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Aging of target lipid parameters in fingermark residue using GC/MS: Effects of influence factors and perspectives for dating purposes

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ABSTRACT

Despite the recurrence of fingermark dating issues and the research conducted on fingermark composition and aging, no dating methodology has yet been developed and validated. In order to further evaluate the possibility of developing dating methodologies based on the fingermark composition, this research proposed an in-depth study of the aging of target lipid parameters found in fingermark residue and exposed to different influence factors. The selected analytical technique was gas chromatography coupled with mass spectrometry (GC/MS). The effects of donor, substrate and enhancement techniques on the selected parameters were firstly evaluated. These factors were called *known* factors, as their value could be obtained in real caseworks. Using principal component analysis (PCA) and univariate exponential regression, this study highlighted the fact that the effects of these factors were larger than the aging effects, thus preventing the observation of relevant aging patterns. From a fingermark dating perspective, the specific value of these *known* factors should thus be included in aging models newly built for each case. Then, the effects of deposition moment, pressure, temperature and lighting were also evaluated. These factors were called *unknown* factors, as their specific value would never be precisely obtained in caseworks. Aging models should thus be particularly robust to their effects and for this reason, different chemometric tools were tested: PCA, univariate exponential regression and partial least square regression (PLSR). While the first two models allowed observing interesting aging patterns regardless of the value of the applied influence factors, PLSR gave poorer results, as large deviations were obtained. Finally, in order to evaluate the potential of such modelling in realistic situations, blind analyses were carried out on eight test fingermarks. The age of five of them was correctly estimated using soft independent modelling of class analogy analysis (SIMCA) based on PCA classes, univariate exponential linear regression and PLSR. Furthermore, a probabilistic approach using the calculation of likelihood ratios (LR) through the construction of a Bayesian network was also tested. While the age of all test fingermarks were correctly evaluated when the storage conditions were known, the results were not significant when these conditions were unknown. Thus, this model clearly highlighted the impact of storage conditions on correct age evaluation.

This research showed that reproducible aging modelling could be obtained based on fingermark residue exposed to influence factors, as well as promising age estimations. However, the proposed models are still not applicable in practice. Further studies should be conducted concerning the impact of influence factors (in particular, storage conditions) in order to precisely evaluate in which conditions significant evaluations could be obtained. Furthermore, these models should be properly validated before any application in real caseworks could be envisaged.

1. Introduction

Fingermarks have been commonly used in forensic investigations for more than a century, essentially for identification purposes [1–4]. However, other useful information could be extracted from fingermarks. For example, the estimation of their age, i.e. the time of their transfer, can be crucial for the resolution of cases. Indeed, this information is directly linked to the relevance of these marks, as it allows determination of whether they were left during the crime or not [5,6]. It is not uncommon that an identified suspect admits to having been at a crime scene, but claims that the contact occurred at a time different to that of the crime and for legitimate reasons. Despite the recurrence of such issues and the research conducted on this topic, no fingermark dating methodology has yet been validated by the entire forensic community [7–9].

Among the fingermark dating methodologies proposed so far, the study of the aging of chemical characteristics seems to offer good perspectives because numerous target compounds can be studied using various analytical techniques, as already foreseen by Olsen [10]. Later, this opinion was also supported by Wertheim, who saw in the analysis of fingermark constituents the opportunity to find compounds deteriorating over time without being too much affected by influence factors,

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which would allow for accurate age estimations [11]. This idea was also mentioned in different reviews on the subject [12–14]. Studies reported that lipids were interesting targets for aging research, as they can be analysed by techniques commonly available in forensic laboratories (e.g., GC/MS or FTIR) and undergo oxidation over time [7–9,12,15–17]. Furthermore, reproducible aging patterns following an exponential decay could be detected, particularly for fatty acids, cholesterol and squalene extracted from fingermarks stored under controlled conditions and analysed using GC/MS [16,17]. However, it was also highlighted that the initial composition and aging of lipids was significantly affected by different influence factors, classified in five groups: the donor, the substrate, the enhancement techniques, the deposition and the storage conditions [12]. Some research using GC/MS were already conducted to study the impact of the donor [17–20], the substrate [17], the application of enhancement technique [19] and the exposition to light [16] on the initial composition and aging of some lipids (particularly fatty acids, squalene and cholesterol). Overall, large variability was observed. The calculation of ratios of compounds using squalene, cholesterol and wax esters was proposed to reduce part of the variability due to donors, substrate and enhancement techniques [17,19]. However, the efficiency of these ratios still needs to be further tested on larger sets of fingermarks exposed to various influence factors. Based on recent papers proposing a formal practical fingermark dating framework [8,9], the five groups of influence factors mentioned above can be further categorised as *known* or *unknown* factors. In fact, as it seems reasonable to consider that fingermark dating questions would mostly be raised after the collection, enhancement and identification of the questioned marks, donor, substrate and enhancement techniques can be considered as *known* influence factors. Thus, specific aging models could be built for each new case taking these particular parameters into account, i.e. using comparison fingermarks collected from the identified donor, on the same substrate and applying the same enhancement techniques to the comparison and questioned marks. The variability brought by these three sources could thus be limited. On the contrary, deposition and storage conditions can generally be considered as *unknown* factors, as they will never be precisely obtained in caseworks. For fingermark dating perspective, it is therefore particularly important to study the effects of these factors on target compounds (e.g., lipids) and take them into account in aging models.

The aim of the present research was to study the aging of target lipid parameters available in fingermark residue and exposed to different influence factors using GC/MS. Whether the residue variability reported in the literature could be reduced in such a way that robust aging models could be developed and used for fingermark dating perspectives was assessed. First, the effects of *known* factors on selected lipid parameters were evaluated over time using principal component analysis (PCA) and univariate regression, in order to collect fundamental knowledge about the variability brought by such factors. Four donors, two substrates (microfiber filters and glass) and one enhancement technique (aluminium fingermark powder, also called *Argentorat*) were tested. Then, the effects of *unknown* factors on lipid parameters were also studied over time with PCA and univariate regression. Partial least square regression (PLSR) was also used in order to evaluate if multivariate regression was more appropriate to model aging affected by these factors. Two different deposition moments (three months apart) and three different deposition pressures (100 g, 500 g, 1000 g) were tested, as well as three different storage temperatures (15, 20 and 25 °C) and two different lighting conditions (darkness and light). Furthermore, in order to evaluate the real potential of such modelling for fingermark dating purposes, blind analyses of eight test fingermarks were carried out and their age was estimated using the relevant PCA, univariate regression and PLSR models. Finally, based on literature on the dating of ink traces [21,22], gunshot residue [23] and fingermarks [8,9], a probabilistic approach using the calculation of likelihood ratios (LR) obtained from a Bayesian network was also tested to evaluate the age of these eight test fingermarks.

2. Material and methods

2.1. Sampling: donors, deposition protocol and data sets

Four different donors were selected for this study, based on their availability during the different research phases. All donors were Caucasian, non-smoker and omnivores who had no diagnosed metabolic disease. Furthermore, these donors were classified based on a recent study proposing a donor classification based on their fingermark lipid composition [20] (Table 1).

All the fingermarks collected for this study were deposited on 25 mm diameter glass microfiber filters (Whatman, Bottmingen, Switzerland) or on glass microscope slides (VWR international SA, China) according to the following deposition protocol:

1. The donors were asked to follow their tasks normally before deposition. The only condition was to avoid hand washing with soap within the last 45 min preceding the deposition.
2. Both thumbs were gently rubbed on the forehead, miming a natural movement.
3. The time of deposition was maintained at 15 s and each fingermark was deposited on a kitchen scale to obtain the desired pressure (variations of ±50 g were observed). During each deposition session, the right and left thumbs were collected.

After deposition, some fingermarks were directly analysed (t = 5–10 min) and others were aged under different storage conditions up to around one month before analysis. For each age, two fingermark specimens were analysed. As mentioned in the introduction, seven influence factors were studied in this work (i.e. donor, substrate, enhancement, deposition moment, deposition pressure, temperature and lighting) and blind tests were also conducted. Table 2 summarised the acquired data sets.

When one influence factor was tested, the others were fixed to the following standard values (unless other precisions are given under *Description* in Table 2):

- Substrate: paper
- Enhancement: none
- Deposition moment: fingermarks used in a specific data set were left within one month
- Pressure: 500 g
- Temperature: 20 °C (climate chamber)
- Lighting: complete darkness.

It is important to note that the effects of enhancement techniques (set #3) were tested using the *Argentorat* powder (Paine Spurensicherung, Arni, CH) because this technique is one of the most widely used on non-porous surfaces such as glass [4]. The application of the powder was made when the fingermarks reached the desired age and analysis was then conducted around 30 min after enhancement. Concerning the lighting conditions (sets #3 and 7), the fingermarks stored in the dark were put in closed boxes kept in an air-conditioned laboratory. The fingermarks exposed to light were stored in an open box in the same laboratory close to the window, being thus exposed to light during the day and to darkness during the night (normal 24 h cycle). Finally, it should be highlighted that, as the analyses were

Table 1
Characteristics of the fingermark donors.

Name	Donor classification	Sex	Age	Cosmetics
A	Medium-rich	F	26	No
B	Medium-rich	M	27	Face cream
C	Medium-poor	F	27	Face cream
D	Poor	F	32	No