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Differentiating individuals through the chemical composition of their fingermarks

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ABSTRACT

Fingermark patterns are one of the oldest means of biometric identification. During this last decade, the molecules that constitute the fingermark residue have gained interest among the forensic research community to gain additional intelligence regarding its donor profile including its gender, age, lifestyle or even its pathological state. In this work, the molecular composition of fingermarks have been studied to monitor the variability between donors and to explore its capacity to differentiate individuals using supervised multi-class classification models. Over one year, fingermarks from thirteen donors have been analysed by Matrix-Assisted Laser Desorption/Ionisation Mass Spectrometry Imaging (n = 716) and mined by different machine learning approaches. We demonstrate the potential of the fingermark chemical composition to help differentiating individuals with an accuracy between 80% and 96% depending on the period of sample collection for each donor and size of the pool of donors. It would be premature at this stage to transpose the results of this research to real cases, however the conclusions of this study can provide a better understanding of the variations of the chemical composition of the fingermark residue in between individuals over long periods and help clarifying the notion of donorship.

1. Introduction

Fingermarks are one of the most commonly used forensic traces bearing biometric information. Their exploitation requires the visualization of the ridge pattern through optical means, combined with the application of physico-chemical techniques [1]. Since 2009, the interest for the additional knowledge that can be extracted from a fingermark, beside the ridge pattern, has significantly increased [2].

The molecular content of the fingermark residue has proven to be a useful source of information to meet both profiling and ridge pattern reconstruction goals [3–6]. To reach these objectives, spectroscopic and spectrometric techniques have been applied to the detection of various (semi-)exogenous molecules and contaminants (e.g., explosives, drugs, pharmaceuticals, condom lubricants, blood) as well as endogenous molecules naturally produced by the body and excreted through sweat (e.g., triglycerides, amino acids, free fatty acids, peptides and proteins) [3,4,7–17].

Among these methods, Matrix-Assisted Laser Desorption/Ionisation Mass Spectrometry Imaging (MALDI-MSI) appears to be one of the most promising ones [3,18]. MALDI-MSI was already successfully applied on various types of substrates and put in sequence with other fingermark detection techniques to either reconstruct the ridge pattern [2,11,14,19–30] or obtain information about the molecular composition of fingermarks [3,15,17,19,31]. Also classified as a class B by the Home Office in fingermark recovery [32], MALDI-MSI is becoming, over the years, an asset for the chemical analysis of fingermark residue. As stated above, this technology offers many possibilities regarding the profiling of individuals, as well as their differentiation [31].

The introduction was completed with the following paragraph: "The idea of profiling donors based on the composition of their fingermarks has already been explored for purposes like sex determination, age estimation or health condition documentation [4, 15–17, 33, 34]. All these scientific developments have shown promising results. For example, the analysis of fingertip smears has recently been successfully applied to the classification of donors according to their breast cancer development stage [34]. Another proof-of-concept study also demonstrated the underlying potential of fingermark composition to determine the sex of an individual with accuracy level ranging from 67.5% to 74.4% [17]. All these

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studies were focused on specific compounds and also raised more fundamental questions regarding the degrees of variability and consistency of the fingermark composition over time and between donors. In this context, a lack of data has been identified with regards to the characterization of donors with regards to their donorship, a characteristic often used during the experimental design step. In 2014, Girod et al. investigated how the lipid composition of fingermarks could help categorizing donors between "rich" and "poor" ones [35]. For forensic research purposes, donorship is a crucial parameter that must be set in the early stages of the research. Indeed, it is often asked to select a range of donors based on their capacity to provide fingermarks reacting more or less with a specific detection technique and classify them according to their donorship. This selection is often performed in an empirical manner, asking several people to deposit fingermarks that are then processed with the investigated technique, or through habits, such as "this person is usually a good donor". Pre-profiling donors from the composition of their fingermarks could help selecting donors with various donorship profiles, or narrowing down the number of donors required. Gaining knowledge about the intervariability of the fingermark composition could also be useful to help refining the concept of donorship, and its stability over time, but also to better assess the potential of the fingermark composition to differentiate individuals among others.

This study aims at investigating the molecular composition of the fingermarks provided by 13 donors of both genders and varying ages, over a year, using MALDI-MSI and supervised multi-class classification models. The presented dataset was already used in a previous work to assess the intravariability of the 13 donors [36] and is now used in R environment [37,38], which is a virtual space that contains all the objects used (i.e., datasets, variables, functions, etc.), to explore the intervariability in-between the 13 donors and to determine the most performing model to differentiate individuals. It is believed that this research will provide new and relevant information about fingermark composition over a long period of time, as well as about the possibility to limit the pool of individuals at the source of a given fingermark using molecular information.

2. Materials and methods

The whole materials, methodology and instrumentation including MALDI-MSI instrument and parameters, matrix preparation and deposition as well the fingermark sampling process are similar and can be consulted in a previously published paper, available in open-source [36]. In brief, all the analyses were conducted on a hybrid mass spectrometer MALDI Linear Trap Quadrupole (LTQ) Orbitrap XL from Thermo Fisher Scientific with a linear trap and coupled to an Orbitrap, equipped with an azote laser of 337 nm. The mass range was set from 100 to 2000 *m/z*, the spatial resolution to 100 µm and the spectral resolution to 60'000. The instrument was used in imaging mode and acquired square areas of several mm2 corresponding to 150–200 pixels. α-CHCA was chosen as the matrix as it has already demonstrated its performance on fingermark analysis and was prepared with 5 mg/mL of α-CHCA in 70:30 ACN:H2O and 0.1% TFA. It was deposited on the fingermarks using an automatic sprayer from SunChrom as follows: first layer at 10 µL/min, second at 20 µL/min, third at 30 µL/min, and from the fourth to the tenth at 40 µL/min with a spray height of 25.32 mm.

A total of 22–30 fingermarks, depending on donor availability due to COVID-19-related restrictions, were collected per individual on glass slides over January to December 2020. After each collection, the fingermarks were aged for 24 h before being analyzed by MALDI-MSI. Two analyses (replicates) were performed on each fingermark, for a total of 44–60 analyses per donor and 716 analyses overall. In this study, only one replicate was used to avoid overfitting the models. The second replicate was considered as a back-up.

Regarding the data preprocessing, the exportation and pretreatments of the data are also identical to Gorka et al., (https://github.com/mgrk-94/ShinyApp_MALDI) [36]. In this research, the chemometric analysis were performed using median normalization followed by logarithm base 10 transformation. For predicting the class of the analysed fingermarks, several supervised classification methods were tested: linear support vector machine (SVMLINEAR), radial support vector machine (SVMRADIAL), random forest (RF), logistic model tree (LMT) and linear discriminant analysis (LDA). These models were encoded through R with the caret package [37]. Correlated variables were deleted with a cut-off of 0.9 [38,39]. The data were then split with a proportion of 65% for training and 35% for testing. Given that the sample size remains small compared to other types of data, the choice was made to use a repeated 10-fold cross validation based on the recommendations of Kuhn et al., 2016 [38] for the "traincontrol" function in R.

3. Results

3.1. Peaks detected and consistency

Subsequent to the analyses of all the fingermarks from the 13 donors, statistics have been carried out to provide insights about the number of *m/z* that are qualitatively consistent between all the fingermarks collected, or a subset of them, for two time periods: one month and one year (Table 1).

As emphasized in Tables 1, 14.3% of the *m/z* detected (i.e. 50 out of 349) are present – on a qualitative aspect – in all the fingermarks that were deposited over one month and 10.5% (36 out of 344) over one year. Most likely, these compounds are part of the endogenous substances common to humans, without encompassing any semi-exogenous substances that are influenced by the health and lifestyle of the individuals. However, 100% of appearance is too a restrictive value to rely on since it does not allow an *m/z* not to be detected once in all the fingermarks. Indeed, for various reasons, a specific *m/z* might not be detected (e.g., intensity below the limit of detection of the instrument, inhomogeneity of repartition on the fingertip surface, internal instrumentation issue). Therefore, taking into consideration the compounds present in 90% of the collected fingermarks, the percentages of *m/z* constantly detected during one month and one year raise to 38.7% (135 out of 349) and 29.1% (100 over 344), respectively. Qualitatively, it is reasonable to assume that mainly (semi-)endogenous substances, produced naturally by the human metabolism, are included in this selection.

Interestingly on another hand, the percentage of *m/z* present in 50% of the fingermarks is almost identical to the after peak-picking selection. This observation is partly explained by the fact that the selected pool of donors all live in the same societal and geographical environment and therefore share many of the same daily habits (e.g., work environment, food, frequented shops, colleagues, etc…). The observed differences stem from their individual behaviour. However, these differences are disparately distributed among the 13 donors (as shown by the 75% and 90% values), which opens the door to the possibility of distinguishing individuals by the investigating the composition of their fingermarks.

Table 1
Number of *m/z consistently* detected in all (100%) or a subset (other % values) of the fingermarks collected during one month and one year. The reference 100% is based by default on the number of *m/z* remaining after the peak-picking process.

	After peak-picking	50%	75%	90%	100%
1st month	349	342	252	135	50
1 year	344	329	169	100	36

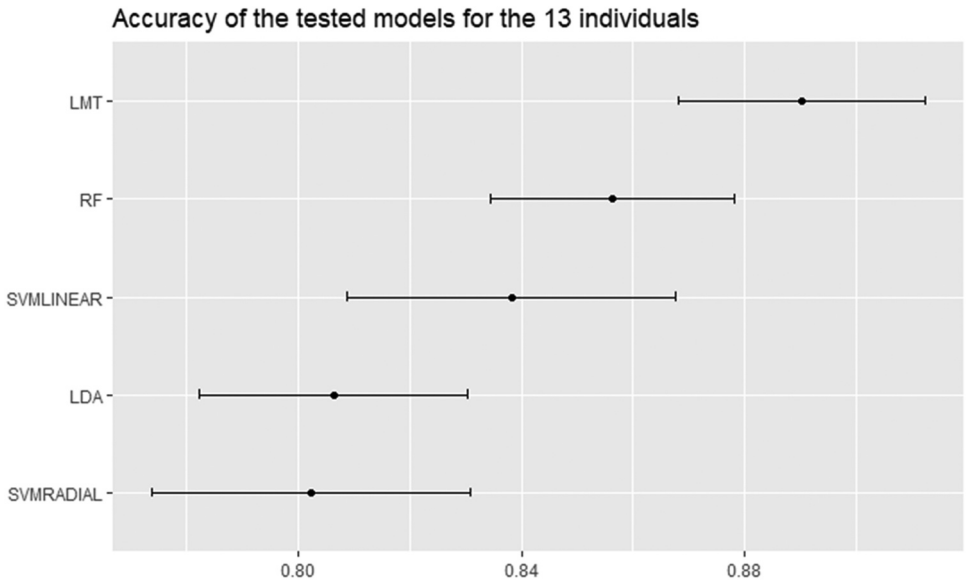

Fig. 1. Accuracy of the five supervised classification models that were considered in this study. Each dark spot represents the average percentage of correct attribution of a given fingermark to its actual donor. The lines on the edges stand for the 0.95 confidence level. Accuracy is a metric that informs on the fraction of correct predictions associated with the investigated model.

3.2. Multi-class classification of fingermarks

3.2.1. Differentiation of the donors

To investigate the possibility to use the chemical composition of fingermarks to differentiate individuals, five supervised multi-class classification models were selected and applied to the 358 fingermarks collected during this study. The objective was to evaluate the percentage of correct classifications that could be achieved by the five models and hence help selecting the most efficient one. The accuracies of the five models after training and resampling are illustrated in Fig. 1.

It can be observed that logistic model tree (LMT) has the best overall performance with more than 90% of correct classification, which means that for 90% of the considered samples, a given fingermark is correctly associated to its actual donor, solely through its molecular composition. Random Forest (RF), another model based on decision trees, is close in terms of performance with 85% of correct attributions. Lower performances were observed with the other models, with accuracy values oscillating between 80% and 85%.

To validate these results, receiver operating characteristic curves (ROC) were generated for the two most performant models (i.e. LMT and RF) and are presented in Fig. 2.

The micro AUC metric showed in Fig. 2 is useful in multi-class classification problems as it can highlight class imbalances to the contrary of the macro AUC which considers all classes equally. Overall, the closer the AUC is to 1, the more efficient is the model.

Fig. 2 depicts a micro AUC of 0.993 for LMT and 0.979 for RF. This means that in 99.3% (LMT) and 97.9% (RF) of the cases, a given fingermark is correctly assigned to its originated donor, and that the capacity of the model to distinguish between donors is almost at its best (100%).

Both models are powerful, but LMT remains superior, confirming the performances in terms of accuracy illustrated in Fig. 1. Moreover, these results emphasize that the modifications of composition over time, for a given donor, encompass some consistency. Indeed, if inconsistent variations of compositions occurred throughout the year for a given donor, subsequent misclassifications would have been expected, impacting both the accuracy and AUC values. These results are also in line with those obtained when monitoring the evolution of the fingermark composition throughout the year [36].

In both models, fingermarks of donors 8 and 9 account for the majority of the false positives or misclassifications. These results suggest that their composition – quantitatively speaking – may be quite similar.

Considering the LMT model, the five most discriminant variables (*m/z*) have been selected and plotted in a stripchart (Fig. 3) displaying their median and their associated interquartile range. This representation allows to visualise the quantitative differences between the 13 donors along one year.

Fig. 3 emphasizes the quantitative differences between donors for five given variables (*m/z*) over a one-year collection timeframe. These results highlight that the chemical composition of the fingermark residue quantitatively varies in between the 13 donors investigated and that this information can be useful to differentiate individuals.

Indeed, some *m/z* (e.g., *m/z 369.4284*) appear to be highly variable from one individual to another, while others vary only for a small part of the donors (e.g., *m/z 294.2407*). Even though two donors can be quantitatively close for some variables (e.g., *m/z 369.4284, 312.3626, and 294.2407* for IND7 and IND8), they may also significantly differ through other *m/z* (e.g., *m/z 283.2632 and 654.0686* for the same individuals) which allows to differentiate them successfully. For instance:

- IND1 and IND6 are well distinguished by all of the 5 *m/z*.
- IND7 and IND8 are close for *m/z* 369.4284, *m/z* 312.3626, and *m/z* 294.2407 but are clearly different for *m/z* 283.2632 and *m/z* 654.0686.
- IND8 and IND9 – as emphasized in Fig. 2 – are always extremely close for the 5 displayed *m/z*, which makes their differentiation difficult and explains the classification errors observed previously.

Overall, these results highlight that the chemical composition of fingermarks can be a powerful tool to differentiate up to 13 donors when using an adapted classification model.

3.2.2. Differentiation of a restricted pool of donors

In casework, one of the most important objectives is to narrow down the pool of suspects. In this context, the same supervised

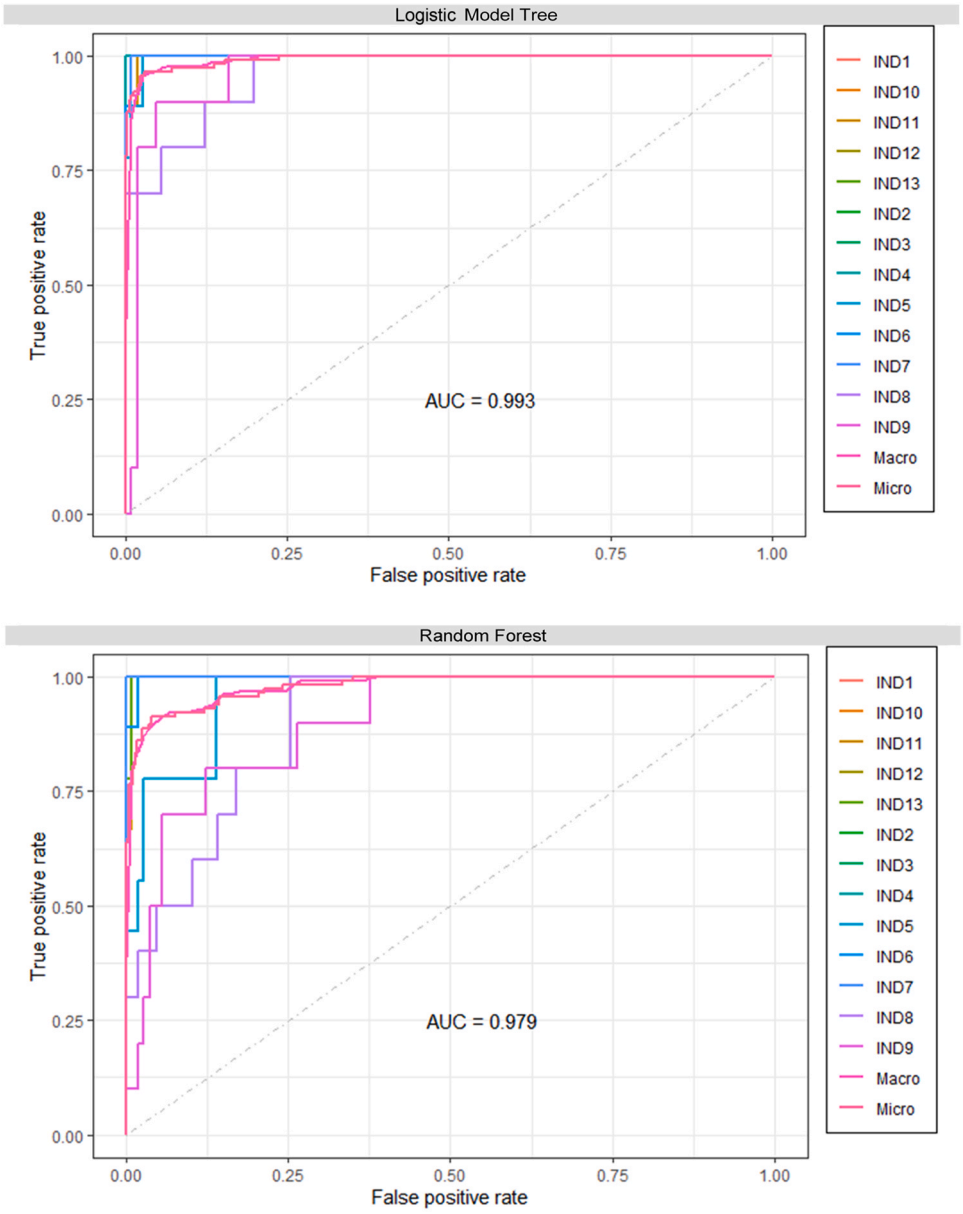

Fig. 2. ROC curves of LMT (upper image) and RF (lower image) models and their macro and micro area under the curve (AUC) value. Macro AUC computes the contributions of each class independently and then takes the average. Micro AUC aggregates the contributions of all classes to compute the average metric.

multi-class classification models were applied on 6 restricted random groups of 6 donors (arbitrary value). This way of doing aims at determining if the performances of the models are influenced by the individuals composing a pool of donors.

For a one year timeframe, the accuracy values of the 6 groups for the LMT model are illustrated in Table 2. The graphics for the 6 groups and the 5 tested models are available in supplementary data.

The results presented in Table 2 highlight the performance of the LMT model over the other ones, with an accuracy varying between 88% and 96% depending on the pool of donors selected. Also, five groups out of the six considered are associated with an average accuracy higher than 90%. These numbers also depict the influence of the donors contained in a pool on the classification results. Indeed, when randomising the donors, the percentage of accuracy varies slightly from one group to another. This emphasizes that some individuals tend to be closer in terms of fingermark chemical composition than others as it was already shown in Fig. 3. Indeed, for example, IND8 and IND9, whom were proved to be quantitatively close in terms of composition, were both part of the random group #2, which have the lowest accuracy. It can also be noted that this closeness is not specifically attributed to the sex of the donors neither their age (results not shown).

Regarding the ROC curves for the LMT model, the micro AUC values are close to one (0.987) for both random groups. These results are similar for all the four other random pools of donors that were generated (data not shown).

Overall these results emphasize that with a reduced number of individuals in a group, the false positive rate remains extremely low, which is of first importance when it comes to forensic evidence. Indeed, higher performance was expected for smaller groups of donors rather than with the original 13 donors' batch due to the greater chance of having different individuals with a restricted pool. Moreover, reducing the number of donors highlighted that some donors can be closer to others in terms of fingermark composition (e.g., IND8 and IND9), therefore influencing the accuracy of the models. However, the global accuracy remains extremely high for all the six groups with an average accuracy for the LMT model of 92.25%.

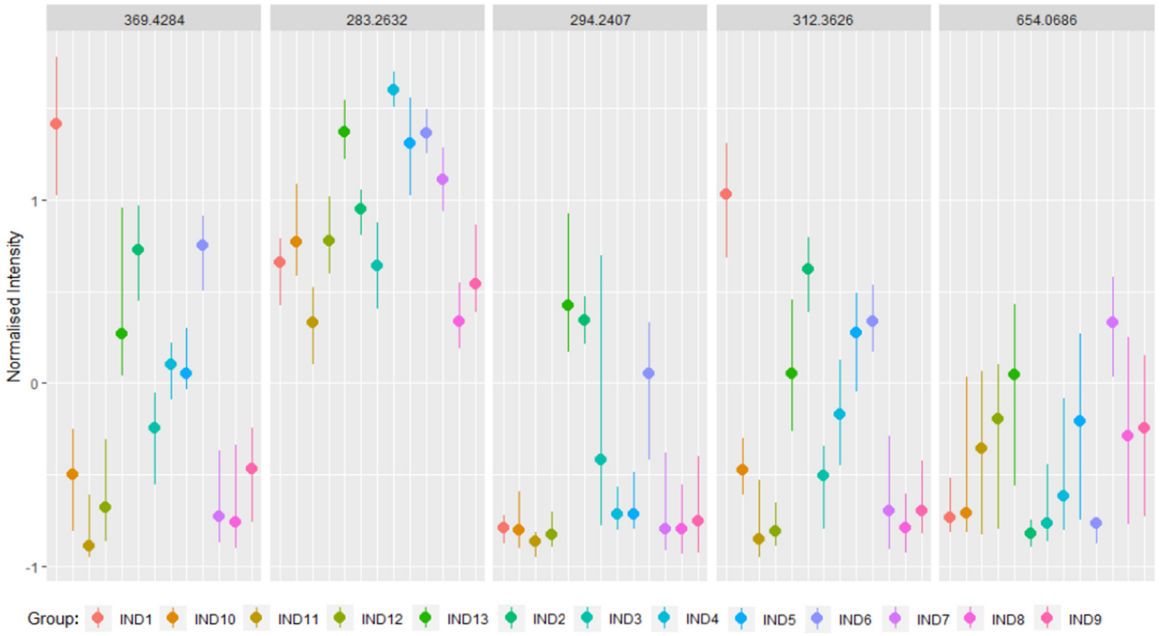

Fig. 3. Stripcharts of the 5 most discriminant variables (*m/z*) according to the LMT model. Dots represent the median of the variables and the lines the interquartile range (IQR). Each color stands for a specific individual (INDx).

Table 2
Accuracy of the LMT model for the 6 random groups, expressed in percentage. All the fingermarks collected over one year were considered.

Random group	#1	#2	#3	#4	#5	#6
LMT Accuracy	95.5	87.7	94.2	92.7	91.8	91.6

For a one month timeframe, the models were also tested for the same six pools of six donors (Supplementary data). Due to the restricted time-frame, only six fingermarks per donor were used to generate these results. Subsequently, the models are less trained and the confidence intervals tend to be wider. To avoid the influence of the size of the pool of data, the number of fingermarks collected in a timeframe of one month should be equal to the ones collected over one year. Nevertheless, due to the fingermark collection calendar for the 13 donors of this research, it was impossible to meet this condition.

However, the results still confirm that LMT remains the most powerful model for fingermarks classification of this dataset even though the average percentage of correct classification tends to be on average 5–8% lower, compared to the one obtained over one year (Fig. 3). These results might seem counter-intuitive with regards to the variation of composition. Indeed, the percentage of correct classification would have been expected to be lower over one year than over one month. However it is believed that the reason for such trends lies rather in the size of the pool of data than in the actual influence of the composition.

The ROC curves (data not shown) are also less defined but confirm that the LMT model offers a low false positive rate with micro AUC higher than 0.93.

These results are encouraging and confirm that the composition of fingermarks may help differentiating individuals when using the appropriate multi-class classification model. The reported results could be of an interest in research or, ultimately, for casework purposes.

4. Discussion

This study aimed to explore the possibility of distinguishing individuals through the chemical composition of their fingermarks, considering a large period of collection time (i.e. one year). Keeping in mind the methodological limitations, the obtained results showed the potential of using the composition of the fingermark residue to associate a given fingermark with its actual donor over a timeframe up to one year.

Indeed, we highlighted that the chemical composition of the fingermark residue varies qualitatively and quantitatively in-between donors, while keeping some inner consistency. When combining the chemical composition of fingermarks with an LMT classification algorithm, it was possible to associate 24-hours aged fingermarks with their actual donors, with an accuracy of up to 96%. Without undermining the observations that were made above, it is crucial to discuss the limitations of the methodology, to get a full picture of the frame in which this study places itself.

Indeed, if such study brings fundamental knowledge about the composition of fingermarks, it is also worth eventually considering a transition to the operational field, in which the composition of fingermarks would be used to prioritise suspects to investigate.

In this context, as this study only considered 24-hours aged fingermarks left on glass, it is first mandatory to evaluate the performance of this approach on fingermarks of varying ages to in order to realistically determine the level of applicability of this technique on aged fingermarks. In a second step, more research needs to be conducted on the type of substrate on which the fingermarks are deposited. It would allow assessing whether this detection method is applicable to porous substrates, only, or to other types of substrate as well. Finally, sequencing with other detection techniques should be evaluated to investigate the impact of chemical and non-chemical detection methods on the composition of fingermarks prior to MALDI analysis. Such results would determine whether or not this detection technique can be inserted in a fingermark detection workflow. Overall, these three additional research steps would help consolidating the results presented in this study and bring MALDI-MSI closer to a reliable operational implementation.

Indeed, when it comes to the operational implementation, it is mandatory to prove that the composition of fingermarks helps providing relevant and robust information to the investigators. Although this study is preliminary and further research is required, as stated above, the following elements of discussion are worth to be

raised. Two topics are to be considered: the fingermarks retrieved on site, and the reference fingerprints taken from a suspect. Additionally, questions related to the MALDI-MSI analysis feasibility are relevant to discuss.

First, as mentioned above, if the proof of concept is demonstrated, the methodology presented in this study could be applied in an investigative forensic strategy to prioritise the suspect to investigate and is implementable in laboratory equipped with a MALDI MSI instrumentation (e.g., legal medicine laboratories) which could lead to new inter-laboratory collaborations. For now, this detection process can only be applied on latent, unprocessed fingermarks or after a touch-DNA sampling if the whole fingermark residue was not swabbed. Indeed, DNA recovery and profiling remain possible after the application of MALDI MSI as the matrix applied in the sample preparation protects the biomolecules and may still allow a successful recovery and profiling of DNA [6]. When it comes to on-site fingermark collection, several studies have already demonstrated that the matrix powder can be used as a detection powder, which already offers an alternative to the traditional detection powders used on crime scenes [11,26,28] allowing the subsequent analysis of the collected fingermarks with MALDI. These findings corroborate that MALDI fingermark analysis could be implemented in forensic laboratories in the near future.

Regarding the collection of the reference fingerprints: when suspects are apprehended, their fingerprints are usually automatically recorded. Therefore, if required, it seems plausible that a suspect is asked to deposit a set of fingermarks on glass slides using the same protocol presented in this study, making the collection of references easy to implement. With further research assessing the impact of other detection methods on the fingermark residue composition and corroborating the obtained results of this study, a so-called "molecular signature" could be extracted from any individual using the same reproducible method that was implemented in this study.

The simplicity of extraction of the .imzML files with the dedicated R Shiny App makes such a concept handy to implement. In this regard, concerning the technical aspect of MALDI-MSI technology and data treatment, several aspects need to be considered. First, the time of analysis of fingermarks by MALDI depends on the required resolution, on the size of the analysed area, and if a visual reconstruction of the ridge pattern is required. For an area of a few mm^2 corresponding to the centre of a fingermark, the analysis takes a few minutes, but no exploitable image can be extracted. Bigger area, such as a whole fingermark, requires one to several hours with the instrument used in this study. But, with the rapid evolution of the MALDI technique, the latest instruments now allow to reduce the analysis time to a few minutes even at high resolutions. In this study, only a square of several mm^2 was considered, and up to 7 fingermarks could be analysed per day. However, the size of analysis was too small to retrieve the ridge pattern. Therefore, choices about size and resolution parameters that will impact the time of analysis have to be made depending on the needs for the investigation: imaging, profiling, or both.

Second, from a practical point of view, the .imzML files and their associated .ibd files from the MALDI analysis of the fingermarks of a pool of suspects can be stored in one computer folder, like a "case database". Once stored, the files have to be selected either in a dedicated software or in the open source R Shiny App used in this research. Each step is detailed in the instructions of the Shiny App panel and only two processing buttons have to be clicked to extract the intensities and obtain a ready-to-use .csv file.

Finally, regarding the LMT model implementation in R, the Caret package was used as mentioned in "2.3.4 Data processing". This is the most common way of performing machine learning in R. Once the code is created, you can always use the same one and only update the data path and the file name. The data treatment process is user-friendly and accessible to anyone with minimum training. The overall computer data treatment took on average no more than 30 min for 13 donors, which remains quick and efficient. Overall, depending on the chosen resolution and size of analysis, the whole processing can be time consuming regarding other techniques. However, it would provide a new discriminant method to exploit the fingermark residue evidence that could also be useful when other detection techniques might not be successful (e.g., when the friction ridge pattern and/or the contact DNA are not of sufficient quality or cannot be exploited).

5. Conclusion

The aim of this study was to investigate the capacity to differentiate 13 donors, men and women of varying ages, based on the chemical composition of their fingermarks, collected during over one year. MALDI-MSI combined with machine learning models led to highly promising results.

Within the methodological frame of this study, the most performant multi-class classification model was determined to be LMT. Correct fingermark classification was determined for up to 90% of the fingermarks, with an AUC of 0.993.

These findings are encouraging in the perspective of the exploitation of the composition of fingermarks, when the ridge pattern is of insufficient quality and touch DNA unlikely to be collected. Using dedicated R tools, the data treatment after analysis do not exceed 30 min and is relatively user-friendly.

This study having been conducted on ideal fingermarks (i.e. 1-day-old marks left on glass), further research is required to investigate the impact of the substrates, of the aging time, or of the fingermark detection techniques on the fingermark chemical composition, and ultimately on the machine learning model performances.

CRediT authorship contribution statement

Marie Gorka: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Aurélien Thomas:** Conceptualization, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Andy Bécue:** Conceptualization, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Conflicts of interest

There are no conflicts to declare.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.forsciint.2023.111645.

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