processing seems to have no significant influence on the quality of development (223). Consequently, it is not recommended to install equipment maintaining constant environmental conditions (note: storage temperature was varying between 22 and 25°C).

Pretreatments - It is commonly accepted that the effectiveness of CA is reduced when dealing with aged or dry marks. Different protocols were consequently proposed to "rejuvenate" old marks before their processing: exposition to acetic acid or ammonia vapours for 15 minutes (208) or to vapours of a 10% w/v methylamine solution (209). Pinto *et al.* observed an increase of the ridge thickness after CA following acetic acid vapours pretreatment, compared to conventionally-processed marks. Another experiment consisted in dusting the marks with valine-containing powders prior to CA (210). The aim was to enrich the sebaceous fraction of the marks (more likely to survive upon aging) with polymerization initiators. The obtained results were mitigated in comparison with the chemical vapour enhancement.

Post-treatments / Sequence - Styryl 11 was shown to give better results than R6G when observed in the near infrared region (NIR; $>700\text{ nm}$) (211). Styryl 11 has a maximum absorbance at 575nm , and a strong emission at 766 nm . One advantage of visualizing marks in the NIR region is that it is unlikely to face an unwanted background luminescence (in comparison with stains emitting in the visible range), for example with aluminium soft drink cans. It is also possible to combine Styryl 11 with R6G ("STaR11") to extend the Stoke's shift. In another study, DMAB was proposed as a vapour-phase stain for CA-treated fingermarks (212). The described procedure consists in leaving the items in a close container with DMAB powder for at least 48 hours, and subsequently observing the detected marks under UV light. The authors report the obtaining of fluorescent marks on substrates not suitable for a conventional liquid-based staining process (e.g. unglazed earthenware flower pot). Finally, the sequential application of CA and VMD on plastic has been studied at a molecular level (213). The sequence "CA/BY40 – gold/zinc VMD" was proved to be adapted to the processing of LDPE substrates.

One-step fluorescent cyanoacrylate - A new generation of CA process has been tested. It consists in fuming and staining the items - simultaneously - by co-evaporating the CA monomer with a fluorescent powder (214). The biggest advantage of a one-step process is to avoid the use of organic solvents (staining post-treatment), which may be time-consuming and detrimental for some substrates. In this study, the prototype of the Polycyano (Foster and Freeman – UK) was tested. The overall quality of the development was comparable to what is

obtained using the current two-step fuming and dye stain procedure (e.g. R6G, Ardrex, RAM). One drawback, that could be noted, is that the process requires the modification of the fuming cabinet to increase the temperature of the heating plate to 230°C. This procedure may be costly and lead to the generation of HCN as stated by Fung *et al.* (207). In another research, people were interested in expanding the excitation range of a dye-stained CA by using a sublimating dye (i.e. Sublaprint Red R70011) (206). The new combination of dyes extended the excitation range from 365-505 nm to 365-530 nm (which is more compatible with the 530 nm single wavelength light source found in some agency laboratories). Finally, the synthesis of CA monomers functionalized with fluorescent groups has also been attempted (215). However, these monomers were unable to detect fingermarks when fumigated (like a conventional CA monomer). The authors proposed to solubilize the fluorescent monomers in xylene and applied them by a quick immersion of the item to be processed.

Quality controls - Two control tests were designed to assess the quality of development upon CA: (a) one based on sodium hydroxide spots, and recommended for blood cases when it is expected to use luminol subsequently on the scene (216), and (b) one based on fingermarks made of artificial sweat (217). Thiburce *et al.* showed that an optimized exposition to CA may have a positive impact on the subsequent application of luminol (or Bluestar[®]), with longer-lasting chemiluminescence (216). On the contrary, an excessive quantity of CA polymer may completely hinder the reaction of luminol with the underlying blood (shoe)marks. Velthuis *et al.* considered a mixture of different compounds (i.e. fatty acids, amino acids, glycerides) to mimic natural sweat, which was subsequently jellified using gelatin leaves (217). The control test consisted in marks left on plastic/glass using a silicone fingertip in contact with the jellified artificial sweat.

Miscellaneous - A study showed the possibility for DNA to accumulate both outside and inside a CA chamber, as well as for DNA to transfer from one exhibit to another during the fuming process (218). Recommendations are given by the authors to limit such unwanted contaminations (e.g. sampling for DNA before the fuming, limiting the number of evidence in the chamber, decontaminating using UV light-equipped chamber). The effect of CA on the composition of polyethylene bags (used to carry illicit drugs, for example) was studied using SPME/GC-MS analyses (219). A portable CA system (SUPERfume[®] from Foster and Freeman – UK) was tested, and its efficiency compared with the use of aluminium powder on crime scene (220). As a result, SUPERfume was more effective on textured and smooth plastic surfaces (and for marks stored at 37°C), whereas aluminium powder was shown to be more effective on glass, enamelled metal paint, and varnished wood (and for marks stored below 20°C). When facing a scene to be processed, the authors consequently recommend to consider each surface independently (if possible) or to dust the surfaces made of glass, enamelled metal paint, and varnished wood before using the SUPERfume equipment.

2.3.3 Lipid stains and lipid-oriented techniques

Evolutions of the Oil red O technique consisted in optimizing the formulation (224, 225), proposing a luminescent alternative (226), and evaluating its performance in sequence with DFO and ninhydrin (227). About the physical developer, quality control procedures were proposed (228, 229), an extended shelf-life has been measured for the new formulation of physical developer (230), and its robustness has been tested (231). The interest of introducing physical developer in a sequence after amino acid reagents has also been confirmed (232).

Finally, *TECTOPO* has been applied on paper (233) and Nile red proposed as a new lipid stain (234).

Used acronyms: *DFO* (1,8-diaza-9-fluorenone), *EDTA* (ethylenediamine-tetraacetic acid), *IND* (1,2-indanedione), *NIN* (ninhydrin), *ORO* (oil red O), *PD* (physical developer), *R6G* (rhodamine 6G).

Oil Red O - An alternative to the original formulation of ORO (see: (226)) is proposed, in which the methanol-based solution is replaced by propylene-glycol (224). This new formulation was proved to be as efficient as the original one, but is safer, quicker, and requires fewer reagents. It however suffers from the same limitations as the original formulation, especially on some kinds of substrates and on aged fingermarks. A sequence of treatment for porous substrates is further proposed: "IND (/HFE-7100) – ORO (/propylene-glycol) – PD" (225). The authors recommended reducing the immersion time in the IND solution to less than 5 seconds, to avoid a detrimental effect on the lipid fraction of the secretion (which could impact the subsequent ORO treatment). Another research aimed at studying the sensitivity of DFO, NIN, and ORO over time, as well as the contrast of the resulting marks, for different kinds of porous substrates (227). It has been shown that ORO could be applied subsequently to the "DFO (/HFE-7100) – NIN (/HFE-7100)" sequence, which could result in an enhancement of the already-detected marks and detection of latent-remaining ones (especially on kraft paper, cardboard and thermal paper). On white and recycled papers, no additional marks were detected by ORO when applied after DFO and NIN. Finally, a luminescent alternative to the original ORO formulation has been proposed, which could be used on wetted and dark porous substrates (226). Substrates of interest are first processed with the original ORO, and immediately followed by the spraying of a R6G staining solution. The marks obtained using this procedure appear as dark ridges on a luminescent background.

Physical developer - In order to test the reliability of a prepared PD working solution, two procedures were proposed: (a) printed standardized test strips using a modified inkjet printer (228), and (b) EDTA spot tests on Whatman #2 filter paper (229). The pattern developed by Kupferschmid *et al.* contains geometric shapes made of ascorbic acid and oleic acid aqueous solutions (used as inks by the printer). The correlation between the number of detected shapes on the test strip and the quality of fingermark development was shown to be high. The EDTA spot test procedure represents an inexpensive, reliable, stable, and rapid alternative to a printed pattern as well as to gold chloride spot tests on filter paper. The shelf-life of the last PD formulation (in which Synperonic N has been replaced by Tween 20) has been thoroughly studied (230). As a result, it has been shown that the shelf-life of the PD working solution was of 10-15 days for the Synperonic N formulation, but rises up to 2 1/2 months when using Tween 20. The robustness of the PD formulation was tested by comparing the efficiency of different working solutions, among which voluntary alterations and modifications were introduced (231). It has been shown that (a) the order in which constituents of the redox solution are added has no influence, (b) Tween 20 is an advantageous alternative to Synperonic N, and (c) maleic acid is to be preferred to malic acid during the prewash. The sequence "DFO – NIN – PD" was tested, and the results confirmed what had already been pointed out about the usefulness of using PD in terms of detection of additional marks and quality improvement of the already detected ones (232).

TECTOPO – This lipid reagent has been used to detect marks on papers using time-resolved luminescence (233).

Nile Red - Nile Red is a newly proposed fluorescent lipid stain to be used on wetted porous substrates. When compared with PD, it appeared that PD remains the most reliable and sensitive technique. Nevertheless, Nile red can be added at the end of the treatment sequence for porous substrates, after PD (234).

2.3.4 Dry micro-/nano-sized powders and powder suspensions

Two different powder applications are described (235, 236), as well as two lifting processes (237, 238). Several new micro-sized powders like magnetic powder based on indigenous minerals (239), iron flakes (240), turmeric (241), silica gel G (242) and synthetic food and festival colours (243) are evaluated. Phosphorescent powder is also discussed (244, 245). Powder application proved to be the best method on particular substrates, such as fruits and vegetables (246). Powder was used to evaluate the potential of fingerprint recovery on cell phones (247).

For the nano-sized powders, various types are discussed. Among them are aluminium oxide (248), calcium carbonate (249) and iron oxide (250). Quantum dots coated with polymers (251) or silica (252) can also be applied, as well as embedded in porous matrix (253-255). Anti-stokes nanopowders are presented (256, 257). Two papers depict the uses of silica nanoparticles (258, 259). All the other articles dealing with the use of functionalized silica nanoparticles to study the composition of secretion residue and allow the detection of exogenous elements in fingerprints using analytical methods (260-262) are described in section "2.4.2. Chemical Imaging". Finally, zinc carbonate is used as wet suspension powder (263-267).

Used acronyms: CA (cyanoacrylate fuming), CdS (cadmium sulphide), CdSe (cadmium selenide), CdTe (cadmium telluride), NIR (near infrared), NPs (nanoparticles), QDs (quantum dots), R6G (rhodamine 6G), SPR (small particle reagent).

Application method and lifting - The cotton wool powdering technique has been evaluated (235). It proved to be efficient and easy-to-use since larger surfaces can be covered. The obtained marks were judged to be of a comparable quality as those obtained with squirrel-hair brush. Powder can also be applied using an aerosol spray (236). After modifications in formulation and in aerosol technology, this technique proved to be a viable method for crime scene use. The amount of powder is controlled and the contact with the substrate is reduced. The gelatin lifting process has also been extensively evaluated (237). Gelatin lifting prior to powdering is less effective than powdering only but it might be considered in case of contaminated surfaces or incompatibilities with powders. It proved to work well on smooth surfaces, but its effectiveness decreases with age of the mark. In the case of fingerprints made of dust or deposited on dusty surface, dust print lifters are effective tools to recover fingerprints when powder dusting cannot (238).

Dry micron-sized powder - A magnetic powder based on an indigenous mineral from Thailand is tested (239). The minerals are grinded and mixed with nickel. The powder is applied with a magnetic brush and marks of good quality are obtained. Iron flakes of different

dimensions, produced with a high-energy milling device, are evaluated for fingermarks powdering (240). 50 µm flakes can sometimes develop marks of better quality than conventional black magnetic powders. Turmeric powder, normally used as an ingredient in Indian food, is used to detect fingermarks (241). This powder, cheap and non-toxic, is efficient on various non-porous substrates. Fingermark detection with silica gel G (242), synthetic food and festival colours (243) are presented. These powders are efficient on non-porous substrates. Phosphorescent (glow-in-the-dark) powder is compared to fluorescent powders (244). The authors stated that a better contrast is generally obtained with glow-in-the-dark powder. Another powder with upconversion properties is investigated by Drabarek *et al.* (245, 268). Anti-stokes phosphor pigments mixed with white powder can detect fingermarks of good quality without background staining, since an infrared illumination is used and visualization made in the visible range.

Fingermark detection on fruits and vegetables is discussed by Fergusson *et al.* (246). The most promising methods are black magnetic powder and black powder suspensions. Recovery of fingermarks on cell phones is evaluated by Lodhi *et al.* (247). Identifiable marks were obtained on 11% of the items (n = 121, 13 marks) using silk black powder.

Dry nano-sized powder – This section is also focused on powdering, but using dry NPs, nanostructured materials or materials containing NPs. Aluminium oxide NPs are covered with two dyes (R6G and styryl 11) and mixed with silver magnetic powder (248). The results can be visualised in both visible and NIR regions. On the same principle, Khokhar *et al.* (249) used nanopowders made of calcium carbonate or copper both coated with an organic compound. Iron oxide NPs coated with a silver layer have been applied under dry form on fingermarks (250).

Different QDs can also be powdered on fingermarks. CdS QDs coated with various polymers are applied on glass and aluminium foils (251). CdTe QDs were also coated with silica layers (252). When applied on fingermarks deposited on non-porous surface, the powder adheres on fingermarks leading to luminescent results. QDs can be embedded in different porous matrix used as a template for the NPs synthesis. CdS QDs (253) and CdSe QDs (254) were embedded in phosphate heterostructures. Gao *et al.* (255) used CdTe in montmorillonite to produce a nanostructured powder. After application on fingermarks, they obtained luminescent results. Anti-stokes nanopowder made of NaYF₄:Er,Yb (256) and YVO₄:Er,Yb (256) have been powdered. There are only two works reporting the use of silica NPs powder for fingermark detection. Liu *et al.* (258) synthesized silica NPs containing R6G. The obtained powder is mixed with magnetic iron powder and applied with a magnetic brush. The second paper is based on lanthanide-doped yttrium zirconate entrapped in silica NPs (259). The obtained powder is applied on fresh fingermarks. These two publications are mainly focused on the optical properties rather on the efficiency to detect fingermarks. No comparison is made with traditional powders.

Other functionalized silica NPs were used as a dusting powder to allow the detection of exogenous pharmaceutical drugs (metabolites) and explosive (contamination) in fingermarks using analytical methods (260, 261), as well as to study the composition of sweat secretion (262) – see section "2.4.2. Chemical Imaging".

Wet powder suspension - New SPR based on the use of zinc carbonate have been studied. Eosin Y (263) or crystal violet (264) are added in the formulation. These methods proved to be efficient on surfaces that have been immersed into water. Zinc carbonate SPR is effective on compact disc and does not interfere with data retrieval (265). A comparison study between

CA, powdering and SPR to recover marks on wet transparent foils has been performed (266). SPR proved to be the most efficient technique, even for marks exposed to water during at least one week. In similar study about the recovery of mark on glass and metal surfaces that have been wet, CA proved to give the best results compared to SPR (267).

2.3.5 Nanoparticles in solution

Uses of nano-sized powders and nanostructured powders have been discussed in the previous section (2.3.4). Only nanoparticles in solution are described here, and are classified by their composition, starting with gold nanoparticles (269-275) and followed by semi-conductor nanoparticles (quantum dots) (276-283). The other nanoparticles types, such as metal oxide (248, 249), silica nanoparticles (258, 259) and up-convertors (256, 257), are only applied as powder and were therefore described in the previous section. An extensive review on the use of nanoparticles applied for fingerprint detection is proposed by Dilag et al. (284). The authors describe uses of gold nanoparticles, fluorescent dye-doped nano-powders and quantum dots. Another review on the same topic is made by Hazarika and Russell (285). An entire book chapter is also dedicated to the fingerprint detection with nanoparticles (286). This contribution deals with the synthesis of different nanoparticles types (gold, quantum dots and silica nanoparticles) and their application to fingerprint detection. It also makes assumptions about the interaction principles between nanoparticles and fingerprint secretion. All the articles dealing with the use of nanoparticles functionalized with antibodies (287-291) are described in section "2.3.6. Immunogenic detection".

Used acronyms: CA (cyanoacrylate fuming), CdS (cadmium sulphide), CdSe (cadmium selenide), CdTe (cadmium telluride), MMD (multi-metal deposition), NPs (nanoparticles), PAMAM (poly[amido amine]), PD (physical developer), QDs (quantum dots), SiO₂ NPs (silica or silicon oxide nanoparticles), SMD (single-metal deposition), VMD (vacuum metal deposition), ZnS (zinc sulphide).

Gold nanoparticles - General uses of gold in forensic sciences are described by Mohamed (269). The review encompasses the topic of illicit drug and describes the use of silver, gold and other metals to detect fingerprints (PD, MMD, SMD, VMD). Works on gold NPs are various. Fairley *et al.* (270-90) propose an evaluation of the effectiveness of various multi-metal deposition techniques (MMD I & II, and SMD). MMD II is considered as the most effective, but since it is more labour intensive, MMD I appears as the best compromise between practicality and effectiveness. In general, it performs better than VMD or CA on cling film and plasticised vinyl, but it is not effective on leather and masking tape. The final results obtained with SMD are not considered as sufficient, due to lack of contrast and consistency. Bécue *et al.* (271) described a new version of SMD. The synthesis of the gold NPs has been simplified, and an amino acid is also grafted onto the surface. The modifications led to a simplification of the synthetic procedure, and the new formulation shows a stronger resistance to pH variations. Results are more reliable than with the previous SMD formulation. Contrary to the two previous works for which NPs were already present in the solution, Hussain *et al.* (272) applied a solution containing gold chloride. In this case, the fingerprint residues act as an initiator for the particles growth. There is an *in situ* formation of NPs onto the fingerprints.

When detecting fingermarks, a contrast is generally obtained by specifically targeting the sweat secretions, but in a recent work, gold NPs have been designed to target the cellulose of the paper instead of the marks (273, 274). The NPs functionalized with a thiolic ligand possess an affinity for the substrate and negative fingermarks have been obtained after a post-treatment using PD. Specific targeting was also possible when NPs (gold and silver) were attracted onto the metallic surfaces by electrodeposition (275). Negative marks were obtained, the secretions acting as an insulator.

Quantum dots - Among QDs applied in aqueous solution, CdTe QDs are the most common. These NPs functionalized with a carboxylic group have been applied on fresh fingermarks deposited on various surfaces (276), as well as on cellulose tape (277), but with mitigate results. The same NPs functionalized with a double carboxylic group were applied on non-porous surfaces (278, 279). The results are obtained after a very short immersion time of 1 to 10 seconds, or by spraying the surface with the solution. Gao *et al.* (280) used another functional group, leading to the formation of positively-charged NPs. According to the authors, these particles give better results than the negatively-charged ones. Contrary to toxic cadmium-based NPs, Moret *et al.* (281) describe the use of ZnS QDs to detect fingermarks in blood. Results obtained on non-porous have been compared to Acid Yellow 7. CdS (282) and CdSe (283) prepared in PAMAM dendrimers and stabilized in water were applied on adhesive tapes and tin foils. Sebaceous fingermarks aged up to one month were effectively detected.

Other nanoparticles - Antibody-functionalized silver NPs were used to specifically target sweat components, followed by chemical imaging using Raman spectroscopy (292). This article is described in details in section "2.3.6. Immunogenic detection". This section also describes the use of other types of antibody-functionalized NPs, which can target specific compounds that can be present in the secretion like drug metabolites (288), explosives (285), cotinine (290, 291) and amino acids (287).

2.3.6 Immunogenic detection (antibody/antigen)

Immunogenic-based techniques have been recently used to specifically target antigens contained in the fingerprint residue. Reviews of the field have been proposed, from the early attempts to the current research strategies (285, 293). All the work done on immunogenic techniques consisted in combining the antibodies with (magnetic) nanoparticles, so that they could target L-amino acids (287), drugs and metabolites (288, 290, 291), human immunoglobulin G (292), and body fluids (289). A new approach is also currently investigated: the use of aptamers (i.e. single-stranded nucleic acid oligonucleotides) to specifically target secretion components (293).

Enantioselective anti-L-amino acid antibodies conjugated to gold nanoparticles were used to detect marks on non-porous substrates (287). Early results were presented, and the field is still being investigated since conventional methods are shown to produce superior fingerprint details on fresh samples. It should be noted that the author emphasized the role played by nutrition habits on the efficiency of the technique (phenylalanine-based sweeteners, for example).

Magnetic nanoparticles were functionalized with a series of antibodies to specifically target the following antigens: morphine (288), benzoylecgonine (288), cotinine (290, 291). All the authors emphasized the high selectivity of the techniques, as well as the possibility to observe the detection in luminescence. However, nothing is said about the introduction of such an approach in the existing detection sequences (forensic context). Despite it is not in direct link with the detection of fingermarks, it is interesting to cite the immunodetection and localization of body fluids (i.e. blood and saliva) on various substrates using magnetic nanoparticles functionalized with the corresponding antibodies (289). Finally, silver nanoparticles functionalized with antibodies able to recognize human immunoglobulins (IgG) were used to specifically target sweat components of fingermarks (292). By doing so, it was then possible to visualize the fingermarks through a Surface Enhancement Raman Spectroscopy imaging process, as described in section "2.4.2. Chemical Imaging".

2.3.7 Substrate - Thermal papers

Among the different techniques developed to detect fingermarks on thermal paper, we can cite: steam (294), controlled application of heat (295), amino acid reagents (202, 296), background darkening treatment (201, 296), as well as iodine to retrieve erased text (297).

Used acronyms: BY40 (basic yellow 40), DABCO (1,4-diazabicyclo[2.2.2]octane), DFO (1,8-diaza-9-fluorenone), IND/Zn (1,2-indanedione/zinc chloride), LED (light-emitting diode), NIN (ninhydrin), PVP (polyvinylpyrrolidone)

Steam / Heat - The use of a fabric steamer to detect fingermarks on thermal paper has been studied (294). The authors concluded that sebaceous marks are more likely to be detected, especially if they are fresh (the rate of successful detection decreasing over time). The results were somewhat inferior to the ones obtained with some conventional techniques, such as acetic acid fumes and NIN (in HFE-7100). In an effort to uniformly apply heat on thermal paper, an apparatus has been designed and tested (295). The sample is placed between two rectangular plates: a brass one (further heated to 44°C) and a glass one (allowing the observation of the detection process). The use of a 465 nm blue LED illumination could help in the observation of the ridges development.

Iodine - The use of iodine vapour on thermal paper could help in retrieving the text that was initially present, but faded over time or erased as a result of the application of a fingerprint enhancement technique (297). The recovered texts may appear as positive (dark writings on white background) or negative (white writings on dark background), and remain visible for several weeks. This technique is also efficient for texts printed using dot-matrix printers, and seems unaffected by the age of the document.

Amino acid reagents - A "dry" application of IND/Zn, with low temperature heat, is reported as a non-destructive way of detecting fingermarks on thermal papers (202). Luminescent fingermarks could be obtained without darkening the background (as it would be the case with the traditional procedure) by placing the sample in sandwich between two reagent-impregnated filter papers and heated at 60°C for 15 minutes in a Ziploc™ bag. Schwarz *et al.* propose different chemical possibilities to deal with thermal papers when applying amino acid reagents (IND, NIN, or DFO): (a) preventing the darkening of the background by including

PVP (Kollidon[®] 12 PF) into a conventional NIN formulation (aka NinK12) (296), or (b) chemically reversing the background darkening by using a "whitening" solution (G3 or DABCO) (201, 296). It has to be noted that the G3 and DABCO solutions erase any printed text present on the document.

2.3.8 Substrate - Metal and cartridge cases

The visualization of fingermarks on metal surfaces through an electrochemical process has been further studied (298-304), as well as electrostatic deposition (305), electrolysis (306), vapours of S₂N₂ (307), and electrochromic process (308, 309). Metal sputtering was also assessed (310), as well as thermal development (311). On cartridge cases, different techniques and sequences of detection were tested (312-314). The possibility to recover fingermarks from cartridge cases submitted to arson conditions (315) is described in section "2.3.11. Scenario - Arson scenes". The use of chemical imaging to detect fingermarks on metal substrates (i.e. platinum, gold, silver, copper and stainless steel) (316) is described in section "2.4.2. Chemical Imaging". Some case reports were also described, dealing with successful fingermark detection on a cartridge case (317) and with the overall recovery rates observed in forensic laboratories (318, 319).

Used acronyms: ATF FSL (Alcohol, Tobacco, Firearms and Explosives Forensic Science Laboratory), CA (cyanoacrylate fuming), DPD (Denver Police Department), R6G (rhodamine 6G), RUVIS (reflected ultraviolet imaging systems)

Metal corrosion effect (brass) - The process leading to the visualization of fingermarks on brass cartridge cases through an electrochemical process (corrosion) has been further studied (298-302). RUVIS was shown to be inappropriate to observe corrosion on fired brass cartridges for which white light is to be preferred (304), combined with the use of a selective colour mapping process (e.g. using Adobe Photoshop[®]) (303). The electrostatic deposition system causes no detrimental effect on the ballistic identification process, once the deposited powder has been washed thoroughly after fingermark detection (305). Other researchers studied the enhancement of fingermarks using electrolysis on fired brass cartridges (306). For this experiment, brass cartridges were immersed in diluted hydrochloric acid (HCl) before applying a voltage to initiate the galvanic corrosion reaction. As a result, a visible contrast appears between the ridges and the metal surface. It has to be noted that the fired group of cartridges showed better results compared to the unfired group. Vapours of S₂N₂ were used to detect marks on metal items, for which the marks have been removed (deliberately or as a consequence of an explosion) (307). S₂N₂ polymerizes around localized physical imperfections on the substrate surface, and by the same way at the level of the corrosive modifications due to the presence of sweat (even if the mark has been erased).

Metal surfaces - The electrochromic enhancement of fingermarks on metal substrates was compared with conventional methods (i.e. dry powder, wet powder and CA) (308, 309). Briefly, the electrochromic enhancement relies on the deposition of a conducting polymer on the metal substrates. The secretions inhibit the polymerization, resulting in a negative image of the mark. In this study, stainless steel samples were exposed to different scenarios (e.g. immersion in water, high temperature, washing using soap) then further processed for fingermark detection. Dry dusting and electrochromic enhancement gave the overall best

results. Metal sputtering (= vacuum evaporation) of copper or gold onto stainless steel substrates allowed the detection of fingermarks (310). It was observed that sputtered gold and copper tend to concentrate in the ridges, resulting in dark ridges on metal background. Gold sputtering was favoured, especially for aged marks and for its stability against oxidation. Thermal development of fingermarks on metal surfaces was also further studied (311). Brass appeared to be the least dependent on temperature up to 600°C, aluminium gave poor visualization when heated above 280°C, and stainless steel only gave good visualization when heated above 600°C (a loss of detail starting to appear when heated a 900°C).

Cartridges cases - A comparison of different techniques to detect fingermarks on fired cartridge cases showed that the Gun blue solution, the palladium deposition, and the CA/BY40 were all successful in detecting marks, with a preference for the first two techniques in terms of mark quality (312). The authors observed that most of the ridge details were detected below the bottom third of the cases. It has to be noted that no fingermark detection was observed with the electrostatic deposition (device built by following the descriptions given in the literature). These results were confirmed by another study for which six enhancement techniques to detect fingermarks on unfired brass cartridge cases were compared (313). Two sequences provided the best results and showed no statistical difference in terms of efficiency: "CA – Gun Blue – BY40" and "CA – Palladium deposition". Powder suspension produced the poorest results. Acidified hydrogen peroxide (H₂O₂) is a technique allowing the detection of fingermarks on brass, and in this case on fired brass cartridges (314). The authors recommend to use it after CA/R6G; they also set a maximum processing time of 75 seconds to visualize ridges (average detection time of 24 seconds), emphasizing that this process could negatively interfere with firearms examinations since noticeable effects on the cartridge characteristics may be observed after 20 seconds of treatment.

Case reports - Babin has reported the successful application of CA on a fired 9mm cartridge casing, with the detection of a partial mark consisting of a smudged core with identifiable ridges above it (317). Among the firearms processed by the ATF FSL in San Francisco over a three-year-period (Jan'07 to Dec'09), a recovery rate of 13% was obtained on firearms (n = 598, 168 identifiable marks), 7.6% on ammunition magazines (n = 423, 46 marks), and 0.12% on cartridges (n = 6,698, 8 marks) (318). Whenever possible, the authors recommend removing the grips of the firearms prior to processing, because the area underneath the grips can yield identifiable marks. A similar study conducted at the DPD over a 2-year period (May'08 to May'10) showed a recovery rate of 3.7% on firearms (n = 189, 7 identifiable marks), 10.0% on ammunition magazines (n = 110, 11 marks), 0.25% on live cartridges (n = 817, 2 marks), and 0% on spent cartridge cases (n = 200, no mark) (319). The firearm evidence items were processed with orange magnetic powder (leading to 6 marks), CA and RUVIS (leading to the successful detection of 14 marks).

2.3.9 Substrate – Tapes and adhesives

In the field of tapes, people were interested in the best way to separate duct tapes stuck together (320, 321) or on paper (322), in determining an optimized detection sequence for processing the adhesive side of tapes (323), especially if blood marks have to be found (324). The use of engineered nanoparticles to detect fingermarks on the adhesive side of tapes (277, 282) is described in section "2.3.5. Nanoparticles in solution".

Used acronyms: CA (cyanoacrylate fuming), RAY (rhodamine – Ardrex – basic yellow 40), SSP (sticky-side powder)

Manual separation was determined to be the only method to separate duct tapes stuck together while still enabling the recovery of latent fingermarks on the adhesive side (320). In another study, the use of liquid nitrogen applied with a cryogun was preferred over gradual force and adhesive neutralizer (Un-Du[®]) (321). In both studies, the use of an adhesive neutralizer was strongly discouraged for duct tape, since its use degraded the adhesive support and consequently the marks in presence. In another study (322), the Turkish solution (a mixture of solvents) outperformed Un-Du[®] to help removing adhesives from papers.

Using RAY as a post-CA dye constitutes an efficient way to process the adhesive side of tapes (323). Moreover, the following sequence has been shown to give optimal results (for the adhesive side): "CA – gentian violet – black/white powder suspension – RAY dye", with results superior to the use of the dye alone post-CA. Amido black proved to be the best method for developing blood fingermarks on the adhesive side of duct tape, when compared with Wetwop, SSP, Liqui-Nox, and gentian violet (324). Moreover, amido black does not seem to hinder the subsequent application of techniques dedicated to non-blood marks. For non-blood mark detection, Wetwop and SSP offered the best results (Wetwop offering the advantage of giving positive results with both blood and non-blood marks).

2.3.10 Substrate – Skin

An extensive review on the recovery of fingermarks on human skin is proposed by Wilkinson (325). Färber et al. (326) report the results of a large European project, and Beaudoin (327) presents a comparison between amido black and ortho-tolidine.

An extensive review on the recovery of fingermarks on human skin is proposed by Wilkinson (325). In a more practical approach, Färber *et al.* (326) report the results of a European project called "Latent Fingerprints and DNA on Human Skin". The purpose was to conduct a systematic research on the recovery of fingermarks and DNA on skin. The marks were treated with magnetic or black powder, and were lifted with a gelatine foil or silicone casting material. The lifts were systematically swabbed and analysed to detect DNA. The authors recommend the use of magnetic powder, lifted with silicone casting material (Isomark[®]).

The recovery of fingermarks in blood on skin was also studied (327). Amido black and ortho-tolidine were compared and despite the toxicity of the latter, it remains the most effective technique. Furthermore, amido black cannot be adequately cleaned and may interfere with the autopsy findings.

2.3.11 Scenario - Arson scenes

The possibility to retrieve fingermarks from items exposed to extreme heat was explored (328), as well as blood marks (329) and marks on cartridge cases (315).

Used acronyms: BY40 (basic yellow 40), CA (cyanoacrylate fuming), NIN (ninhydrin), VMD (vacuum metal deposition), WPS (white powder suspension)

The possibility to recover fingermarks from glass and white ceramic tiles exposed to fire showed that ridge details are still retrievable, especially if the marks were protected from direct exposure to heat above 350°C (328). The most efficient technique was found to be CA followed by BY40 stain (except at 200°C, for which iron powder suspension gave better results). As the temperature increased, it has been observed that the effectiveness of both techniques decreased. However, if the items have been wetted, CA should be discarded and replaced by powder suspensions. It should be noted that the detergent within the powder suspension formulation may help removing the soot on some of the items. Finally, silver VMD could represent a potential alternative for items exposed to higher temperatures (>700°C).

Mock scenes - Mock scenes into which blood marks were planted on various common substrates (e.g. glass & plastic bottles, knife, envelope, magazine) were set in fire then extinguished to assess the possibility to recover marks (329). The author observed that (a) the likelihood of retrieving marks is related with the average temperature in the scene, (b) marks of various qualities (from poor to excellent) were recovered using a LASER (optical observation), acid violet or NIN, and (c) Kastle-Meyer tests (presumptive blood test) gave sometimes negative results on known blood marks.

In another experiment, shotgun cartridges cases bearing latent fingermarks were left on mock scenes which were set in fire, then extinguished (315). In a first attempt, the following sequence was applied on the retrieved cases: "soot removal – visual observation – CA/dye – WPS", but led to no mark detection. In a second set of experiments, electrostatic deposition of charge powder (developed by Bond) was applied and led to the detection of ridges on some (undamaged) cases.

2.3.12 Scenario - Blood marks

A review of the blood fingerprint detection techniques was made by Bossers et al. (330), in which the different techniques are classified according to the detection principles involved. Studies about the detection of marks in blood mostly consist in comparing different known techniques, considering various substrates (324, 327, 331, 332). Two studies are dedicated to the development of new reagents based on wet powder suspension (333) and semi-conductor nanoparticles (281). The compartment of latent fingermarks exposed to blood is the subject of two studies (334, 335). The other publications are focused on the detection of bloodstains, either chemically (336-339), or optically (340-344). The possibility to recover blood marks on common substrates submitted to arson conditions (329) is described in section "2.3.11. Scenario – Arson scenes". The use of functionalized nanoparticles conjugated to antibodies specific to blood and saliva (289) is described in section "2.3.6. Immunogenic detection".

Used acronyms: AB (amido black), AY7 (acid yellow 7), IR (infrared), LCV (leucocrystal violet), SERS (surface-enhanced Raman spectroscopy), TiO₂ (titanium dioxide), UV (ultraviolet).

Comparison studies - The formulation of AY7 has been modified (345). In the new protocol, the fixing step was merged with the staining step. The optimized formulation is easier to use

and appears to be more sensitive. The pH of the solution was also modified to increase the chance of obtaining a DNA profile. Agarwal *et al.* (331) compared Phloxine B and AY7 when applied on dark-coloured substrates. AY7 appears to be more effective than Phloxine B, due to the fluorescence properties of the former. A comparison between three reagents (AB 10b, TiO₂ in methanol and AY7) applied on knives with black handles was performed (332). In this study, AB was shown not to be suitable for the detection whereas the two other reagents lead to good results. TiO₂ is recommended for the detection of aged blood marks and AY7 appears to be a better option for the use on crime scenes, for fresh marks. Other techniques were evaluated on various substrates. AB proved to be the best method for developing blood fingermarks on the adhesive side of duct tape (324). Wetwop also gave positive results with both bloody and non-bloody marks. White Wetwop, containing TiO₂, was proved to enhance the quality of marks in blood on non-porous surfaces (333). It can be used alone or in conjunction with acid dyes, but it is detrimental to DNA. In another study, ortho-tolidine appears to be the best technique to detect fingermarks in blood on skin, compared to AB (327). Semi-conductor zinc sulphide nanoparticles (ZnS) doped with copper have also been used to detect blood fingermarks (281). These nanoparticles proved to be better than AY7 on most substrates.

Blood exposition - Two papers studied the behaviour of a latent mark exposed to blood. Reitnauer (335) focused on sebaceous marks deposited on painted drywalls. It appears that a sebaceous mark exposed to heavy blood deposit will develop the furrows of the impression. Praska and Langenburg (334) did a similar study with marks deposited on glass. They found that a latent mark can be developed after exposition to dilute and whole blood, but this phenomenon did not appear consistently. These marks are distinguishable from blood marks, but may appear as genuine marks after an enhancement with AB and LCV.

Bloodstain detection (chemical) - Three different chemical enhancement techniques for latent bloodstains were evaluated (luminol, Bluestar[®] and Hemascein[®]) (338). All reagents are highly sensitive, but with a surface dependency. Luminol and Bluestar performed similarly, while Hemascein gives poor results on wood surface. It also cross-reacts with many substances, giving more false positive results than the two other techniques. The sensitivity and reliability of Hemascein was also tested (337). It is found to be reliable up to a dilution of 1:100,000 on light-coloured surfaces. Effect of Hemascein on subsequent DNA analysis has yet to be determined. Performances of luminol, fluorescein, hydrogen peroxide, as well as optical methods like UV and IR were evaluated to detect bloodstains on dark surfaces (339). Sensitivity, specificity, ability to work on various surface types and further DNA analysis were evaluated for each method. For the authors, the use of hydrogen peroxide (H₂O₂) is the most efficient method.

Bloodstain detection (optical) - IR photography has been used to detect and localize latent bloodstain evidence lying beneath a layer (or multiple layers) of paint, using a tungsten halogen lamp as source of visible and IR light (341). Blood marks have been detected beneath up to six layers of paint under reflected IR, depending on the characteristics of the paint (especially their IR transmission capability). In addition to IR, bloodstain beneath layers of paint can also be detected using an alternative light source, Bluestar, luminol and fluorescein (336). All techniques are said to be effective. It is recommended to apply the optical methods before any chemical enhancement. Among all chemicals tested, Bluestar produced the best results. IR imaging has also been tested to distinguish bloodstains on fabrics from stains of fruits and vegetables (344). This technique proved to be effective to differentiate blood from

other stains. SERS was also used to detect blood (340). This technique is non-destructive and can successfully detect blood with a dilution of 1:100,000.

Hyperspectral imaging has been used to observe bloodstain patterns on black fabrics, hardly visible to the naked eye (343). In another study (342), the authors describe an evaluation of three different types of light source. These articles are described in section "2.4.1. Photography and alternative light sources".

2.3.13 Fingerprint detection and DNA analysis

The effect of fingerprint detection techniques on subsequent DNA analyses was extensively studied, in terms of potential detrimental effects (346-349), or contamination risks during the detection process (218, 346). Choosing between DNA or fingerprint is also addressed (350), especially when dealing with firearms (351).

Used acronyms: CA (cyanoacrylate fuming), DFO (1,8-diaza-9-fluorenone), PD (physical developer), VMD (vacuum metal deposition)

The effect of fingerprint detection techniques on subsequent DNA analyses was extensively studied (346-349). As already known, most of the techniques do not affect DNA analysis (e.g. dry powders, wet powders, CA, DFO, VMD), but some were identified as deleterious (e.g. PD and silver nitrate). Bhoelai *et al.* also showed that the washing steps (e.g. during CA dye staining) reduced the amount of DNA, and that any immersion step could lead to DNA contamination between samples (346). Norlin *et al.* showed that fingerprints on adhesives (enhanced by wet powders) gave the highest DNA amount, most certainly due to cell shedding caused by the adhesive layer (348). A study showed that DNA could accumulate both inside and outside of a CA chamber, as well as to be transferred between items if processed simultaneously (218). The risks are low but could become problematic if DNA typing systems become more sensitive. The authors propose a number of recommendations to be taken in consideration.

Ferraro discussed about the choice that has sometimes to be made between "swabbing for touch DNA" or "processing the items for fingerprints" (350), as for seized firearms (351). A survey has been conducted with firearms seized within an Indianapolis police district over a two-year-period (Jul'07 to Aug'09). Touch DNA (collected using TriggerPro kits) produced a much larger volume of usable forensic evidence than fingerprints (65.0% vs. 14.3% of the cases, respectively), but identification outcomes for the two methods were equal (2.5% vs. 2.7%, respectively). Considering this, and given that touch DNA takes more time to generate results and is more costly, fingerprint detection remains the most cost-effective technique.

2.3.14 CBRNE-related evidence

Only explosive-related scenario seemed to have been covered during these last three years. Post-blast mock scenes were processed to estimate the chance of recovery of planted marks (352-355). Another goal was to identify the presence of explosive contaminants in fingerprints using analytical techniques (356, 357). It should be noted that all the articles dealing with chemical imaging of explosive-

contaminated fingermarks (261, 358-360) are described in section "2.4.2. Chemical Imaging".

Used acronyms: CA (cyanoacrylate fuming), CBRNE (chemical, biological, radiological, nuclear and explosive), LCV (leuco crystal violet), NIN (ninhydrin), SR-FTIR (synchrotron radiation-based Fourier transform infrared micro-imaging), PETN (pentaerythritol tetranitrate), RDX (research department explosive), RUVIS (reflected ultraviolet imaging system), SPR (small particle reagent), TNT (trinitrotoluene), VBIED (vehicle-borne improvised explosive device).

Explosive threat - The likelihood of recovering fingermarks on various materials used to build an explosive device (e.g. initiators, containers, electrical components, timing mechanisms, adhesives) was discussed (353). Batteries, switches, adhesives, tape, paper and cardboard are the surfaces from which fingermarks could most likely be recovered. Post-blast evidence can be processed by the conventional fingermark detection techniques (e.g. CA, powders, Wetwop[®], LCV). The effects of blast to latent fingermarks left on items present in a VBIED (e.g. cell phone, computer hard drive) and on the vehicle surfaces were studied (354). Observation using a RUVIS, CA with dye staining, metallic dry powders (e.g. gold, copper), as well as SPR were efficient in recovering the latent marks. It should be noted that a large number of the fingermarks in presence were unaffected by the blast effect. The effects of using a water-based disrupting device on a VBIED were also studied, in terms of fingermarks and DNA recovery (355). Fingermarks were left inside and outside the vehicle as well as on objects present inside (i.e. glass and plastic bottles). The disrupting device left a sticky residue after its use (due to the gel sometimes added to the water) which then dries. Fingermarks were successfully detected using dry powder or CA (for small items). Most of the marks were left unaffected (especially outside the vehicle). Metal corrosion on post-blast copper pipe fragments allowed the detection of fingermarks through visual examination or using a selective colour mapping process (352).

SR-FTIR has been used to identify contaminants present in fingermark secretion (e.g. cream, drugs, explosives – PETN, TNT, RDX) (356). It is possible to transfer the marks from a hard-to-reach place, using a Mylar foil as a lifting medium, so that it could be subsequently analysed. The authors emphasize that the location of the latent fingermarks as well as the substances being the source of the contamination were known a priori. A wide-field Raman imaging was used to detect traces of explosives in fingermarks left on problematic Raman surfaces (e.g. plastics, painted metals), using an automated background subtraction process (357).

2.3.15 Miscellaneous detection techniques

A lot of studies dealt with low-pressure sublimation of reagents (361), materials (362-364) or metals (362, 365-367) to detect marks on various substrates. The processing of grease-contaminated marks and substrates has also been a hot topic (368-372). The detection sequence for plastic packaging films was updated (373), and two iodine-fixing reagents were proposed (374, 375). Among the remaining miscellaneous techniques, it is possible to cite: update on the use of silver nitrate (376), electrochemiluminescence to detect marks on conductive substrates (377-379), use of aqueous electrolytes to detect marks on metal (380), measure of the

decay of surface charge to detect marks on plastic (381, 382), spraying of diacetylene monomers (383), chemical lifting of fingermarks from non-porous substrates (384), and electrodeposition of Prussian blue (385).

*Used acronyms: BV2 (basic violet 2), BV3 (basic violet 3 = gentian violet), BY40 (basic yellow 40), CA (cyanoacrylate fuming), CAST (Home Office Centre for Applied Science and Technology), CTF (columnar thin film), ESDA (electrostatic detection apparatus), FTIR (Fourier transform infrared spectroscopy), HDDCPU (2,4-hexadiyne-1,6-bis[*p*-chlorophenylurethane]), HDDPU (2,4-hexadiyne-1,6-bis[phenylurethane]), IND (1,2-indanedione), NIN (ninhydrin), NY3 (natural yellow 3 = curcumin), PET (polyethylene terephthalate), PGME (1-methoxy-2-propanol), PVC (polyvinyl chloride), R6G (rhodamine 6G), Rubpy (ruthenium[III] tris[2,20-bipyridyl]), SB (solvent black 3 = sudan black), SPR (small particle reagent), TPE (tetraphenylethene), uPVC (unplasticized PVC), VMD (vacuum metal deposition), ZnO (zinc oxide).*

Low-pressure sublimation - A prototype system was proposed to allow the application of 7 common reagents (i.e. iodine, CA, CA/R6G, CA/fluorescent dye, IND, NIN, fluorescent powder) on porous and non-porous substrates, without the need of any solvent (361). The technique is based on vacuum sublimation combined with a gas injection delivery system, allowing the reagent to come in contact with the exposed surfaces. The quality of development was shown to be comparable to the traditional protocols (no statistical difference). This process also presents the advantage to have no negative effect on drug chemistry and DNA, but some reagents could have a detrimental effect on inks (forensic document). In the field of VMD, the traditional gold/zinc process was used to visualize grab impressions on fabrics (365). Ridge details could only be obtained on smooth non-porous fabrics (such as nylon), whereas other fabrics lead to the visualization of contact area, which could be further processed for DNA taping. Vacuum deposition of ZnO yielded to the detection of fingermarks on PET plastic substrates, without the need for gold seeds (366). The technique is said to give better ridge details for aged marks (e.g. 45 days). Finally, the sublimation of copper phthalocyanine can lead to deep blue-coloured ridges on light-coloured substrates (367). The technique is efficient on porous substrates (such as paper), but requires exposure times of at least 30 minutes, and was shown to be inefficient on non-porous items (such as glass, uPVC, or ceramic tiles). A specific thermal evaporation of chalcogenic glass or gold under vacuum allowed the visualization of fingermarks on non-porous substrates (e.g., glass, plastic and tape) through the deposition of a small layer of nanoscale wires (i.e. CTFs) (362). The technique is particularly seen as a way to study the topological details of the secretions and determine the sequence of deposition of overlapping marks. The same technique has been applied on untreated marks, as well as on CA-fumed and dusted ones (363), or by thermally evaporating calcium fluoride and silica (364). In this last study, the marks were further enhanced using R6G or IND, which acted as fluorescent dyes for the CTFs.

Grease-contaminated marks/substrate - The detection of fingermarks using lipid staining agents (i.e. SB, BV2 and BV3) is reported (368). The authors observed differences in the staining of sebaceous components by the three reagents, proposing to use them in sequence to increase the likelihood of detection. They also proposed a reduction of the dye stain concentrations by 25% without having a detrimental effect on the staining efficiency. Finally, they identified BV2 as a promising lipid stain (over BV3), mainly for its fluorescence

properties. Another large scale study, encompassing more than 35 domestic greasy contaminants and several detection techniques, led to the proposition of recommended detection sequences (369). Two formulations of SB were compared: one made in ethanol and one made in PGME (370). The PGME-based formulation was preferred in terms of effectiveness and safety of use (lower flammability). The authors also recommend reducing the staining time to less than 2 minutes, to avoid heavy background staining. They also emphasized that old staining solutions could be used, even if it is currently recommended not to use solutions older than 1 month. Finally, a new dye-staining reagent was tested: NY3 (371). Given its fluorescence properties, this stain could replace SB for the processing of contaminated marks (i.e. animal fats and vegetable oils) left on dark non-porous surfaces. In another context, when an item has been (voluntary) exposed to a petroleum-based contaminant (e.g. WD-40, gasoline, kerosene, oils), the use of heptane could help degreasing it before it is processed for fingerprint detection (372). The procedure consists in applying heptane (CO₂-propelled, for example by using "Paslode Degreaser" or "Dynamo") to remove the contaminant, then letting the surface dry before applying a conventional detection technique. By decreasing order, SB, SPR, CA followed by powder, and powder alone allowed the observation of ridge details. A degradation of the marks was observed, especially after 2 weeks in contact with the contaminant.

Flexible plastics - The CAST conducted a study to re-assess the best sequence for processing flexible plastic packaging films (e.g. supermarket bags, trash can liners, protective product films), while voluntarily omitting PVC-based plastics such as cling film and shrink wrap because fingerprint recovery rates from these materials are known to be low (373). The previously-stated most effective technique to be applied on packaging film was VMD (study from 1986), but a decrease of the efficiency of the VMD was recently observed, supposedly due to changes in the chemistry of the plastic material. The new recommended technique is CA/BY40. Powder suspensions are also recommended, as they develop as many fingerprints as CA and present the advantage of working on wetted items. VMD could still be applied, but it is recommended to introduce it after CA or powder suspension.

Iodine - Brucine has been proposed as an efficient way to fix iodine-processed marks, which are known to fade out quite quickly (374). After the fixing process, marks remain visible for one week on non-porous substrates and one month on porous ones. It should be noted that this fixing step seems to have a detrimental effect on the subsequent NIN process, since no marks (or very faint ridges) were visible after applying the amino acid reagent. α -naphthyl amine is also proposed to be used as pretreatment vapors, before iodine fuming (375). As a result, the detected marks appear as red-colored, not fading with time.

Miscellaneous - The use of silver nitrate to detect marks on porous substrates has been re-evaluated by Schwarz & Hermanowski (376). They concluded that silver nitrate could give results on modern papers, but is not recommended for use regarding the appearance of the marks and background staining, especially when compared with the conventional amino acid reagents (e.g. NIN).

Electrochemiluminescence was used to detect latent fingerprints on conductive substrates (377-379). The ridge pattern acts as an inert mask, resulting in negative images of the fingerprints in presence. The visualization is caused by the electroluminescence reaction between Rubpy and tri-n-propylamine, occurring only where the metal remains untouched by the fingertip (377, 378). In another study, rubrene was applied, according to two different

application protocols: being a lipophilic compound, it can be applied to stain the sebum-rich ridges, or it can also be applied to stain the background (379).

Aqueous electrolyte solutions were used to detect fingermarks on metal (i.e. copper, aluminium, iron, brass, zinc) and non-metallic substrates (i.e. glass, plastic) (380). The technique simply consists in immersing the samples in solutions of different pH values (using sulfuric acid or sodium hydroxide) and observing the fingermarks appear.

A method based on the decay of surface charge measured by an electric potential sensor is proposed to detect marks on plastic (381, 382). This technique is different from the ESDA, which is based on the application of a large electric field. It is hypothesized by the authors that the decay of the surface charge may constitute a way to date or estimate the sequence of deposition of the marks in presence.

The spraying of two diacetylene monomers in acetone (i.e. HDDPU and HDDCPU) was shown to successfully detect fingermarks (especially sebum-rich ones) on both porous and non-porous substrates, leading to a purple-on-white contrast (383). Due to the chemical structures of the reagents, it is also possible to chemically image the marks left on an illustrated substrate using FTIR.

Chemical lifting of fingermarks from non-porous substrates using a thermoplastic polyurethane resin combined with fluorescein is reported (384). The marks appear in red, after exposing the film a few seconds to hot air (i.e. 100°C). TPE solution was shown to aggregate into sebum-rich secretion residue left on non-porous substrates (386). Given that TPE is non-luminescent in the soluble state but becomes luminescent after forming aggregates, the resulting marks become blue-luminescent under UV light.

Spatially selective electrodeposition of Prussian blue (385) was performed to visualize fingermarks on conductive substrates. The marks act as masks preventing the deposition of the dye, resulting in a blue-coloration of the substrate only.

2.4 Photography, forensic light sources, and digital/chemical imaging

2.4.1 Photography and alternative light sources

Digital imaging was shown to be useful when suppressing an unwanted background illustration or dealing with round objects (387), as well as for enhancing a coloured mark on a coloured substrate (388, 389). The smart combination of observation filters is not to be neglected given the enhancements that could be obtained (390). Some studies showed the advantages of using laser (391, 392) or LED (393) to record fingermarks. Imaging in the UV (394, 395) and in the IR range (341, 343, 396) showed their advantages. All the articles dealing with the recording of blood marks using alternative light sources (336, 342, 344) are described in section "2.3.12. Scenario – Blood marks".

Used acronyms: CCD (charge-coupled device), DEUS (digital enclosed ultraviolet imaging system), DFO (1,8-diaza-9-fluorenone), IND/Zn (1,2-indanedione/zinc chloride), IR (infrared), LED (light emitting diode), NIN

(ninhydrin), NIR (near-infrared), RUVIS (reflected ultraviolet imaging system), UVC (ultraviolet C)

Photography - Two examples of how digital imaging could help in visualizing fingermarks were proposed (387). The first case consists in suppressing the contribution of an illustrated background for a DFO-processed mark on a printed document. The second case consists in overlapping sequential pictures of friction ridges on a cartridge case, then to merge them to generate a flat panoramic view of the detected mark. The use of colour channels in Adobe Photoshop[®] is illustrated to enhance a NIN-processed mark on a coloured substrate (388, 389). Dalrymple demonstrated that the combination of narrow bandpass filters with a barrier filter could be advantageous when capturing a fingermark in luminescence, especially when the fluorescence of the background may be problematic (390). Optimal conditions (filtering between 470-575 nm) for the recording of NIN-developed marks were investigated as a function of various substrates (397).

Laser - A study aimed at evaluating the best light source to visualize fingermarks detected using IND/Zn, as well as using two emerging amino acid reagents (i.e. genipin and lawsone) (391). The Coherent TracER lasers (460 nm, 532 nm, 577 nm) proved to be the most sensitive at detecting untreated fingermarks, and led to higher ridge clarity. Genipin and lawsone gave unsatisfactory results, and require more development before becoming competitive (formulation and detection protocols). A pulsed Nd-YAG laser and a cooled CCD camera with an image intensifier were used to visualize fingermarks on porous substrates bearing printed texts (392). For this experiment, paper sheets were black-printed using different laser and inkjet printers. The native fluorescence of the marks was observed using optical filters and a time-resolved method. The fluorescence of most printed papers is weak, because ink or toner absorbs the fluorescence of the paper. Excitation at 280 nm is preferred (over 230 nm).

LED - A LED emitting in the IR range (i.e. 940 nm) was used to non-destructively record fingermarks powdered at crime scene, before lifting them (393). By recording in the IR range (900 – 950 nm), black-powdered ridges appear black while multi-coloured background or printings disappear or appear as a single bright colour.

UV - Three fingermark imaging systems based on UVC light source were compared: (a) a DEUS system (home-made UVC-sensitive back-thinned CCD and camera), (b) a RUVIS system UVC-sensitive image intensifier, and (c) a flatbed scanner fitted with a UVC light source (394). The DEUS system gave the best results on porous and non-porous substrates, followed by the RUVIS and the flatbed scanner. It should be noted that using a digital camera with real-time output (i.e. "live" mode) increases the effectiveness of imaging fingermarks. Reflected UV to visualize or enhance latent marks has been extensively described and explained by Richards and Leintz (395). This article is more focused on bitemarks and shoemarks, but constitutes a good overview for people interested in buying the adequate equipment to record reflected UV images (to visualize fingermarks).

IR - A CONDOR Hyperspectral Imaging System was used to visualize untreated fingermarks present on various substrates (e.g. paper, adhesive, aluminum) (396). Data was collected from 400 to 720 nm and digitally processed to reduce the background interference and increase the resulting contrast. This non-destructive method could have its place when chemical treatment is not possible, for example on delicate supports. A visible/NIR CONDOR Hyperspectral Imaging System (650 to 1100 nm) has also been used to observe bloodstain patterns on black

fabrics, hardly visible to the naked eye (343). This technique combines digital imaging with conventional spectroscopy for analysis of samples. In another study, IR photography has been used to detect and localize latent bloodstain evidence lying beneath a layer (or multiple layers) of paint, using a tungsten halogen lamp as source of visible and IR light (341). Blood marks could be detected beneath up to six layers of paint under reflected IR, depending on the characteristics of the paint (especially their IR transmission capability).

2.4.2 Chemical imaging

Chemical imaging has for aim to provide additional information, more than just the morphological one (ridge pattern) (160, 285), for example by enhancing the presence of explosives or metabolites in the sweat residue. Some are non-destructive (e.g. FTIR, Raman, OC-LIBS, CWL), while others require covering the fingermark with a matrix before allowing the analysis (e.g. MALDI). From a chemical point of view, specifically modified CA monomers or reagents were synthesized to be suitable for chemical imaging (221, 222, 383). SERS was used to specifically visualize or target secretion components (292, 398) as well as exogenous contaminants (360, 399). The use of a CWL sensor has been extensively studied to estimate the age of fingermarks (177-182), but also to separate overlapping marks (400) or localize marks on various substrates (401-407). A group of researchers proposed to use a new kind of powder to detect fingermarks and allow their analysis using a MALDI-MS(I) technique (408-410). MALDI-MS can be used in an extended range of scenario (408, 410-412), but is mainly used to visualize exogenous materials contained in the secretions metabolites (408, 410, 412-415), as well as trying to determine the sex of the donor (416). Among the miscellaneous techniques, it is possible to cite: the use of OC-LIBS to localize explosives in secretion residues (358), use of SECM to image fingermarks (359, 417, 418), ToF-SIMS to determine the chronology of events between writing and fingermark deposition (419, 420), ESDA to reach the same goal (421), and SALDI-ToF-MS to detect exogenous material in secretion residues (260-262), and finally capillary-scale ion chromatography to detect gunshot residues (422).

Used acronyms: ATR (attenuated total reflectance), CA (cyanoacrylate or cyanoacrylate fuming), CHCA (α -cyano-4-hydroxycinnamic acid), CWL (chromatic white light), DART (direct analysis in real time), DNT (dinitrotoluene), ESDA (electrostatic deposition detection apparatus), FTIR (Fourier transform infrared spectroscopy), GC (gas chromatography), MALDI (matrix assisted laser desorption ionisation), MeV (mega electron volt), MNT (mononitrotoluene), MS (mass spectrometry), MSI (MS with imaging), NIN (ninhydrin), OC-LIBS (optical catapulting in combination with laser induced breakdown spectroscopy), SALDI (surface-assisted laser desorption ionization), SECM (scanning electrochemical microscopy), SERS (surface-enhanced Raman spectroscopy), SIMS (secondary ion mass spectrometry), TNT (trinitrotoluene), ToF (time of flight), VMD (vacuum metal deposition), XPS (X-ray photoelectron spectroscopy).

A review about the advantages and use of chemical imaging has been proposed by Hazarika and Russell (285), and a comparison between various analytical techniques (e.g. MALDI-MS,

ToF-SIMS, MS, XPS, ATR-FTIR) by Bailey *et al.* (160). In this study GC/MS was found to be the most sensitive to amino acids, MALDI to lipids and peptides, and XPS to the carbon configuration and inorganics. XPS, MeV-SIMS, ToF-SIMS, and ATR-FTIR spectroscopic imaging present the advantage of requiring no sample preparation.

FTIR - Tahtouh *et al.* synthesized modified CA monomers specifically designed to optimize their visualization through an FTIR-based chemical imaging process, while keeping their ability to be fumed on marks (221, 222). Highly interesting results were obtained with one of the monomers (1-cyanoethyl 2-cyanoacrylate) on Australian polymer banknotes, especially on the intaglio printings. De Grazia *et al.* imaged marks processed using diacetylene copolymers on both porous and non-porous substrates (383).

Raman and SERS - SERS was used to visualize fingermarks through the targeting of lipids and amino acid components (398). For the SERS effect to occur, it is necessary that metal nanoparticles are in contact with the analytes. Antibody-functionalized silver nanoparticles were also used to specifically target sweat components, followed by SERS imaging (292). To allow an optimized visualization, the nanoparticles were also functionalized with a Raman probe, i.e. 4-mercaptobenzoic acid, for which the Raman peaks were identified and easily imaged. A semi-automated Raman-based chemical imaging was used to visualize fingermarks, as well as to identify threat materials present in the secretions (e.g. drugs, explosives) (360). To gain a lot of time, only a limited number of points of interest were analysed, selected on the basis of the fingermark optical images. This method also works if the fingermark has been processed with CA. Finally, fingermarks contaminated with β -carotene and fish oil were imaged on various substrates (e.g. paper, cardboard, metal, adhesive) using a line-scanning Raman imaging system (399).

CWL sensor - The CWL sensor is a technology that makes use of the chromatic aberration of light to generate a topographic image of the sample. CWL sensors were used to separate overlapped fingermarks (400), to localize marks on various non-porous substrates (e.g. glass, varnished wood, metal, plastic) (401-403), and is seen as a key element of a contact-less acquisition device (404), which could be used on crime scene (405). A CWL sensor has also been used to estimate the age of fingermarks left on various substrates (177-182), as described in "2.2. Composition, aging and persistence of fingermarks". Another application of a CWL sensor aimed at classifying the surfaces according to texture parameters, and hopefully allowing the detection of fingermarks (406, 407).

MALDI-MS - Contrary to the other non-destructive techniques (such as FTIR or Raman), MALDI-MS requires covering the fingermark with a specific matrix before performing the analysis. A two-step matrix application method is commonly applied in this context, i.e. the "dry-wet" method, for which the matrix is first dusted with CHCA onto the sample then solvent-sprayed (408, 409). More recently, curcumin was proposed as an efficient, natural and colored matrix for MALDI-MS analysis, in replacement of CHCA or as solvent-free matrix (410). The authors using that method emphasize the fact that the powdering step allows by the same way the visualization of the latent marks, given that the matrix absorbs UV light and fluoresces. A review of the use of MALDI-MSI to visualize fingermarks is proposed by Francese *et al.* (411). In details, MALDI-MSI was used to visualize fingermarks (or separate overlapping fingermarks) using ion signals that are characteristic of secretion endogenous species (e.g. amino acids, lipids) (408, 410, 412), metabolites (413), and contaminating substances such as condom lubricant (414, 415), antiseptic (408), or drug (410, 412). Some authors also claimed being able to determine the sex of a fingermark donor by using MALDI-

MS (success rate from 67.5 to 85%) (416). This study was based on multivariate modelling of mass spectrometric profiles of fingermark peptides and small proteins contained in the secretion.

Miscellaneous - OC-LIBS has been used to analyse explosive residues (i.e. TNT, DNT and MNT) in contaminated fingermarks left on glass (358). Discrimination between explosive and non-explosive materials is possible.

SECM has been applied on fingermarks which were artificially contaminated with an explosive (i.e. picric acid) (359) or left on glass and detected using an alternate VMD process (i.e. Al-ZnO) (417). SECM has also been applied to detect fingermarks on metal substrates (i.e. platinum, gold, silver, copper and stainless steel) (316), or various substrates (418). SECM is a technique based on the response given by a local oxydo-reduction reaction, which takes place if a target is present in the sample.

ToF-SIMS chemical mapping was used to determine whether a fingermark has been deposited before or after a text was written (419) or printed using a laser printer (420). This technique requires: (a) the presence of some endogeneous ions in sweat and not in the laser ink (e.g. Na^+ , K^+ and C_3H_5^+ ions), and (b) the visualization of these ions only if the mark is deposited above ink (and has consequently been left after the ink was printed). However, the ink signal could sometimes be visualized from beneath the ridge, or be lower than expected even when lying on top of the fingermark (419). The ESDA was also shown to allow determining the order of deposition (fingermark or ink) when processing laser-printed documents (421). If a text is printed after a mark has been left, the ESDA will result in unbroken white lines, whereas the opposite scenario (i.e. a mark left on a printed text) will result in dark lines bearing ridge details. It should be noted that: (a) the technique can be applied after a NIN process, and (b) the sequence determination success rate drops quickly as the mark age. The order of deposition between latent fingermarks and laser printed ink has been examined using chemical mapping with secondary ion mass spectrometry (423). Blind testing on 21 samples results in correct determination for all samples.

SALDI-ToF-MS was used to detect terbinafine (i.e. a medication) as a metabolite in sweat secretions (260). To reach this goal, magnetizable carbon black-doped silica nanoparticles were used to dust the fingermarks and act as signal enhancing agents for SALDI-ToF-MS. The same particles were used to detect the presence of explosive in sweat secretion on various substrates (i.e. stainless steel, glass, paper, plastic bag, metal drinks can, wood laminate, adhesive and white ceramic tile), using SALDI-ToF-MS and DART-MS (261). Seven common explosives were used (i.e. six nitro-organic- and one peroxide-type) and were detected in the nanogram range. The same nanoparticles and analytical technique were also used to study the composition of the secretion residue in terms of polar and non-polar constituents (i.e. amino acids and squalene / fatty acids, respectively) (262).

Capillary-scale ion chromatography was applied to detect gunshot residue or black-powder contamination in secretion residue, as well as exogeneous species in the sweat of smokers (422).

3 Miscellaneous marks

3.1 Earmarks and earprints

The possibilities of identification offered by the comparison between earmarks and earprints are still the subject of a few publications. The operational successes obtained in the region of Hamburg have been reported (424). The paper also provide an extensive bibliography related to the early work carried out in Germany in that area.

A method of earprint deposition has been proposed (425). An apparatus based on an ear defender headset, integrating a spring that allows controlling the force with which an ear is pressed to a substrate is presented. The ears were coloured beforehand with yellow vegetable dye. High reproducibility of the measured variables on different earprints, taken by different operators, was achieved.

A pilot study of ear identification based on photographs, aiming at the investigation of personal identification by the ear from surveillance videos has been carried out (426). The authors divide the ear into four regions (concha, helix, antihelix and lobe), and measure the relative surface (with respect to the entire ear) of these different surfaces. Good reproducibility (within and between observers) is found, and a low probability of observing the same measurements on two different ears has been computed using a parametric model.

One study (427) investigates sex differences in the external ear of the Indian population and finds that there are differences between male and female donors with respect to lobe length and breadth as well as ear length, breadth and the height at the base of the auricle.

Junod and colleagues (428) presented an automatic system allowing the matching between earmarks and earprints. The system also allows assigning weight of evidence (in the form of a likelihood ratio) to each comparison undertaken. The authors detailed the system performance including measures of the rates of misleading evidence. For mark to print comparison, the equal error rate is 2.3%. The system has been tested on a database of 1229 donors and also in cases from police forces. A review of automatic systems used from earmarks and earprints has also been published (429).

3.2 Foot morphology

The link between foot dominance and morphological characteristics as well as the link between foot and hand dominance have been investigated (430). These links would allow, from barefoot impressions from the crime scene, to determine first the dominant foot and then the dominant hand. Foot width and two foot lengths (related to the first and second toe, respectively) were used as descriptors. Results did not show very clear relationships between these factors.

Hammer and colleagues (431) studied the possibility of carrying out comparisons between the impressions on shoe insoles with inked comparison material. Both impressions from insoles and inked materials were used for these comparisons. A number of measurements (chosen for discrimination as well as discernibility on the insoles) were carried out and compared, and overlay comparison was also used. Like-to-like (insoles to insoles) comparisons showed more similarity when indeed from the same source; it was however still possible, in this study, to

attribute the impressions on shoe insoles to the right source using inked impressions as a reference. In casework involving a question about the mark on the insole of a shoe, the authors recommend using shoes known to have been worn by the putative source as comparison material.

3.3 Lipmarks

Reviews on lip prints as well as their forensic use have been carried out in the time covered by the present review (432-434). Vanishree and coauthors (435) describe detection techniques useful for the visualisation of latent lip marks. Three dyes (Sudan Black, vermilion and indigo) have been compared for the visualisation of lipmarks left with classic or long – lasting lipstick on china as well as cotton and satin fabric (436). Their performance has been found to be similar.

Several studies assess the frequencies of different lip patterns in populations (437-445); with the exception of (441) and (444) these studies also find differences in the frequencies of patterns between the genders. Verghese and Mestri describe frequencies (446) and furthermore exclude a link between lip patterns and blood group. The relationship between sex and lip patterns (447) and between age, sex, and lip patterns (448) has been investigated in more detail; sex differences have been found (447), but in (448) they depended on the age class of subjects. Ludwig and Page (440) also investigate the comparison between photographs of lips and lip impressions using more intricate detail than just the classification results, and present such comparisons in detail. One study aims at establishing uniqueness of lip prints based on a sample of 200 individuals, including five pairs of twins (449). The authors also show some similarity of patterns between parents and children on the basis of five families (consisting of mother, father and 2 children). Finally, the lip prints of 20 individuals were recorded at a 3-month interval to show permanence (449). A similar study investigating individuality and permanence (over the time of one year) has been carried out (450). Three other studies investigated the question of uniqueness on a sample of 100 individuals (451), one of 200 individuals (452) and on one of 124 individuals (453). Choraś (454) proposes a method for automated feature extraction from lips.

3.4 Identification of deceased individuals

Campbell (455) describes the retrieval of a fingerprint from the underside of the epidermis of a body whose outer epidermis was too decomposed to obtain a good image. Subsequently, a hit in the AFIS was obtained. A revivification method of the epidermis used in Germany has also been published (456).

An analysis of the identification methods used on 134 bodies of unknown identity shows that 10 were identified by their fingerprints. Such identification was only carried out when a pre-mortem set of prints of the suspected identity was present in the national database (457). The admissibility of fingerprint evidence, in particular in the U.S. and Canada, is mentioned in an article detailing different means of identifying deceased individuals (458). In order to properly identify deceased individuals in an institute of legal medicine, livescans of two fingers were taken from bodies upon entry, and the identity verified when the bodies were released (459). This was used as an additional insurance of the proper identity. Identification of deceased individuals when identity theft has occurred is the object of another article (460); several case reports including cases where identification through fingerprints is problematic due to identity

theft are presented (460). The fingerprints of 109, up to then unidentified human remains, have been sent to larger fingerprint databases (Department of Homeland Security Biometric Support Center and the FBI Criminal Justice Information Services Special Processing Center rather than the local database). This allowed the identification of 51 of these cold cases (461). The special case of a 2650 year old body has been reported during the period of review (462); using photography and the image enhancement tools of an automated fingerprint identification system, the general patterns of the fingers of the right hand were still visible, and there were enough minutiae on the image of the right thumb for an individualisation. The practice of retrieving latent impressions at the residence of a presumed identity of a deceased individual in order to identify this individual is described and defended using Occam's razor (463). The same argument is also applied then to fake fingerprints, stating the fact that in general, the most simple explanation of the presence of a mark on a scene is touch by the finger rather than planting.

3.5 Various subjects

In order to detect / avoid tampering with raw fingerprint images in biometric systems, a watermarking method is proposed by Li (464).

The marks left while wearing gloves have been studied (465). The authors indicate how impressions from friction ridge skin may be left even when gloves are worn, when the material constituting the glove is very thin and flexible.

4 Crime scenes and case reports

Used acronyms: CA (cyanoacrylate fuming), DFO (1,8-diaza-9-fluorenone), ORO (oil red O), RAM (rhodamine – Ardrox – methylene blue)

Beaudoin has reported the use of ORO on a 21-year-old cold case involving the processing of papers (used to start a fire) (466). DFO was applied first, giving negative results, and was followed by ORO, which led to the detection of two fingerprints. In this article, the recipe and application protocol are described.

The use of a 532 nm TracER laser led to the observation of an additional fingerprint on a duct tape processed with superglue followed by TapeGlo (fluorescent stain) (467). This mark was barely visible using a conventional alternate light source (Omniprint 1000B).

Successful recovery of latent marks on an *Agave Americana* (six-foot-plant with thick green leaves) was reported in the context of a home robbery (468). Black (magnetic) powder followed by lifting, and CA followed by RAM dye staining were chosen.

The inside of interior door handles should not be neglected when processing a (stolen) car, given that very good quality marks may be detected (469).

The case reports related with cartridge casings (317-319) are described in section "2.3.8. Substrate – Metal and cartridge cases".

Rubber-like casting materials (i.e. Accutrans and Reprorubber) were chosen to allow the fingerprinting of an Egyptian mummy without causing damage to it (470).

Wendt *et al.* (471) presents the setting of the new fingerprint detection laboratory in Kiel (Germany).

The management issues associated with a fingerprint unit are covered by Tomaszycski (472).

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