Chapter 3: Estimating the age of fingermarks: relevance, potential approaches and perspectives

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Abstract

The age estimation of (latent or visible) fingermarks recovered at crime scenes may be particularly useful to be able to discern traces relevant to the investigated event or to help place a particular event in time. The measurement of physical or chemical changes of fingermark characteristics over time has been proposed as a mean to achieve this goal. However, fingermark depositions are complex matrices that are influenced by many factors besides time, such as donor, transfer and environmental conditions to cite only a few. Thus, the main challenge resides in the identification of aging parameters that evolve over time with as little influence as possible from such factors. The development of reliable aging models that can be implemented in practice to place fingermarks in time requires the acquisition of large amounts of fundamental data on the aging processes of target compounds, if possible, using versatile and easily available analytical techniques. The importance of the data interpretation stage should not be underestimated either. Thus, while age estimation of fingermarks would be particularly useful for crime reconstructions or as evidence in court, much more information is currently needed on the aging of natural fingermarks. A research cycle is proposed to address this issue in a more systematic manner, and several cases in which fingermark dating was either attempted or debated are discussed from a forensic perspective.

Keywords: Fingerprint, latent and visible fingermarks, composition, aging, dating, time.

Acronyms

AFM: Atomic Force Microscopy

CWL: Chromatic White Light

DNA : Deoxyribonucleic Acid

FTIR: Fourier Transform Infrared Spectroscopy

GC: Gas Chromatography

HIS: Hyperspectral Imaging

LC: Liquid Chromatography

LR: Likelihood Ratios

MALDI: Matrix Assisted Laser Desorption Ionization

MS: Mass Spectrometry

PA: Peak Area

PCA: Principal Component Analysis

PLSR: Partial Least Squares Regression

QCM: Quartz Crystal Microbalance

RMSE: Root Mean Square Errors

SIMCA: Soft Independent Modelling of Class Analogy

SIMS: Secondary Ion Mass Spectrometry

TLC: Thin-Layer chromatography

TOF: Time-of-Flight

UP: Ultra-Performance

UV-VIS: Ultraviolet Visible Light

3.1. Introduction

Determining the time of fingermark deposition has long been a major concern for forensic scientists and still remains a complex challenge in practical settings [62,66,80,81]. Information on time of placement serves several purposes. On one hand, when the time of the offense is (approximatively) known, it enables the selection of relevant traces[1] to be collected for further examination [61] i.e., only traces transferred during the criminal event, rather than before or after, are collected. Furthermore, it allows the selection of adequate detection techniques to enhance latent traces. For example, while the ideal enhancement techniques for fingermarks should work on all kinds of traces, some are reported to perform better on fresher or older marks, such as iodine vapors or physical developer, respectively. This is especially important when the elapsed time between the offense and the investigation reaches a significant interval (e.g. several weeks, months or even years), thus increasing the risk of degradation of the pertinent enhancement targets, as well as the risk of contamination of the crime scene. On the other hand, when the time of the offense remains unknown, estimating the age of relevant traces may help reconstruct the investigated events in time (e.g. estimation of the time-of-death through the measurement of body temperature).

While the main use of fingermarks by law enforcement agencies remains the identification of its source (i.e. **who** has left the trace), the answers to **how** and **when** were the marks transferred on crime scene are mostly implicitly inferred from **where** the marks were detected. Indeed, a (latent) fingermark discovered on a glass or bottle of wine will have less significance than one on the murder weapon, particularly when the identified person had a legitimate reason to visit the crime scene before the crime occurred (e.g. family, friends, acquaintances). The location of fingermarks at the scene thus helps reconstruct the activities of the people potentially implicated in the crime (e.g. drinking a glass of wine or handling a knife). However, the question of time has to be explicitly addressed and becomes highly relevant when a person admits to having handled the knife – generally

[1] A trace can be defined as a mark, signal or object. It is an observable sign (not always visible to the naked eye), the vestige of a presence or an action at the place of the latter.

well before the crime occurred – but for legitimate reasons, such as cooking. In such cases, the identity of the person is no longer under question, but the time at which the mark had been transferred is [44]. Failing to consider the question of **when** in such cases could lead to potential wrong convictions, which highlights the relevance of estimating the age of fingermarks, as well as other forensic traces.

Focusing principally on fingermarks, three main approaches have been discussed in the literature to situate them in time, considering [44,81]:

1) investigative information (e.g. if windows are regularly cleaned, then the maximum interval between transfer and discovery of traces on the glass can be extrapolated)
2) chronology of deposition (e.g. when two traces overlap, determining which one was deposited first)
3) physical and chemical changes of fingermark characteristics over time (e.g. degradation of the friction ridge skin detail[2] or chemical components)

This chapter will mainly focus on the third approach, i.e. the measurement of changes, generally aging processes, to estimate time-since-transfer. The requirements for the development of dating methods will first be detailed. Then, the potential and limitations for implementation in practical settings will be discussed through the presentation of case examples.

3.2. Requirements for the development of an approach to estimate the age of fingermarks

As previously described in the literature, the development of a methodology to estimate the age of fingermark residue is a very complex challenge [18,44,66]. While operational methods have been reported for the dating of other forensic traces, such as ink or gunshot residue [6,8], their implementation in practice still remains to some extent a contentious

[2] Friction ridge details comprise the combination of friction ridge flow, friction ridge characteristics (including level I, II and III) and friction ridge structure

issue. Girod *et al*. [44] proposed a research cycle to develop and test such methods for practical application (Figure 1).

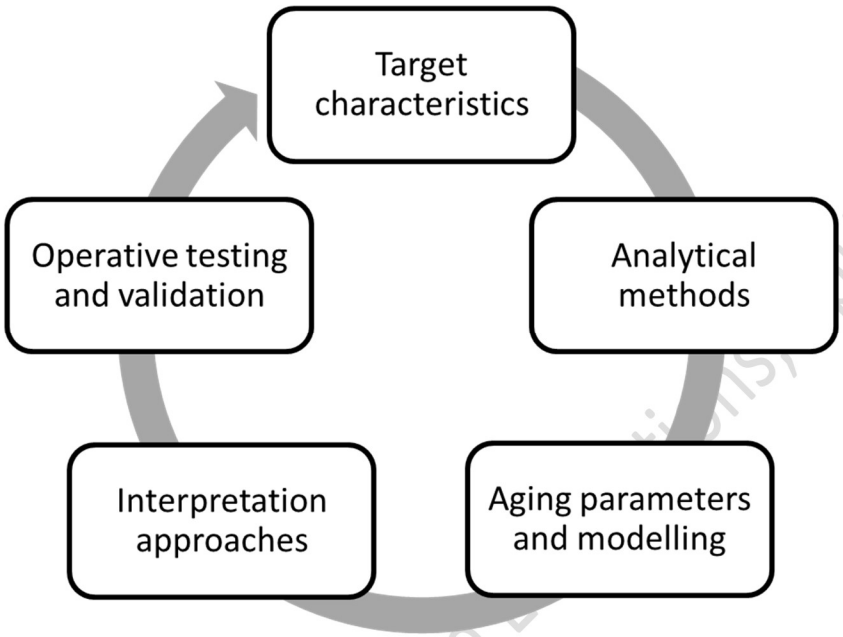


Figure 1 – Proposed research cycle to develop dating methods of forensic traces (adapted from [44]).

3.2.1. Target characteristics

The key strategy to develop dating methods based on aging processes is to select target characteristics that actually change over time in a reproducible and measurable way (e.g. the decrease of squalene[3] in the residue [11,82] or the minutiae count in the ridge pattern [23] after fingermark deposition). These characteristics should ideally be present in the residue of all donors in order for the method to work on a broad range of fingermarks found at crime scenes [15,39,47]. Furthermore, the targets should be easily analyzed using analytical methods available in forensic laboratories. Promising candidates are discussed below (see also Table 1). See also Chapter 7 for detailed descriptions of the chemical degradation processes.

[3] Squalene is an organic compounds found in human sebum, and is an essential precursor in the biosynthesis of cholesterol, steroids and vitamin D.

In addition to the visual characteristics linked to the ridge detail, many potential target compounds have been identified in fingermark residue and mainly originate from the secretory glands found in the dermis (i.e. the bottom layer of skin): the apocrine, eccrine and sebaceous glands [14,40,45]. Apocrine glands are found only in the genital, mammary, inguinal and axillary regions and thus, their secretions do not represent particularly interesting targets for enhancement or dating purposes (i.e. compounds should be found in a majority of fingermarks to be considered relevant targets). Eccrine glands on the other hand, are found over the whole body with greater masses on the palmar and plantar surfaces. While the initial water content of eccrine secretions can be significant (generally between 20 to 70% of the fingermark's initial weight, and up to 90% when hands have been recently washed [56]), several other components have been identified as potential targets to monitor fingermark aging such as proteins, peptides, amino acids and chlorides. Finally, sebaceous glands are not present on the friction ridge surfaces of the hands and feet. However, many sebum components are still found in fingermark residue because of frequent contact between the hands and body parts such as the face and hair. Lipids secreted by the sebaceous glands have been identified as particularly interesting target compounds to study aging. Natural fingermark residue is generally composed of a mixture of eccrine and sebaceous secretions with external contaminants (e.g. cosmetics, food residue, dust, bacteria, nicotine). The latter are rarely targeted as they are not found in all fingermarks. However, cosmetics often contain compounds that are also found in natural fingermarks, such as fatty acids or wax esters. This can increase the potential of such compounds for enhancement of fingermarks and may influence their aging processes.

Targets	Occurrence	Studied time range	Analysis method	Selected references
Ridge detail	in relevant marks	up to several years	2D Visual examination 3D Imaging HSI AFM TOF-SIMS	[19-23,65,68,72]
Water	in all fingermarks	a few minutes	QCM	[56,57]
Chloride	in all fingermarks	*ca*. 200 days	Silver nitrate	[10,16,45]
Tryptophan derivatives	in all fingermarks	a few weeks	Spectroscopy TLC	[79]
Lipids	in all fingermarks	several weeks	FTIR RAMAN GC-MS LC-MS/MS MALDI-MS	[11,29,38,46,48,52,54,71,82,85]
Amino acids	in all fingermarks	*ca*. 200 days	RAMAN GC-MS	[15,16,24]
Proteins	in all fingermarks	a few weeks	Gel electrophoresis LC-MS/MS	[31,69]

Table 1 – List of possible target characteristics/compounds reported in the literature to describe fingermark aging. QCM: Quartz Crystal Microbalance, HSI: Hyperspectral Imaging, AFM: Atomic Force Spectroscopy GC: Gas Chromatography, LC: Liquid Chromatography, MS: Mass Spectrometry, TOF-SIMS: Time-of-Flight Secondary Ion MS.

It was first proposed to investigate fingermark aging through changes in the quality of the visible two dimensional ridge details and loss of water [18-23,46,72,80]. While still intuitively used by some investigators, such observations were not deemed reliable for operational purposes yet and extreme caution should be taken when attempting to correlate pattern quality with time since deposition. Concerning water loss, a recent study indicated that initial water content varied significantly between marks and that evaporation occurred within minutes after deposition [56]. Given the short timeframe of water loss, it

cannot be considered as a relevant target component to study aging from a forensic perspective. Since then, several parameters have been suggested to quantitatively describe physical changes in the ridge details over time: minutia count, color contrast, ridge discontinuity, ridge width and height [19-21,23]. While promising results were obtained, additional multi-variate data is necessary to further investigate fingermark aging using such characteristics.

Lipids are one of the main classes of compounds of interest in aging studies. Free fatty acids, sterols, wax esters, mono-, di- and triglycerides were previously identified in the fingermarks of many donors [13,40,45]. Several studies indicated that fatty acid evolution did not show a clear tendency over time and were thus barely investigated to monitor aging [11,82]. However, the degradation of two unsaturated fatty acids (Δ6-hexadecenoic acid and Δ8-octadecenoic acid) was recently studied over 14 days, as well as products of the oxidation process of these two target compounds [71]. Squalene, a precursor of steroids, and cholesterol were detected in fingermarks of all donors and decreased relatively quickly in the first days after deposition, following an exponential decay up to *ca*. 30 days. However, large differences were detected between donors and normalization of the data was proposed to improve reproducibility [11,29,30,38,46,82]. Many wax esters were also identified in fingermarks [59] and some were found to decrease over time, such as isopropyl dodecanoate [46]. Wax esters were, however, detected in relatively small amounts in fingermarks compared to sterols and fatty acids [47] and their decrease over time has not been systematically characterized yet. Finally, intact di- and triglycerides have recently been characterized, and shown to decrease rapidly over time following deposition [41,42,52,70]. Degradation of the above lipids appears to be largely due to ozonolysis, a process by which ozone reacts with the double bonds of unsaturated compounds, yielding a number of ozonides (1,2,4-trioxolanes) and shorter chain oxidation products. More systematic studies might yield useful additional data to investigate fingermark aging using such target compounds.

Many studies into fingermark composition actually use "groomed"[4] marks to increase the amount of lipids in the residue. Donors are usually asked to rub their fingers on their forehead or neck before deposition. Thus, most studies focusing on lipids will actually report quantities that may be significantly higher than in natural marks. While some of these studies proved the feasibility of studying the degradation of fingermark lipids under laboratory conditions ("proof-of-concept"), it would be necessary for operational applications to study the initial lipid composition and subsequent aging of natural fingermarks instead of "groomed" ones [7].

Eccrine components have been less studied than lipids and it was only recently suggested that their degradation patterns over time may be of interest for fingermark aging/dating purposes [69]. Amino acids were characterized in natural and groomed fingermarks [15]. However, no data was reported about their aging in ambient conditions and these compounds were thus not considered yet for estimating the age of fingermarks. While still present in older fingermarks [50], a decrease in amino acids was reported in several publications over longer time periods or due to thermal degradation [16,24]. Thus, these compounds might present interesting alternatives to study longer time spans, if the observed decrease can be confirmed to occur in a reproducible manner. Proteins and peptides were also characterized in fingermarks and recently proposed to monitor aging processes [31,69]. Among 31 identified proteins, 4 keratins and dermcidin showed significant (increasing or decreasing) trends over 16 days [69]. Possible modifications occurring over time include alkylation, disulfide reduction, ammonia loss and potassium adducts. As sample preparation can also influence degradation, further tandem mass spectrometry studies over longer time periods would bring a better understanding of these aging processes and potentially help identifying further potential targets.

A decrease in the auto-fluorescence of fingermarks was also observed and attributed to protein-bound tryptophan derivatives, including indoleacetic acid, (nor)harman and xanthurenic acid [79]. These compounds were also suggested as potential aging markers, but changes were not studied quantitatively over time. Finally, the diffusion of chloride

[4] "Groomed" refers to fingermarks that are artificially charged, for example with sebaceous secretions by rubbing fingertips on the forehead before deposition.

components was proposed over 40 years ago as an interesting parameter to study the aging of fingermarks on paper substrates [10,16]. However, the reliability of this approach was not demonstrated, and chlorides were not further evaluated as potential aging markers.

The main difficulty when selecting target compounds to model aging lies in the fact that many intrinsic (related to the fingermark itself) and extrinsic (related to the substrate and environment) factors influence the aging processes of fingermarks [12,13,45] (Figure 2). It was previously demonstrated that fingermark composition can vary significantly between different donors physiologically, but also due to activities such as food handling, hand washing, sweating or use of cosmetics (t<0). Then, the initial composition transferred to the substrate will additionally be influenced by the deposition conditions (t=0). Factors such as substrate type, temperature, pressure and duration of the contact thus have a significant influence. Finally, the conditions and duration of storage will have a major impact on the aged composition of the trace found during crime scene investigation (t>0). Target compounds should ideally be influenced as little as possible by these factors, or the aging model should be able to account for their impact.

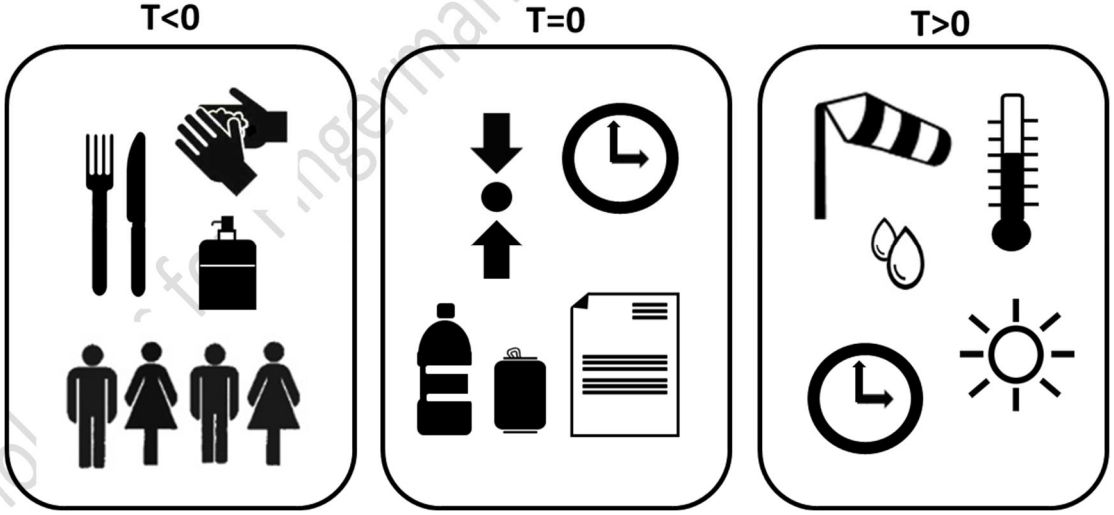


Figure 2 – Factors influencing the composition of fingermark residue over time. The deposition occurs at time t=0.

3.2.2. Analytical methods

The main forensic criteria for adequate analytical methods are cost, rapidity, simplicity, reliability, non-invasiveness and versatility (see Table 2). Thus, selected techniques should be able to carry out analyzes efficiently (with minimal resource requirements), in a reproducible way and detect a wide range of targeted compounds with minimal sample preparation in order to allow subsequent analysis steps. As some approaches may be destructive of the ridge details, a fingermark will generally have to be detected (possibly using enhancement techniques) and recorded before age estimation analysis. Furthermore, if a relevant mark is of insufficient quality to infer who has deposited it, then the sampling and analysis of contact DNA (or touch DNA) will generally be attempted. Thus, analytical techniques and methods of interest for age estimation should be compatible in sequence with both fingermark enhancement techniques and DNA analysis [59,77].

Several (mainly optical and non-destructive) techniques have been reported to describe physical changes that occur on a fingermark ridge pattern over time. Natural and groomed fingermark aging was studied over 20 hours using hyperspectral imaging (HSI) [63]. Atomic force microscopy (AFM) was also suggested to measure changes in the physical characteristics of fingermarks over time [25,27,72]. A decrease in the height of the ridges and horizontal migration across (non-porous) substrate was recorded over 2 months [72]. Physical changes in fingermarks have also been studied over *ca*. 1 month using electrochemical impedance spectroscopy [73]. Measurement of the contrast modifications between fingermarks ridges and background using a chromatic white light (CWL) sensor and/or high resolution images was also proposed [21,64]. More recently, very simple optical approaches have also been introduced to study visible changes occurring in the 2- and 3-dimensional pattern, including minutiae, ridge continuity, contrast, ridge width and height [19-23]. Optical examination combined with quantitative measurements would make such approaches ideal from a forensic science perspective. See also Chapter 4, 5 and 6 for further details on the measurement of physical characteristics.

On the chemical composition, while less detailed information is generated using optical methods, these are still generally favored for forensic operational purposes, as they

comply with several main criteria such as non-invasiveness as well as simplicity, rapidity (minimal sample preparation) and lower costs, compared to more specific analytical approaches. Fourier Transform Infrared Spectroscopy (FTIR) was one of the main optical techniques used to characterize fingermark composition, specifically the lipid components [43,55,76,83]. The amino acid serine was also targeted in one study [55]. While the influence of the donor's age was evaluated using FTIR [51], fingermark aging was also studied over approximately one month, taking into account the influence of substrate, light and moderate temperature [48,54]. While FTIR is a promising approach to study lipid aging, limitations have also been identified. Indeed, this technique is only semi-quantitative and cannot be applied on all type of substrates. It works best on aluminum foil and other non-porous substrates such as glass but cannot be applied on porous substrates such as paper, thus limiting its applicability in forensic practice.

An optical alternative was recently suggested using Raman spectroscopy. While mainly used to identify exogenous compounds such as drugs [17], it has also been applied for the unspecific detection of eccrine and sebaceous secretions [9,28,36,85]. Some signals attributed to squalene and fatty acids, showed a decrease over 1 month. However, high variability between measurements were also discussed and results should be carefully interpreted using sufficient replicate analyses of specimens [9].

Many analytical techniques have been proposed to monitor the chemical composition of fingermarks over the years. While generally more invasive than optical methods, they are also more specific, quantitative and reproducible. Their potential for fundamental research is evident, however they are also often more expensive, time-consuming and more complex to apply, and thus less adapted to forensic routine practice. While thin-layer chromatography (TLC) was first employed [26,33], it was soon replaced by other, more specific techniques such as gas chromatography (GC) or liquid chromatography (LC), nowadays generally coupled to mass spectroscopy (MS). Both techniques allow the study of a wide range of compounds from sebaceous and eccrine secretions. GC-MS generally targets more volatile and thermally stable compounds (e.g. small lipids and amino acids) [11,38,46,71,75,82], while ultra-performance (UP)LC-MS/MS is suited to larger molecules

such as glycerides, peptides and proteins [41,67,69,71] as well as to elucidate the structure of the target compounds and aging products [29,41,67].

Several alternative mass spectrometric methods have also been used to study the fingermark chemical composition, and more specifically chemical imaging of endogenous and exogenous compounds [37,53,84]. Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) imaging was used to study diffusion of saturated fatty acids over time [68]. Some techniques, using lasers and or electrospray ionization, are minimally invasive at least for less volatile compounds such as lipids and proteins (as most techniques require a vacuum for ionization of the molecules), however such instrumentation is very expensive, and their use requires a high level of skill and maintenance, making them less adapted to routine application. While optical methods do not suffer from these disadvantages and are generally more easily applicable directly on the crime scene (i.e., when portable devices are used) [34], they are also less selective and reliable to track quantitative changes in several target molecules over time. Thus, no method fulfils all the forensic criteria and the methods listed in the two central columns of Table 2 would represent the best compromise to follow the chemical degradation of fingermarks, with more reliability and selectivity for GC-MS and LC-MS and better availability and less invasiveness for HSI and FTIR.

Forensic criteria	Optical examination	Spectroscopic methods	Basic separation methods	Advanced technologies
Examples of techniques	Light source / Imaging (3D OP)	HSI, FTIR, RAMAN	GC-MS, LC-MS	UPLC-MS/MS MALDI-TOF-MS
Purchase and maintenance costs	low	low to medium	relatively high	very high
Analysis time	quick	a few minutes (+ calibration)	30-60 min. (+ calibration)	variable, longer for imaging (+ calibration)
Simplicity	basic skills	basic to advanced skills	advanced skills	expert skills
Reliability	low to medium, mainly qualitative*	high, but mainly qualitative	high, quantitative	high, quantitative
Non-invasiveness	yes	yes	no	Variable
Versatility and selectivity	visible pattern	class of compounds, imaging potential	volatile and thermally stable lipids, derivatized amino acids	larger lipids, peptides and proteins, imaging potential
Availability	All forensic laboratories	Broadly available	Specialized laboratories	Rarely available
Portability	Yes	Yes	Limited	No

Table 2 – List of criteria for the techniques able to detect fingermark aging characteristics (physical and chemical) over time. *Quantitative data can be extracted from high resolution 2D and 3D imaging (3D Optical Profilometry), this would potentially increase the reliability of some optical examination techniques.

Aging parameters and modelling

Once potential target characteristics have been identified, their aging processes must be examined and modelled as a function of time, including the effect of influencing factors to reproduce realistic conditions as encountered in practical settings.

One study focusing on the measurement of the (color) contrast differences between ridges and background proposed an aging parameter based on pixel colors to model fingermarks aging over a period of 24 hours. It was reported that aging followed a logarithm curve. However, aging could not be modelled over a longer time span (due to large inter- and intra-variability), thus limiting the potential of this method for operational purposes [64]. Several recent publications proposed to follow the evolution of minutiae count, color contrast between ridges and furrows, ridge discontinuities and ridge width as aging parameters over 6 months using linear regressions for simplicity purposes [19-21,23]. Their results suggested that the unique combination of environmental factors had a significant distinct impact in ridge degradation. In some instances, no visual degradation could be detected (e.g. sebaceous-rich fingermarks on glass exposed to direct light), while in other cases complete obliteration of the mark was observed (e.g. eccrine-rich fingermarks on plastic stored in darkness) over the exact same time periods. This demonstrated, again, the relevance of the interaction of factors, and the value of each aging parameters, and that they should be considered in the aging models in a multivariate approach to increase reliability.

Concerning chemical target compounds, it was observed that different donors present very variable initial fingermark composition, at least quantitatively. Furthermore, while inter-donor variability is significantly higher than intra-donor variability, variation has also been measured between fingermarks from the same donor [47,49]. Thus, the calculation of ratios between endogenous compounds has been suggested as an approach to significantly decrease the effects of quantitative variability of individual compounds. The ratio of two main compounds (i.e. the peak area (PA) of squalene was divided by the PA of cholesterol) was proposed to characterize the decrease of these lipid compounds over time [82]. In order to further reduce variability, other pre-processing steps were tested

using the PA of compounds analyzed by GC/MS. The best pre-processing technique was determined to be the square root of the PA of a target compound normalized to the sum of PA of other compounds [46]. For the sum of the denominator, 7 lipids were selected: isopropyl dodecanoate (IDP), three squalene derivatives (SD1-3), cholesterol (Chol), myristyl palmitoleate (MPO) and myristyl palmitate (MP):

$$\text{Aging parameter MPO} = \sqrt{\frac{PA_{MPO}}{PA_{IDP}+PA_{SD1}+PA_{SD2}+PA_{SD3}+PA_{Chol}+PA_{MPO}+PA_{MP}}} \qquad \text{Equation 1}$$

Exponential linear regressions were used to model the decrease of the normalized aging parameters. While squalene was reported as a promising target [46], the aging of other compounds can be modelled, as shown in Figure 3 for the aging parameter MPO (Equation 1).

Ratios were also proposed to follow triglyceride degradation [70]. The decrease of the ratio TG48:1/TG48:0 was followed over 24 days, as well as the increase of the ratios of monoozonide products of TG48:0.

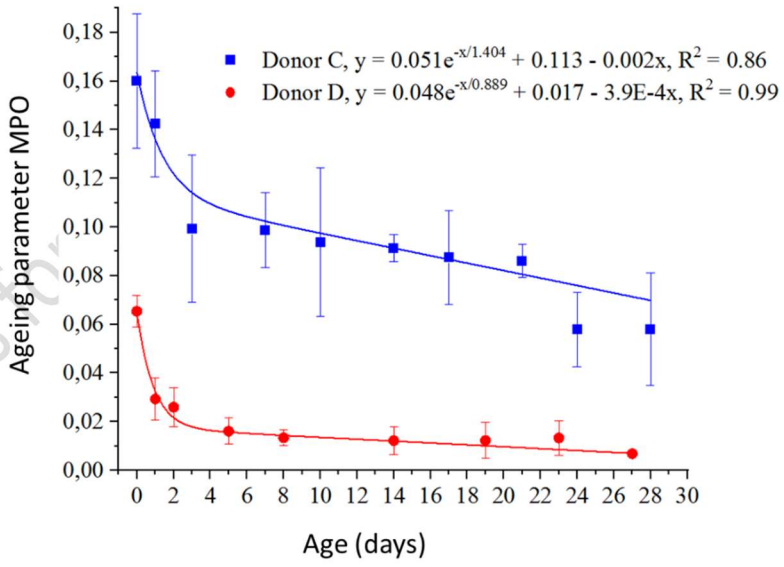


Figure 3 – Aging curves for two donors using myristyl palmitoleate (MPO) as the target compound. Fingermarks were deposited on paper and stored in the dark during 28 days before extraction using dicholoromethane and analysis using GC/MS [46]. The aging parameter is represented as a function of time and fitted using exponential linear regressions.

A second approach to follow fingermark aging focused on the use of multivariate models, thus combining the potential of several aging parameters in one model. Partial least squares regression (PLSR) was tested for this purpose, and it was possible to differentiate fresh fingermarks (t ≤ 3days) from older ones (t ≥ 10 days). However, the PLS models showed large spreading due to the variability among the fingermark samples of the same donor (intra-variability) even when using pre-processing techniques. This type of model may thus not be the best choice to infer about the age of fingermarks [46].

As described above, many factors influence fingermark composition and its aging. In an attempt to propose solutions to deal with this issue, factors were classified in two groups [46]. The known factors, such as the donor (if a suspect has been identified), the substrate (on which the fingermark has been detected) and the enhancement techniques (applied to the mark before analysis) can be determined by the forensic investigators, while the time of deposition, pressure and environmental conditions are more difficult to reconstruct. Known factors can be included in the aging models since it has been demonstrated that substrate and enhancement techniques also have significant influence on the initial quantity recovered and/or the aging processes [46]. The considered time range will depend on the hypotheses formulated during the investigation. Short time spans (e.g. up to 1 month) would be relatively easy to reconstruct during an investigation using the suspect's marks, however longer time ranges would require additional resources that may not be available even for serious crimes. Thus, research should endeavor to study fingermark aging for longer time periods, for example up to 1 year. For such longer periods, lipid components will probably be less relevant targets than amino acids or proteins, as their degradation is rather rapid. While pressure has been shown to influence fingermark length [35], it seems to have less influence on chemical degradation than storage conditions [46]. Light was found to accelerate the aging of lipids in particular, while small changes in temperature (15 - 25°C) had a more moderate impact [9,46,54,67,70]. Reconstructing the storage conditions (approximate temperature, humidity and light exposure) may be possible over short periods of time (using weather reports as done for entomological purposes). Naturally occurring indoor storage conditions will probably have a lower effect on fingermarks aging than outdoor environments. However, exposure to light should still be accounted for as it can significantly influence the degradation

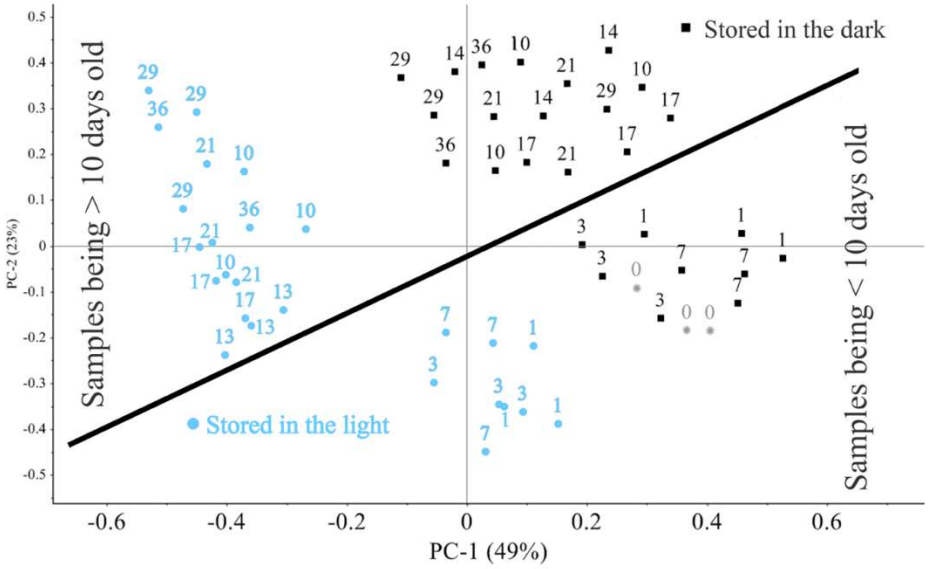
processes of some lipids, such as squalene and triglycerides [41,46,67,70]. The impact of substrate type, initial composition (e.g. groomed fingermarks) and exposure to light were also found to modify ridge pattern characteristics [19-21,23].

3.2.3. *Interpretation processes, operative testing and validation*

Several approaches to interpret the age of fingermarks can be developed depending on the information available to investigators (e.g. known vs. unknown suspect). From an investigative perspective, particularly if a suspect has not yet been identified, the objective is to situate the traces in time either to evaluate the relevance of the detected traces or to evaluate when the investigated event (or particular actions) occurred [58]. Due to the fact that in such cases the donor would not have been identified yet, the interpretation models would aim to determine an approximate timeframe during which the mark has been left, taking into account the circumstances and including if possible, the influence factors (e.g. substrate type, storage conditions). Up to date, no interpretation process including validation steps were published using physical characteristics as aging parameters. Concerning chemical characteristics, three different interpretation models were developed based on lipid target compounds and tested for fingermarks left on glass and stored in an office drawer or on an office desk near a window for up to 34 days (see Figure 4, 5 and 6 adapted from data acquired in [46]).

The first model was based on principal component analysis (PCA) combined with Soft Independent Modelling of Class Analogy (SIMCA) in order to predict to which pre-defined age groups a questioned mark belonged. Validation of this supervised classification method was conducted by an internal cross-validation study and then applied to specimens unknown to the principal investigator through blind testing: substrate and donor were known variables, while age and storage conditions remained unknown to the forensic analyst. Fingermarks were classified as being under or over 10 days old regardless of their storage conditions (dark vs. light). While the model showed very promising results, the storage conditions had a significant impact on the aging of older fingermarks (see Figure 2). Two false results were observed, i.e. two 22- and 34-day old fingermarks stored in a drawer were classified as being less than 10 days old, showing that aging is much slower when marks are not exposed to light. Moreover, a 34-day old

fingermark exposed to light aged to such an extent in the given timeframe that the model was unable to classify it. Thus, such approaches should take into account storage conditions and particular care should be taken when evaluating the age of potentially older marks, as some specific conditions can significantly slow or accelerate aging processes and yield erroneous classification. The practical feasibility of such models should also be further tested using fingermarks from unknown donors and using further apart aging periods, such as very fresh (e.g. less than 3 days) compared to older marks (e.g. more than 2 weeks, 1 month, 1 year). This could potentially increase the feasibility of the model in specific conditions (i.e. pre-defined timeframes).



Samples	Real age, storage	SIMCA
FM1	22d, light	N.A.
FM3	1d, light	< 10d
FM5	1d, dark	< 10d
FM7	22d, dark	< 10d

Figure 4– PCA model using 6 aging parameters and based on latent fingermarks deposed on microfiber filters and left to age in the dark between 0 and 36 days before extraction using dichloromethane and analysis using GC/MS [46]. Based on this model, fingermarks were classified in two groups: < 10 days old or > 10 days old (d = day). One PCA per observed group was built in order to classify blindest samples (e.g. FM1, FM3, FM5 and FM7) in one or the other group using SIMCA. The classifications in green were correct, while the ones in red were wrong.

The second model was based on univariate exponential linear regressions (see examples in Figure 5). While most studies do not explicitly propose interpretation models, they often present their results using univariate aging curves. The results obtained for 8 test marks using a 99% confidence interval yielded 5 correct and 3 incorrect estimations [46]. As the model was constructed independently of factors such as light and dark conditions, the errors were quite large, and a correct result could be quite imprecise. For example, one mark was correctly classified as being more than 3 days old but was actually 34 days old. The fresh and old marks' ages were generally correctly determined (even if sometimes imprecisely) using this model. The errors actually occurred for the ages that fell along the center of the regression curve. This may support the hypothesis that it is only possible to differentiate very fresh (less than 3-days old) from older (more than 30-days old), at least based on the rapid decrease of squalene over the first days after deposition. It would be interesting to test other compounds that are known to decrease more slowly for a finer age discrimination and/or estimation over longer time spans.

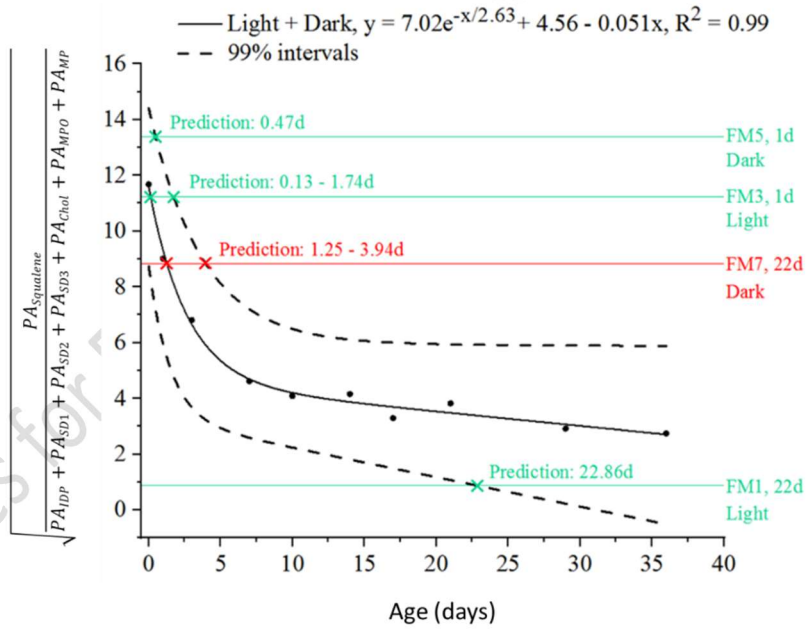


Figure 5 – Exponential linear regression using the aging parameter of squalene. Fingermarks deposed on microfiber filters and stored in the dark during 36 days before extraction using dichloromethane and analysis using GC/MS [46]. Based on this regression and using 99% confidence intervals, the ages of blindtest samples (e.g. FM1, FM3, FM5 and FM7) measured in days (d) were predicted. The predictions in green were deemed acceptable (actual age correctly predicted within one day), while the one in red was incorrect (more than 18 days off).

A multivariate PLS regression was also tested to predict the age of unknown specimens including a prediction error [46]. The robustness of the model was evaluated through the root mean square errors (RMSE) and the coefficients of determination (R^2). Mean RMSE were of *ca*. 8 days and obtained R^2 for calibration (0.58) and prediction (0.48) were actually quite low. Thus, predictions were not very precise, and 3 errors were produced with older fingermarks (22 and 34 days old) as the model could not accurately predict aging in both dark and light exposure conditions. Results varied more significantly over time, and thus prediction of older samples became more difficult if the storage conditions were unknown. These results tend to confirm that storage conditions should be predictable (in addition to substrate and donor) to accurately predict the age of fingermarks over *ca*. 1 month. Indeed, a PLSR model created specifically for fingermarks deposited on filter paper for one donor and stored in a drawer yielded lower RMSE (*ca*. 3 days) and higher R^2 (above 0.80). Such a model has also been tested on aluminum and analyzed using FTIR [48]. The regression models showed good linearity for both storage conditions (R^2> 0.90) and errors were relatively low (between 2 - 3 days). Storage conditions had less influence on the results in this study compared to previously obtained results [46]. Unlike GC/MS, FTIR spectra were obtained using continuous measurements from the same fingermarks over time as specimens were not destroyed by the analysis. The influence of initial composition was thus minimized when building the aging model and expected errors may actually be significantly higher in more realistic conditions.

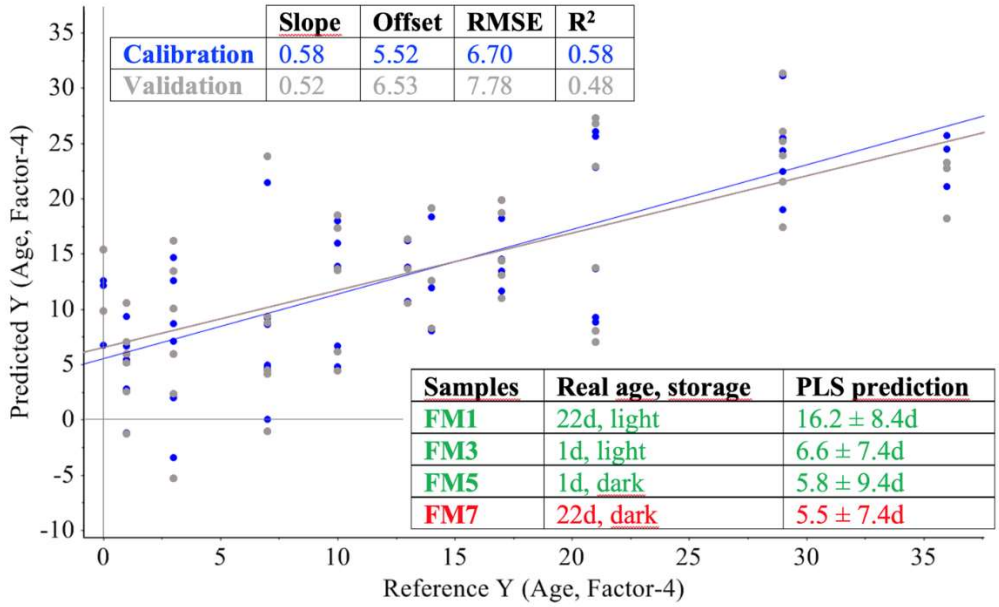


Figure 6– PLS regression using 6 aging parameters and based on fingermarks deposed on microfiber filters and stored in the dark between up to 36 days before being extraction using dichloromethane and analysis using GC/MS [46]. Based on this regression, the age of blind test samples (FM1, FM3, FM5 and FM7) in days (d) was predicted. The predictions in green were correct (however with large errors), while the one in red was wrong.

It is interesting to mention that a fourth type of interpretation model was proposed to allow the comparison of two alternative propositions for the age of a questioned mark, for example in a court context [46]. For this purpose, likelihood ratios (LR) were calculated using a preliminary Bayesian network. The selected alternative hypotheses for the purpose of illustration were the following: the questioned mark was deposited less than 10 days prior to analysis (i.e. age ≤ 10 days old) or it was deposited more than two weeks before (i.e. age ≥ 14 days old). Two age estimate errors were obtained using the same blind test set (mentioned above), again for older marks of 22 and 34 days stored in drawers. LR values were generally very low (below 4), except for two older marks exposed to light and for which LR values reached *ca*. 10 and 12. These results highlight, as previously mentioned, that models cannot accurately predict the age of older fingermarks if they do not include the storage conditions (i.e. dark/light exposure). However, when the

likelihood ratio approach was tested using separate models for the two storage conditions, it yielded correct estimations for all the tested specimens. Obtained LR values were still very low for dark conditions but reached high values for marks exposed to light. Investigating larger time/age differences would be interesting for further research, particularly for dark conditions under which aging occurred more slowly.

While research continues to gather further fundamental knowledge about fingermark composition and aging, the development and validation of interpretation models remain largely overlooked. In order to develop a reliable fingermark aging method fit for practice, it is essential that additional studies focus on this particular topic. If the validation step fails, the research cycle has to be started anew, focusing either on new target compounds, more reliable analytical methods or alternative aging and interpretation models. Based on the literature overview, it appears that fingermark dating methods cannot be implemented universally to all encountered cases using the same aging and interpretation models, but should instead be adapted to specific cases considering known substrate, donor, storage conditions and relevant time span. Even if the feasibility of fingermark dating is limited, acceptable reliability could be guaranteed through adequate testing of the statistical models (including the determination of error rates) and probabilistic approaches. It must be also noted that the majority of research on the composition and aging of fingermarks has been conducted using groomed samples. However, before being able to apply a method in forensic practice, its implementation should also be tested on natural residue, adding another level of complexity to the interpretation. While there are several proposals to control fingermark deposition and environmental conditions, as well as chemical composition for research purposes [30,74], scientists must be aware that the implementation of dating models in practice needs to be based on results obtained using realistic and uncontrolled specimens and include blind testing in casework conditions.

3.3. Practical implementation of dating models

A review of 28 court cases between 1961 and 2011, in which age estimation of fingermarks had been attempted, indicated that most cases were burglaries or robberies, attempts included (61%) followed by homicide (32%) [44]. The two remaining cases

concerned (sexual) assault and kidnapping. Fingermark age estimation was generally based on the quality of the enhanced friction ridge patterns and experience of the forensic expert, rather than experimentally-drived data. While 16 of these judgements were confirmed on appeal based on fingermark evidence and/or other pieces of information, 12 others were reversed on appeal.

It is interesting to note that in the 5 cases where the defense presented alternative explanations for the presence of the traces, the judgment was reversed [1-4] with the exception of a robbery case in which one suspect was released while the other was convicted [5]. One of these cases concerned an homicide [3]. A 72-year old man was found dead by his niece in the living room of his house. He had been restrained and shot in the head. The house had been ransacked and his watch and the money in his wallet were missing. The only evidence which connected the defendant to the murder was a partial fingermark discovered on a small metal box located in another room. The fingermark specialist rightly testified based on his experience that “the length of time a latent print would stay on a metal object like the box found on the desk in the den would vary depending upon conditions, but that a print could stay there for a period of weeks." The niece of the deceased testified that the defendant was unknown to her, that the box was usually kept in a filing cabinet and that it was only handled by family members. Based on these elements, the defendant was convicted. In the appeal it was stated that, as the fingermark could have been a few weeks old, “during that period the defendant could have either entered the porch unlawfully without the occupant's knowledge or lawfully to transact business with the deceased.” This alternative explanation about the presence of the fingermark prompted the judgment to be reversed.

In two burglaries, fingermarks were detected on broken glass fragments [2,4]. In both appeals, it was concluded that the windows were accessible to the public and the marks could have been deposited for legitimate reasons, such as looking through a shop window. In Townsley v. United States [4], the court also noted that touching a window might be a trivial incident and it would be normal that the defendant had no memory of it when arrested several months later. In Solis v. People [2], the forensic expert testified “that latent prints would remain for about two years, unless they were exposed to the elements”. As

no other evidence was offered by the prosecution, the appeal was accepted in both instances. The fingermark expert's observations regarding the persistence of traces were confirmed by other forensic scientists as reported in the literature [14].

The two last cases dealt with objects that could have been handled by many people in stores before having been purchased and subsequently seized at crime scenes (i.e. a map found in a car and an air freshener found in a bathroom, respectively [1,5]). In the latter, the court noted the importance of establishing the time of deposition of fingermarks to avoid jeopardizing "the liberty of every person who ever touches anything later found at the scene of a crime [1]." This ruling clearly emphasizes the importance of estimating the age of fingermarks (and other traces) detected on everyday objects found on crime scenes to demonstrate their relevance to the investigated event. While the question "**who?**" can be answered by dactyloscopic comparison and the location of a fingermark on a crime scene/object largely answer the question **"where?"**, the questions "**when?**" and to some extent "**how?**" remain mainly unanswered if the fingerprint residue cannot be situated in time. Indeed, an object could either have been handled in a shop, or at the crime scene weeks before the crime occurred, or during the investigated event, and through legitimate (e.g. selecting products during shopping) or illegal (e.g. conducting a search during a burglary) actions (how and for what purpose?).

There are, of course, several other ways to answer the questions mentioned above. Investigative information collected by the police (e.g. testimonies, circumstantial information) should not be ignored, as they might be more straightforward, and at the current stage of knowledge, even more reliable than the age estimation of fingermarks. As discussed in section 2, the implementation of dating methods could involve significant costs and resources and may only be justified in serious crimes such as homicides or sexual assaults. Furthermore, fingermark dating would currently mainly be feasible if samples were collected relatively quickly after the occurrence of the criminal event. Based on chemical degradation research, it would be feasible to differentiate fresh fingermarks from significantly older ones (e.g. a few hours/days vs. a few weeks/months) [46]. For example, a fingermark found on a box kept in a closed cabinet (known substrate, storage conditions and suspect, as in State v. Scott [3]) could probably be classified as having

been left a few hours before the crime, rather than several weeks before it occurred. The metallic box was kept in a closed cabinet and storage conditions could be inferred relatively easily. Using a combination of ridge pattern degradation parameters, a few weeks old fingermarks may be distinguishable from months old traces in certain conditions [21].

During the investigation, it should be noted why a particular object was examined for fingermarks (was the object displaced or used during the burglary?) and how many traces were found (including those of the legitimate users). If only a few fingermarks are recovered on a questioned object, it probably means that this object is cleaned regularly, and the maximum possible age of the collected fingermarks may be estimated. However, as previously mentioned, some objects can often be manipulated by different persons. This is why it is necessary to note the exact placement of the collected fingermark on the object, as its position could give information about the activity conducted when leaving the mark (legitimate or illegal). Such investigative information is very important to be able to estimate fingermark age. Moreover, in order to base these estimations on sound data, research should also focus on the persistence mechanisms of marks on different objects routinely used within our everyday activities. This could help answering questions such as "Is it possible that a fingermark can persist for several weeks on an object that has been bought by other people in a shop?". In the air freshener case example mentioned [1], if the questioned fingermark had been found on the spray button (typical position when using the spray, rather than by touching it in a shop), it would probably not persist for very long, particularly if the air freshener spray was being used regularly. However, experience has also shown that fingermarks of good quality can still be detected several years after deposition on glass windows and on the interior surfaces of apartments as reported in Solis v. People [2,14], while fingermarks on a drinking glass or a knife would probably not persist after washing the dishes. Knowledge of the persistence mechanisms of fingermarks is thus highly interesting in such contexts.

In summary, situating fingermarks in time could help identify relevant traces (investigation stage) and avoid the conviction of innocent people (court stage). Age estimation can be based on measurable chemical or physical characteristics over time and/or other relevant

information about the deposition and persistence processes. However, further research is needed, as suggested in previous sections following the cycle proposed in Figure 1, in order to develop reliable fingermark dating methodologies. In the meantime, while age estimation is not yet deemed feasible, it is important to consider fingermarks not only at the source level, but also at the activity level. This is why crime scene investigation is crucial. Precise information about the placement of marks should always be recorded to enable sound interpretation of the evidence, particularly when a suspect had legitimate reasons to visit the crime scene (e.g. he/she was living in the apartment several months before the crime occurred) or to handle objects later found at the crime scene (e.g. air freshener or metallic box). Thus, situating fingermarks in time should be carried out in a global investigative framework endeavoring to answer all relevant questions surrounding the criminal event (who, what, where, when, how, why, how often, etc.).

One also has to consider that in cases in which a partial mark of poor quality is encountered, the risk of erroneous identification is increased and age estimation cannot always provide a solution [14]. In the Brandon Mayfield case for example, the fingermark found at the crime scene in Madrid was actually relevant to the investigation as it led to the identification and arrest of a suspect, but it was not left by Brandon Mayfield as initially thought by several fingermark experts [78]. Thus, particularly when a fingermark is partial and/or of poor quality, extreme care should be taken in the interpretation of the data and probabilistic approaches should be preferred to categorical identification to avoid erroneous conclusions and decisions [32,60].

3.4. Conclusion and perspectives

Beyond the basic question “who?”, answers to other relevant queries such as “when?”, “where?” or “how” could help minimizing erroneous judicial decisions in criminal inquiries. This requires close collaboration and communication between police investigators, crime scene technicians and forensic scientists.

In this context, situating fingermarks in time would be particularly useful to select relevant traces at crime scenes and, subsequently, if the persons at the source of those traces are identified, to infer their presence (or actions) at the time of the investigated event. While forensic scientists have often attempted to estimate the "age" of fingermarks (i.e. their time of deposition on objects), a reliable dating method has yet to be reported. It is widely accepted that fingermark patterns, at least on porous surfaces, can persist for very long times when not exposed to extreme conditions (e.g. direct exposure to water, light or higher temperatures). Thus, visual observation of the quality of a fingermark conducted with the naked eye cannot be easily correlated to its age and more data is still needed to build and test reliable aging models based on the objective measurements of physical characteristics.

Physical and chemical changes occurring over time have frequently been proposed to estimate the age of fingermark residue. For example, lipid components are known to decrease in quantity over time through diverse degradation and diffusion processes. Squalene, a steroid precursor generally present in relatively high quantities in the initial fingermark residue, has received particular interest and its decrease after deposition was studied over several weeks. While several potential aging and interpretation models have been proposed (for squalene and other target compounds), many factors play a role in the aging processes and should be further studied to evaluate the practical feasibility of such approaches. It has been suggested that the person at the source of the questioned trace must be identified to reduce the high variability of the initial composition of inter-donor fingermarks. The ideal model should also strive to include substrate type and storage conditions to minimize the risk of making erroneous age estimations. Dating methodologies based on measuring changes in the chemical composition of fingermarks, while interesting and promising, are complex and may never be suitable to determine precise ages. Preliminary results indicate, however, that such approaches could be able to discriminate between fresh (i.e. a few days old) and older fingermarks (i.e. a few weeks old). Blind tests should be systematically carried out at the end of the validation process to evaluate the methods' feasibility and reliability. Furthermore, as the development and application of dating approaches for practical implementation demands large amounts of

resources, age estimation could primarily be applied in the investigation of serious crimes such as homicides, even if it would be useful for any type of crime.

Academic research on the composition and aging of fingermark residue also plays an important role in the development of dating methodologies. Future research should focus on the systematic acquisition of fundamental data following the cycle proposed in this chapter. At the current stage of knowledge, it is advised to promote in-depth studies of selected aging parameters and processes (including 2 and 3D changes), rather than preliminary proof-of-concept studies. In fact, new technologies might be less crucial in resolving the dating challenges than data processing and interpretation. With this objective in mind, much more information is currently needed on the aging of more *realistic* specimens (i.e. natural fingermarks stored under different conditions) in order to build reliable aging and interpretation models.

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