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Study of latent fingermarks by matrix-assisted laser desorption/ionisation mass spectrometry imaging of endogenous lipids

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Identification of suspects via fingermark analysis is one of the mainstays of forensic science. The success in matching fingermarks, using conventional fingermark scanning and database searching, strongly relies on the enhancement method adopted for fingermark recovery; this in turn depends on the components present in the fingermarks, which will change over time. This work aims to develop a robust methodology for improved analytical detection of the fingermark components. For the first time, matrix-assisted laser desorption/ionisation mass spectrometry imaging (MALDI-MSI) has been used to image endogenous lipids from fresh and aged, groomed and ungroomed fingermarks. The methodology was initially developed using oleic acid which was detected along with its degradation products over a 7-day period, at three different temperatures in a time-course experiment. The optimised methodology was then transferred to the imaging analysis of real fingermark samples. Fingermark patterns were reconstructed by retrieving the *m/z* values of oleic acid and its degradation products. This allowed the three aged fingermarks to be distinguished. In order to prove that MALDI-MSI can be used in a non-destructive way, a simple washing protocol was adopted which returned a fingermark that could be further investigated with classical forensic approaches. The work reported here proves the potential and the feasibility of MALDI-MSI for the forensic analysis of fingermarks, thus making it competitive with other MSI techniques such as desorption electrospray ionisation (DESI)-MS. The feasibility of using MALDI-MSI in fingermark ageing studies is also demonstrated along with the potential to be integrated into routine fingermark forensic analysis. Copyright © 2009 John Wiley & Sons, Ltd.

Fingermarks are the result of material from the surface of the skin transferring to another surface on contact. They are distinct from fingerprints, which are control prints where the donor is known and either ink is used or the fingertip is scanned directly. Fingermarks can be composed of endogenous and exogenous substances, depending on what the depositing skin has been in contact with. The glands on the palms of the hands and feet secrete different substances from those on other parts of the body; however, in many cases, due to regular contact with other areas, such as the face, fingermarks are composed of substances from both sources, in particular lipids, fatty acids, amino acids, vitamins and water.[1] Fingermarks that are composed of opaque materials, e.g. blood, dirt, paint, are termed patent marks and are easily visible; however, fingermarks that are made of transparent materials, e.g. gland secretions, are termed latent marks and are not easily visible.

In order to make a comparison, a latent fingermark must be made visible using a development technique, preferably one that is targeted at the substances present in the mark. In addition, it has been proven that changes in composition occur with time as components degrade.[2] In order to maximise the effectiveness of enhancement techniques, work on fingermark composition has been carried out in several areas; the determination of the range of substances typically found in fingermarks over time is one of these areas. Recent studies have analysed classes of components as a whole, e.g. lipids and proteins,[3] specific members of a class, e.g. proteins,[4] and another study also examined the changes in lipids over time using gas chromatography/mass spectrometry (GC/MS).[5] A further study captured images of cyanoacrylate-enhanced marks over time but, on that occasion, did not investigate the chemical changes taking place.[6]

Another area of research concerns the chemical imaging of compounds present in fingermarks using fluorescent antibodies[7] or advanced analytical techniques such as surface-assisted laser desorption/ionisation time-of-flight mass spectrometry (SALDI-TOF-MS),[8] desorption electrospray ionisation mass spectrometry imaging (DESI-MSI)[9] and attenuated total reflectance Fourier transform infrared (ATR-FT-IR).[3]

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Hazarika and collaborators reported an application combining the properties of magnetic powders with that of antibody recognition through the formation of antibody–magnetic-particle conjugates in order to detect drugs and drug metabolites within latent fingermarks.[7] High-definition fluorescence images of the fingermark pattern were then obtained which demonstrated not only the opportunity to detect drug use, but also the possibility of identifying an individual with the ease of application and removal of magnetic fingermark powder.

A few other groups have reported similar applications in which they reconstructed the fingermark pattern by imaging exogenous compounds such as explosives, drugs and their metabolites by SALDI-TOF-MS[8] and DESI-MSI.[9] These techniques make no use of molecular probes of any kind, thus improving the time- and the cost-effectiveness of the analysis. DESI also proved to be capable of imaging endogenous fatty acids (e.g., palmitic and oleic acid) and triacylglycerols in groomed fingerprints. A major advantage of the DESI technology, especially in this context, is the potential for it to be made portable, meaning that analyses could be carried out on site. Although DESI has been demonstrated to be applicable to many fields of forensic science,[10] in the context of fingermarks, sensitivity and spatial resolution aspects, as well as minaturisation, need to be addressed. Rowell and collaborators[8] reported the development of a mass spectrometry methodology for direct analyses of pre-dusted lifted fingermarks. The aim here was to detect contact residues co-deposited onto surfaces following handling of illicit drugs, or drugs and metabolites secreted from the body onto the skin following their use. In the methodology as reported, a dusting agent was used, for locating/visualising the marks and, at the same time, also acting as a SALDI-TOF-MS enhancer for analytical instrumental detection.

Despite the success of such approaches, the targeted analytes were drugs of abuse rather than endogenous substances. Clearly, this will be useful in some but not all cases as only a proportion of the deposited marks will contain these substances. Ricci and collaborators successfully imaged natural secretions of fingermarks, thus making their technology more specific and of a wider applicability.[3]

The development of methods to physically or chemically visualise fingermarks depending on their composition is currently attracting a great deal of attention in the forensic investigation of latent fingermarks. Various methods have been investigated including powders, nanoparticles and metal deposition.[11–13] Despite all this work and the fact that fingermark examination is one of the approved and official methods of forensic analysis used for suspect identification, the need for ever more effective methods of visualisation, particularly analytical, is still 'strongly felt'.[3,10]

In the work reported here, two areas of forensic investigation are addressed, namely the enhancement and identification of the fingermark. Matrix-assisted laser desorption/ionisation mass spectrometry imaging (MALDI-MSI) has been employed as a molecular non-destructive imaging technology to analyse latent fingermarks. In this context, 'non-destructive' means that, upon analysis, the forensic evidence is not lost; not all the material from the fingermark is in fact, desorbed and it is possible to wash away the matrix and enhance the fingermark for analysis with classical forensic protocols. MALDI-MSI enables images of the distribution of xenobiotics and biomolecules such as lipids, peptides and proteins to be obtained directly from intact tissue sections.[14] Like DESI-MS and spectroscopic imaging techniques such as Raman and FT-IR, images are retrieved without resorting to antibodies or molecular probes of any kind. Since the first paper on MALDI-MSI was published in 1997,[15] the technique has had enormous impact on several lifescience fields including medicine, pharmaceuticals, biotechnology and microbiology. Here, for the first time we demonstrate the application of this technique to forensic science and we propose this as a new complementary approach to fingermark identification. The opportunity to use MALDI-MSI in a non-destructive way, as shown in this work, makes it an even more powerful tool for supporting forensic investigation. In this approach, a wider range of information is obtained, complementing that derived from classical forensic approaches, as well as keeping the evidence intact. As lipid components are generally present in all marks and, since these will persist on wetted as well as unwetted items, we investigated the potential of MALDI-MSI for detecting and imaging a range of fingermark lipids from groomed and ungroomed fingermarks; groomed fingermarks are artificially loaded with sebaceous secretions by contact with the face and ungroomed fingermarks are deposited with no sample preparation. The possibility of monitoring changes in the lipid composition over time and at different temperatures was also investigated and preliminary results are shown here for oleic acid.

EXPERIMENTAL

Materials

Oleic acid (~99%), trifluoroacetic acid (TFA), ALUGRAM® SIL G/UV$_{254}$ pre-coated aluminum sheets and α-cyano-4-hydroxycinnamic acid (CHCA) were purchased from Sigma-Aldrich (Poole, UK). Acetone and acetonitrile (ACN) were obtained from Fisher Scientific (Loughborough UK). MALDI target OPTI TOF 192-well inserts and spotless inserts were purchased from Applied Biosystems (Foster City, CA, USA). Double-sided conductive carbon tape was purchased from TAAB (Aldermaston, UK).

Instrumentation

All mass spectrometric analyses were performed using a modified Applied Biosystems API Q-Star Pulsar i hybrid quadrupole time-of-flight (QTOF) instrument. The orthogonal MALDI source has been modified to incorporate a SPOT 10 kHz solid-state laser (Elforlight Ltd., Daventry, UK), having a wavelength of 355 nm, a pulse duration of 1.5 ns and producing an elliptical spot size of 100×150 μm (the details of this modification will be reported elsewhere). Images were acquired using 'oMALDI Server 5.1' software supplied by MDS Sciex (Concord, Ontario, Canada). Latent marks were sprayed using a 'SunCollect' auto-spraying system (Sunchrom GmbH, Friedrichsdorf, Germany).

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Preparation of oleic acid samples

The oleic acid (OA) solution was prepared in ACN at a concentration of 6.3 mmol/L; 0.5 μL was spotted onto an OPTI TOF 192-well insert and allowed to dry for preliminary MALDI-MS and -MS/MS experiments. A set of similarly spotted OA samples was stored at three different temperatures, 4°C, 37°C and 60°C. The target plates were kept in a Petri dish, in the dark, and stored in those conditions over 7 days in a time course to conduct ageing experiments. Four time points were investigated (0, 1, 4 and 7 days). When ready, samples were spotted with 0.5 μL of the matrix α-CHCA, prepared at a concentration of 10 mg/mL in a 70:30 ACN/0.1% TFA solution which was allowed to dry for 10 min before analysis. These ageing experiments were repeated four times.

MALDI-TOF-MS and -MS/MS analysis of OA samples

MALDI-MS spectra were obtained in positive ion mode in the mass range between *m/z* 100 and 1000. Declustering potential 2 was set at 15 arbitrary units and the focus potential at 10 arbitrary units, with an accumulation time of 0.117 min. MALDI-MS/MS spectra were obtained in the mass range between *m/z* 50 and 284, using argon as the collision gas. In separate MS/MS experiments, the protonated oleic acid (*m/z* 283.27) and the protonated dehydrated oleic acid (dehydrated OA, *m/z* 265.25) were selected as precursor ions; declustering potential 2 was set at 15 and the focus potential at 10, the collision energy was set at 40 arbitrary units, and the accumulation time was 0.117 min. Ageing experiments were performed by MALDI-MS where, at each ageing time point, the relative signal intensities of the protonated OA, dehydrated OA and di-dehydrated oleic acid (di-dehydrated OA, *m/z* 247.24) peaks were plotted against the time points investigated for the three different temperature conditions analysed.

Fingermark preparation

Latent groomed fingermarks were prepared by rubbing the fingers on the forehead, nose and chin, five times, to produce a sebum-rich mark[5] that contained an abundance of the fatty acids of interest, such as OA. In order to produce ungroomed fingermarks, hands were washed using alcohol wipes and normal work activities were continued for a period of 15 min before deposition. The marks were laid onto the ALUGRAM® SIL G/UV_{254} pre-coated aluminum sheets after scraping off the silica with acetone. For ageing experiments, groomed marks were replicated three times for each of the temperature conditions (4°C, 37°C and 60°C) and stored for 7 days to allow the ageing of the sebaceous content. Once the marks had aged for the allocated amount of time, they were sprayed with an α-CHCA matrix solution using the SunCollect auto-sprayer. The matrix was prepared at a concentration of 5 mg/mL in a 70:30 ACN/0.1% TFA solution and sprayed in four layers at a speed of 1, 2, 2 and 2 μL/min.

MALDI-MSI analysis of aged latent fingermarks

MALDI-TOF-MS analysis of fingermarks was performed at a resolution of 150 μm × 150 μm using 'continuous raster imaging' at a laser repetition rate of 10 kHz. Differently from the classic 'stop and go fashion' of MALDI-IMS, in this option, the laser is moved continuously in rows across the sample surface providing time savings at high image resolution. In this way, fingermark samples of the size of ~1.7 cm × 2 cm were imaged in ~44 min instead of 8 h. Data processing was performed by using BioMap 3.7.5 software (Novartis, Basel, Switzerland).

Fingermark recovery post-MALDI-MSI and enhancement

α-CHCA was removed by submerging the support with the fingermark in a 70:30 ACN/0.1% TFA solution which was then tilted and dried at room temperature for 10 min. Magnetic powder was then used to enhance the fingermark and an image was taken by means of a scanner at 9600 dpi. The support was in one case a glass slide and, in the other, an aluminium sheet.

RESULTS AND DISCUSSION

MALDI-MS analysis of OA samples and ageing experiments

The experimental methodology was developed in a MALDI-MS study of the endogenous monounsaturated omega-9 fatty acid oleic acid (OA, MW 282.2559). A range of MALDI matrices (DHB, sinapinic acid and α-CHCA), matrix preparation conditions (different solvent combinations such as ethanol and ACN) and matrix additives such as sodium acetate was explored (data not shown). The best results were achieved using α-CHCA (5 mg/mL prepared in 70:30 ACN/0.1%TFA solution) as matrix. Preliminary MALDI-MS and -MS/MS spectra were acquired in order to evaluate the OA ionisation yield. Figure 1(A) shows the positive ion MALDI-MS spectrum of OA. This exhibits the expected $[M+H]^+$ peak at *m/z* 283.27 and two further ions at *m/z* 265.26 and 247.24, corresponding to the protonated OA after loss of one and two molecules of water, respectively (dehydrated OA and di-dehydrated OA). The two OA dehydrated species result from OA fragmentation under the MS instrumental conditions used. These three ions are also found in the MALDI-MS/MS spectrum of protonated OA (Fig. 1(B)) whereas di-dehydrated OA is present in the MS/MS spectrum of dehydrated OA, thus demonstrating that the dehydrated species originate from OA (Fig. 1(C)).

Several two-fold dilutions from a 6.3 mmol/L solution of OA were analysed by MALDI-MS and it has been estimated that the lowest detectable amount of OA is ~0.9 ng.

Ageing experiments were also performed by spotting a solution of OA on three target plates and storing them at 4°C, 37°C and 60°C, respectively, over a 7-day period. The samples were analysed after 0, 1, 4 and 7 days in four replicates by MALDI-MS and the results are shown in Fig. 2. As expected, the low temperature (4°C) at which OA was stored did not produce degradation of the lipid. Its integrity was preserved, as is shown by the constant signal intensities of OA, dehydrated OA and di-dehydrated OA throughout the 7-day storage period. It can be deduced

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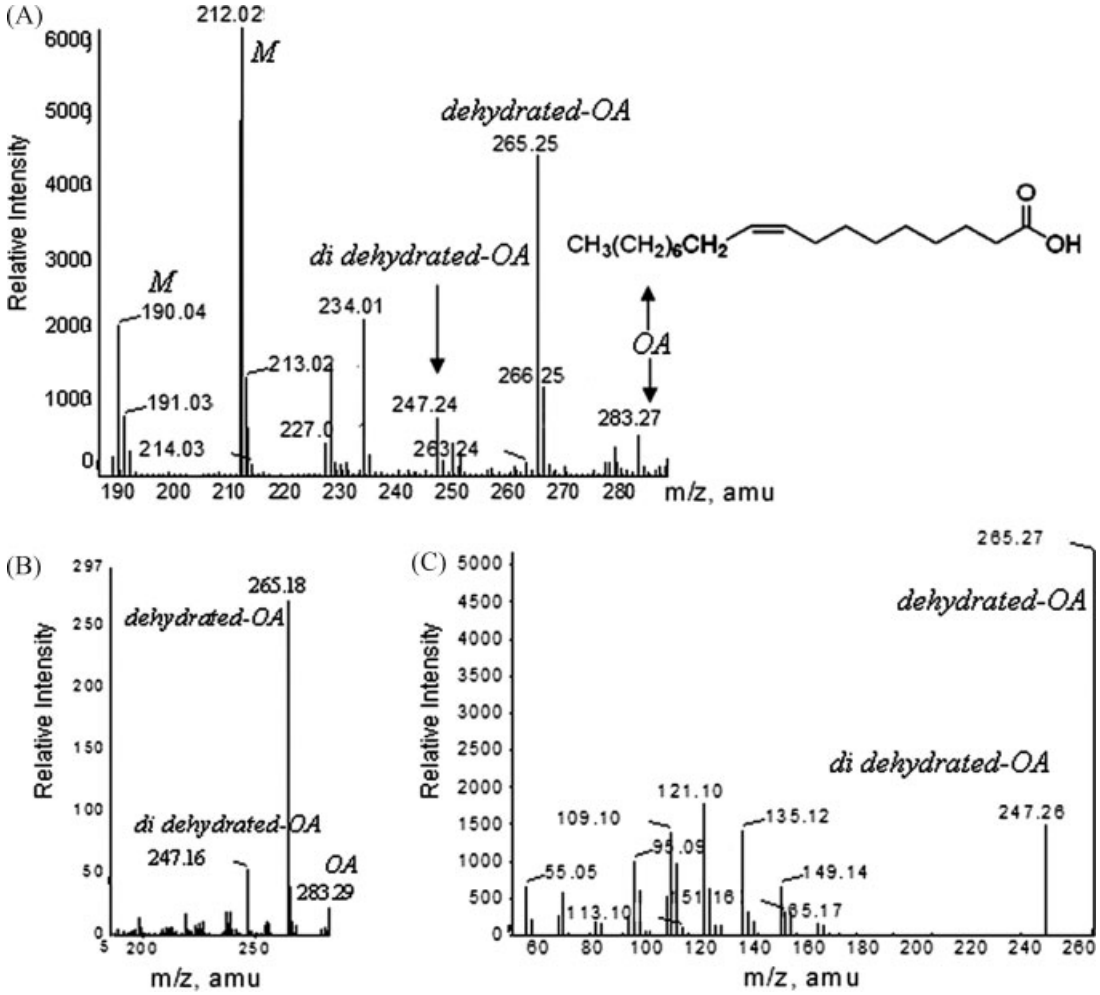

Figure 1. MALDI-MS and -MS/MS analysis of standard oleic acid (OA). (A) Molecular structure of OA and its MALDI-QTOF-MS spectrum exhibiting a peak at *m/z* 283.27 corresponding to protonated OA. An ion is observed at *m/z* 265.25 corresponding to the loss of a molecule of water from OA (dehydrated OA). A second molecule of water is then lost generating the species at *m/z* 247.24 (di-dehydrated OA). In (A) the matrix ion signals are also labelled (M). (B, C) MALDI-QTOF-MS/MS spectra of OA and dehydrated OA, respectively, confirming the above attributions as well as showing the progressive loss of alkyl groups.

then, that, in this instance, the formation of dehydrated OA and di-dehydrated OA occurs in the mass spectrometer due to the instrumental conditions used. At 37°C it is possible to observe ageing of OA as its intensity constantly decreases throughout the 7-day period. Dehydrated OA, instead, undergoes a more important decrease over the same period whereas the ion intensity of di-dehydrated OA shows a rapid decrease at 37°C in 1 day, and then remains fairly constant in the remaining period of storage. At 60°C, the intensity of the OA signal exhibits a similar trend to that observed at 37°C, which is still consistent with an ageing process although there seems to be a higher rate of decrease in the first day. Compared with the analysis at 37°C, at 60°C dehydrated OA and di-dehydrated OA exhibit a higher degradation rate over 4 days, whereas the ion intensities remain fairly constant between 4 and 7 days storage although with very low signal-to-noise. This would suggest accelerated analyte dehydration at higher temperature.

MALDI-MS analysis of latent fingermarks

MALDI-MS reference spectra were recorded for both ungroomed and groomed fingermarks in order to evaluate the ionisation yield of OA, dehydrated OA and di-dehydrated OA in both cases. Fingermarks were sprayed with α-CHCA matrix and spectra were acquired in five random regions. Representative MALDI-MS spectra are shown in Fig. 3. Although well detected, in both the ungroomed (Fig. 3(A)) and groomed fingermarks (Fig. 3(B)), the above species exhibit very low relative ion intensity (see enlarged insets). In particular, as expected, the ion intensity of the three species is higher in the groomed fingermark, as more sebaceous material has been accumulated.

MALDI-MSI analysis of latent fingermarks

An ungroomed fingermark was deposited onto an aluminium TLC sheet, auto-sprayed with α-CHCA and analysed by MALDI-MSI (see Experimental section) to obtain a reference image.

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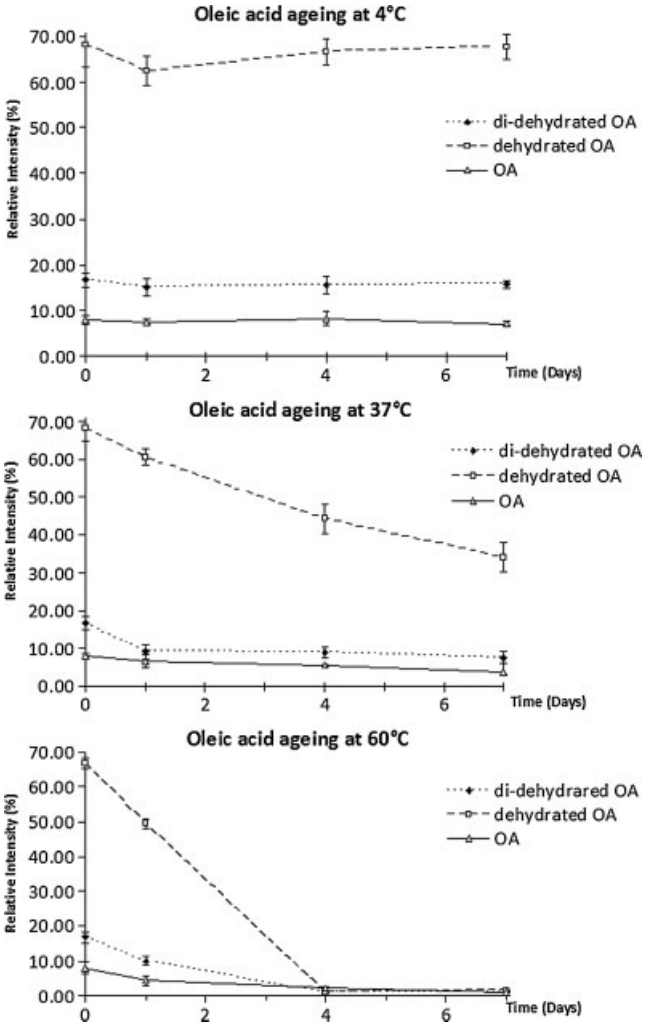

Figure 2. Oleic acid ageing analysis. The graphs plot the intensity of OA, dehydrated OA and di-dehydrated OA ion signals against the ageing time points selected (0, 1, 4, 7 days). Ageing temperatures were 4°C, 37°C and 60°C. Error bars are displayed and obtained for four replicates.

Interestingly, prior to analysis, a scan image of the fingermark was taken, which exhibited poor visualisation of the salient physical features usually used in forensic classical methodologies to match suspects' fingermarks (data not shown). In contrast, by using MALDI-MSI, an image of the distribution of a ubiquitous species at *m/z* 230 (Fig. 4(a)) was obtained which showed the whole fingermark and its ridge pattern (not normalised but optimised for contrast and intensity). Although it can be speculated, on the basis of the *m/z* value and its MS/MS spectrum, that this ion is a fatty acid (13-aminotridecanoic acid), in this context, the identity of this ion is not important as it can simply and efficiently act as a diagnostic ion to reconstruct whole fingermark pattern. At this point, is worth mentioning that, perhaps surprisingly, the matrix ion signals did not interfere with the detection of the species of interest.

As ridge patterns are the features by which classical forensic analysis bases a suspect's identification, this feature makes MALDI-MSI both an enhancement and a detection technique. In addition, MALDI-MSI allows chemical imaging, thus making it a very specific technology; in a single analysis, it was possible to observe the distribution of many endogenous lipids (constituting potential individual features) including cholesterol, palmitoleic acid (de-hydrated, and also found as the sodiated species), stearic acid, OA, dehydrated OA and di-dehydrated OA, nonadecanoic acid, phosphoethanolamine and diacylglycerols. An example of the corresponding images of the distribution of these ions in an ungroomed fingermark is shown in panel 1 of Figs. 4(b)–4(g) and some of the corresponding mass spectra are reported in panel 2 of Fig. 4. The images were characteristic for each of the species detected. Incidentally, as reported in Fig. 4(g), an ion was also detected at *m/z* 550.6 which is compatible with the presence of the dimethyldioactadecylammonium ion. This species corresponds to an exogenous compound, previously reported by Manier and co-workers[16] as a contaminant originating from lotions, hair products, body washes and paper used for tissue products. The MS/MS spectrum of this ion is also in agreement with that reported by Manier and co-workers (data not shown).

This was the first very clear demonstration of the potential application of MALDI-MSI in the forensic investigation of fingermarks thus making it competitive with other MSI techniques such as DESI. Notably, whilst DESI has been shown to generate mass spectrometric images for fatty acids from sebum-rich (groomed) fingermarks, MALDI-MSI provided fatty acid images from ungroomed fingermarks which are more representative of the type of fingermark found at a crime scene. This circumstance would relate to the current superior sensitivity of MALDI-MSI over DESI-MSI.

An ageing study was also performed on groomed fingermarks in an effort to correlate lipid distribution density maps with age under particular 'degradation conditions'. The authors are currently investigating a range of ageing conditions and, for this work, they focused on the effect of temperature, selecting 7 days as the only time point. Although a single analysis gives information on many lipids, endogenous OA has been the primary target, as previous *'non in situ'* ageing experiments have been conducted with this lipid for this study. In order to investigate the difference between fingermarks of the same age but aged at different temperatures, fingermarks were deposited, aged for 7 days at 4°C, 37°C and 60°C, spray-coated with matrix and submitted to MALDI-MSI analysis. The MS images obtained allowed a few observations to be made (panel 1, Fig. 5); the density map of OA from a non-aged groomed fingermark shows an important and almost ubiquitous distribution of OA whereas dehydrated OA exhibits a much lower density map and di-dehydrated OA is only represented by a few pixels (Figs. 5(a)–5(c)). Compared with this, a 7-day-old fingermark stored at 4°C exhibits a slightly lower OA distribution (in agreement with previous ageing analysis on OA, see above) and the dehydrated species exhibit a very similar density map to that shown for the reference groomed fingermark (Figs. 5(d)–5(f)). The similar distribution of the dehydrated species in the non-aged groomed fingermark and in the one stored at 4°C is again in agreement with their formation in the mass spectrometer, due to the instrumental conditions selected, and not an indicator of an ageing process. The MS images show a progressive degradation of OA as the storage temperature

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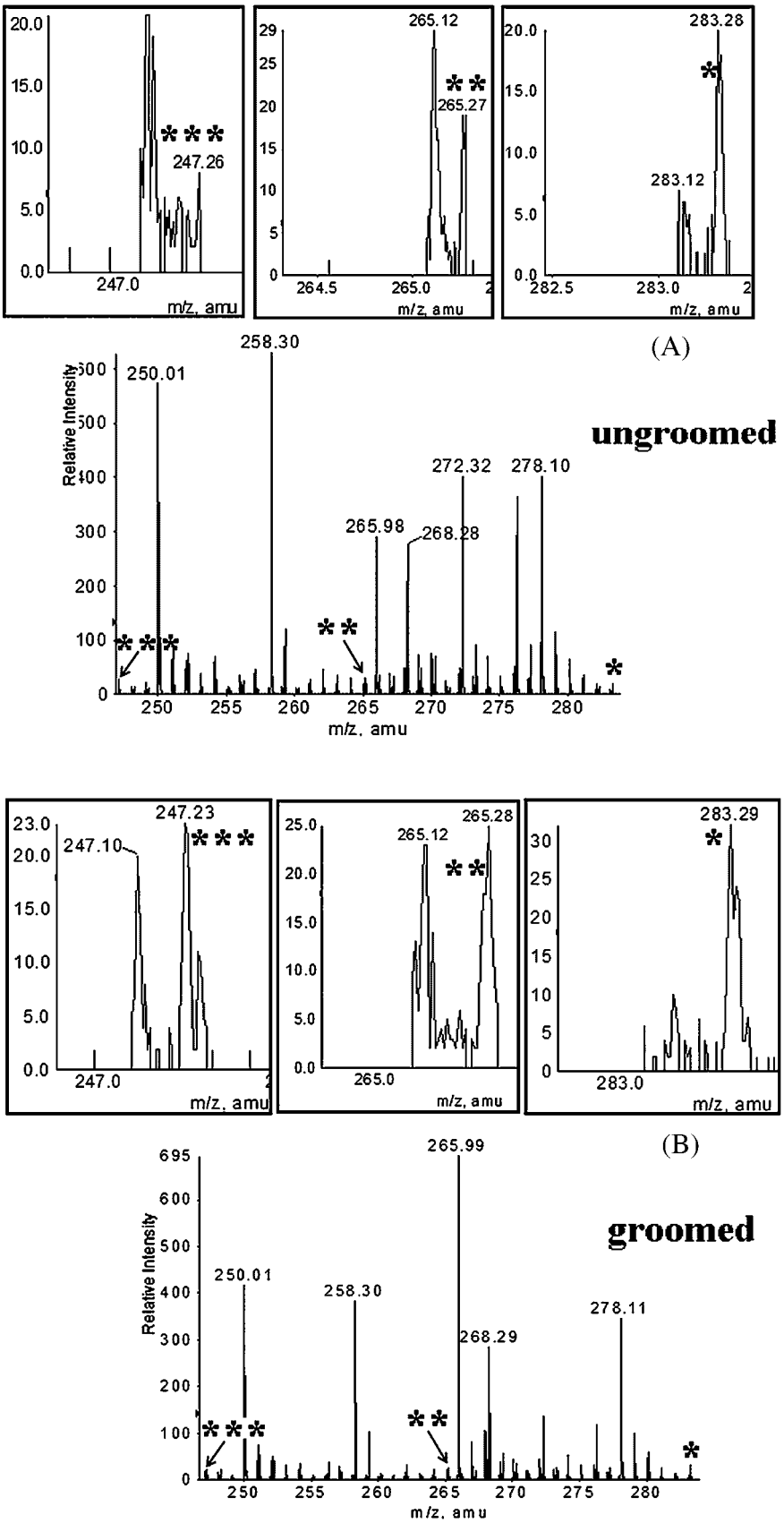

Figure 3. MALDI-MS spectra of ungroomed and groomed fingermarks. MALDI-MS spectra were extracted from ungroomed (A) and groomed (B) fingermarks. The enlarged insets show, for each fingermark type, OA (*) and the species generated by the single (**) and double (***) loss of water under the experimental conditions used.

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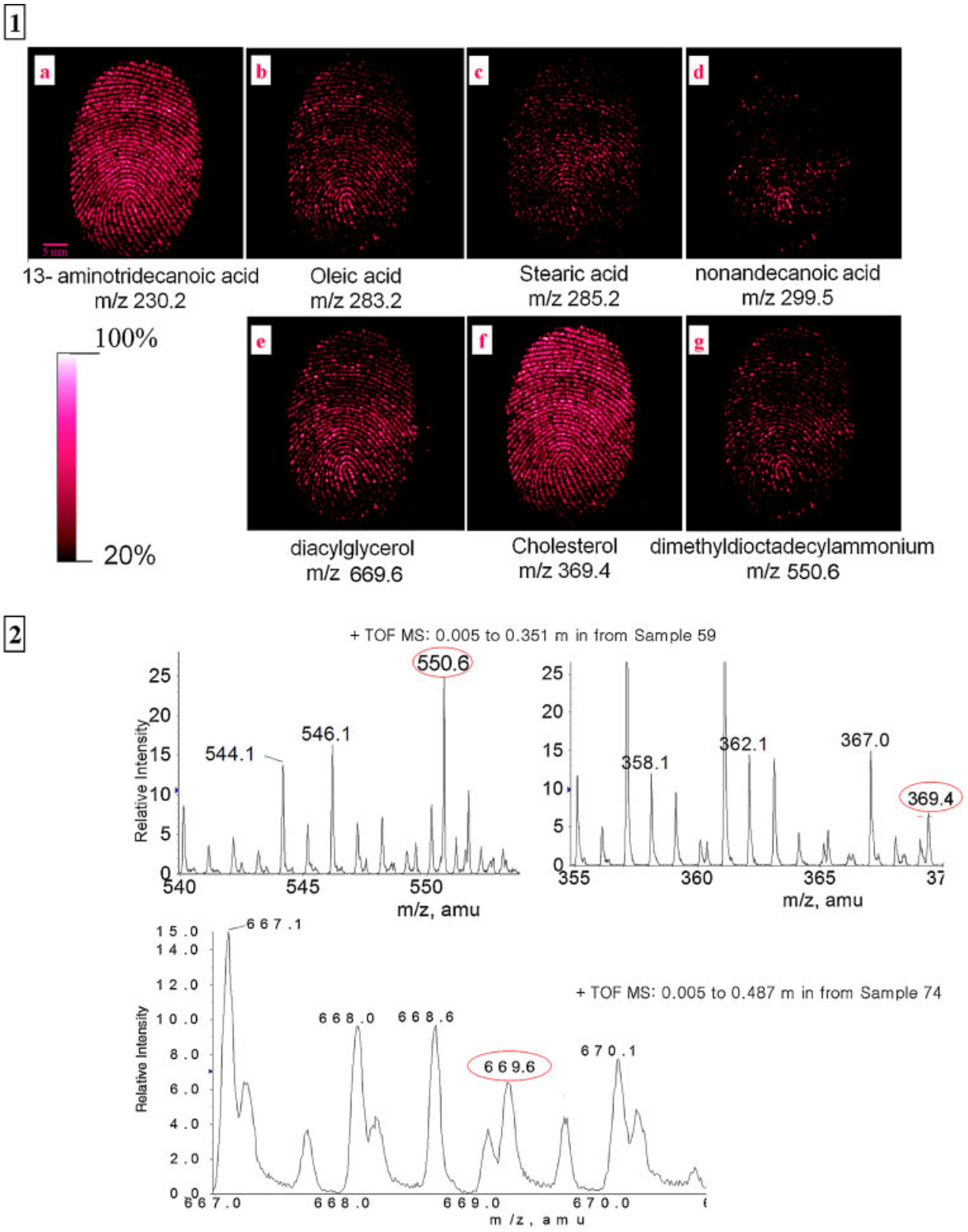

Figure 4. MALDI-MSI analysis of an ungroomed, fresh fingermark. (a) Fingermark MS image reconstructed from the ion signal at *m/z* 230.2. (b–g) MS images of distribution of OA, stearic acid, nonadecanoic acid, diacylglycerol ($C_{43}H_{72}O_5$), dehydrated cholesterol and dimethyldioactadecylammonium, at *m/z* 283.2, 285.2, 299.5, 669.6, 369.4 and 550.6, respectively. Panel 2 reports representative MS spectra extracted from the images in panel 1.

increases from 4 to 60°C (Figs. 5(d), 5(g) and 5(j)), which is, again, in agreement with the preliminary degradation studies carried out on OA aged on MALDI inserts. Panel 2 of Fig. 5 displays the corresponding MALDI-MS spectra of OA showing the decrease of its signal intensity as the temperature increases.

The trend described is supported by images of the distribution of dehydrated OA, the density maps of which are different for the three ageing temperatures and show a pronounced increase in its formation when the storage temperature is increased from 4 to 37°C whereas the species is very difficult to detect after 7 days at 60°C (Figs. 5(e), 5(h)

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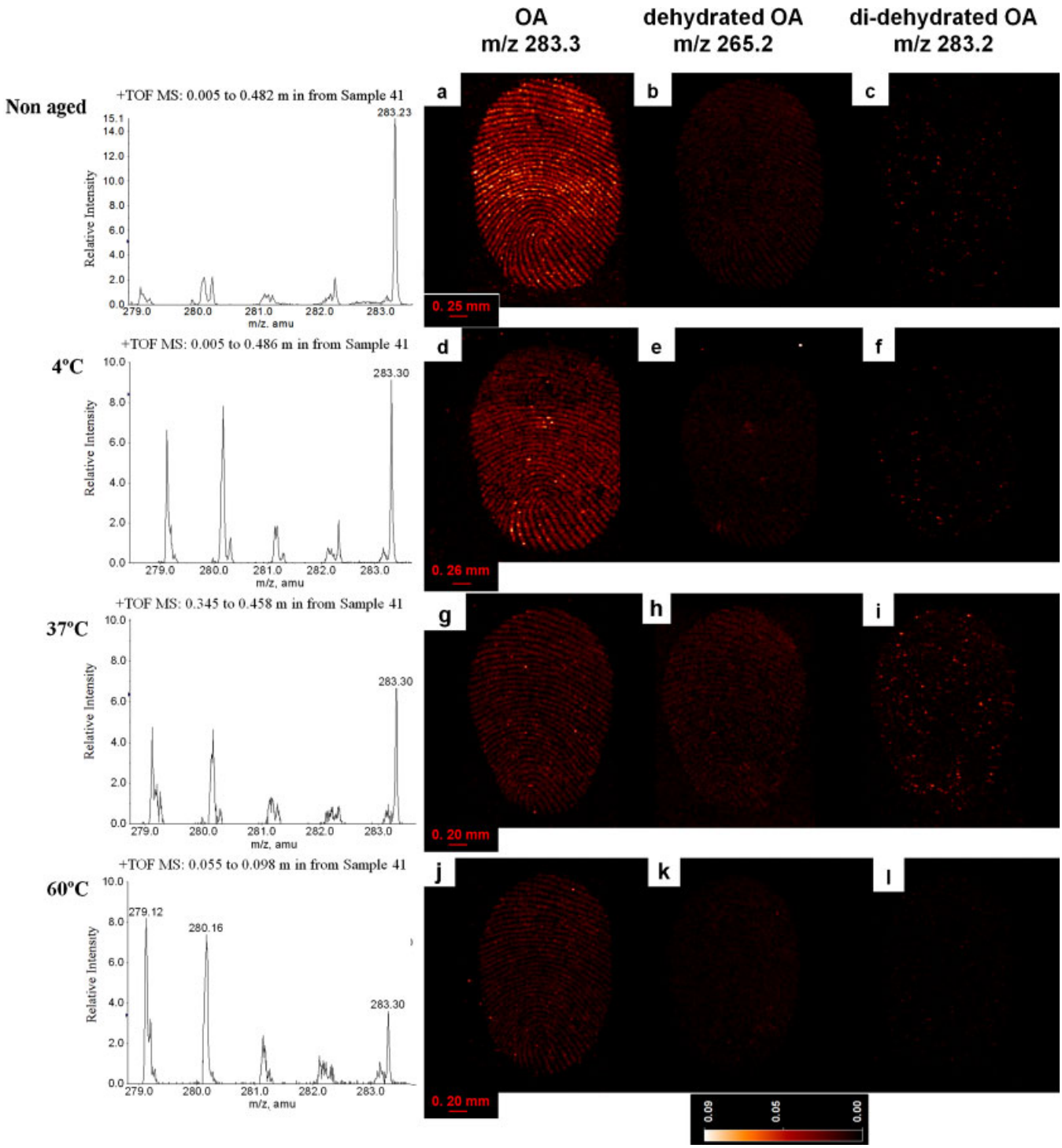

Figure 5. Groomed fingermark ageing analysis by MALDI-MSI. Panel 1, insets a–c show the MS images of OA (*m/z* 283.27), dehydrated OA (*m/z* 265.25) and di-dehydrated OA (*m/z* 247.24) in a non-aged groomed fingermark. The same set of MS images for 4°C, 37°C and 60°C are shown in panels d–f, g–i and j–l, respectively. Images indicate lipid degradation, as the temperature increases, with distribution maps for the species of interest that significantly change between 4°C, 37°C and 60°C. Panel 2 reports MALDI-MS extracted spectra of OA from the corresponding fresh and aged fingermark images, showing the decrease in OA signal intensity upon storage temperature increase.

and 5(k)) presumably due to degradation of the fatty acid carbonyl structure. A similar trend to that of dehydrated OA is shown by the di-dehydrated OA throughout the range of temperatures investigated (Figs. 5(f), 5(i) and 5(l)).

These observations suggest the feasibility of this technology for image reconstruction and discrimination of aged fingermarks. Additional studies are required to confirm the potential of MALDI-MSI to date fingermarks, taking account of environmental conditions. If this possibility is realised, the technology could be used to place a suspect on the scene of crime, to any time before, during or after the crime. The investigation of aged fingermarks is ongoing in the authors' laboratories.

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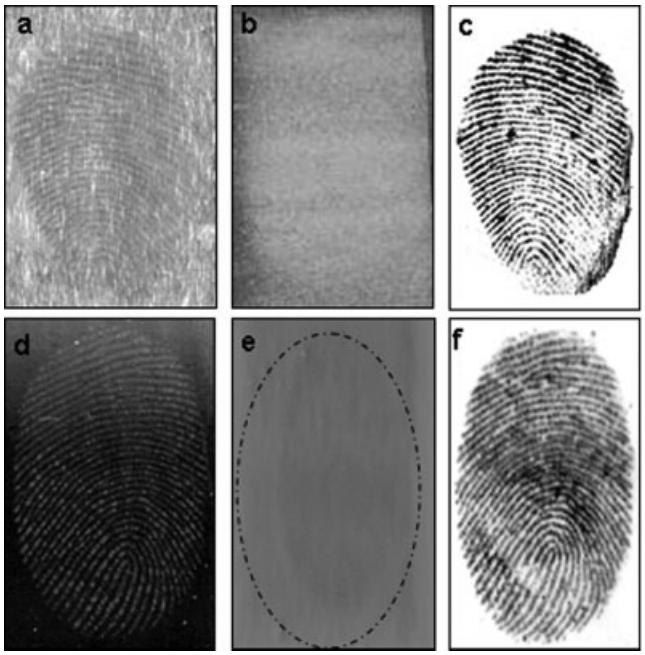

Figure 6. Scan images of an ungroomed fingermark (a) before matrix deposition, (b) after matrix deposition and (c) after matrix removal on an aluminium sheet. The same set of images is shown when glass is used as deposition surface (d–f).

Fingermark recovery post-MALDI-MSI and enhancement

Since MALDI-MSI is generally thought to be a destructive technique, we used a simple protocol to demonstrate that, in fact, post-MALDI-MSI analysis, the fingermarks can still be analysed by more classical forensic methods. A simple wash of the fingermark in the mixture of solvents used for the matrix preparation (see Experimental section) would not remove the non-desorbed lipids, whereas the matrix would be washed away. This is confirmed by a second MALDI-MSI analysis upon matrix re-application; the intensity of OA is comparatively much lower than is observed in the first analysis as are the intensities of the dehydrated OA although these species were able to generate MS images, whereas di-dehydrated OA is no longer detected (data not shown).

The 'washed' fingermark could be enhanced with magnetic powder and a scan image was taken at 9600 dpi showing ridges and minutiae which can be further searched and matched in the existing forensic databases. Two deposition surfaces were tested, aluminium sheet (Figs. 6(a)–6(c)) and glass (Figs. 6(d)–6(f)) and, in both cases, fingermark enhancing post-matrix removal was successful.

CONCLUSIONS

The forensic analysis of suspects' fingermarks can now benefit from a powerful technology, namely matrix-assisted laser desorption/ionisation mass spectrometry imaging (MALDI-MSI). For the first time, the potential and the feasibility of MALDI-MSI were demonstrated by imaging endogenous lipids in groomed and ungroomed fingermarks. In addition, the successful discriminating analysis of aged fingermarks by MALDI-MSI further demonstrates its employability on an even higher level of analysis complexity. Much older fingermark samples, stored under a variety of conditions, are currently being analysed to improve the robustness and reliability of the methodology as well as to establish links between age, ageing conditions and lipid degradation (mass images). The possibility of re-using fingermarks, already analysed by MALDI-MSI, in classical forensic approaches, makes this technology non-destructive and empowers fingermark identification.

Currently, the authors are developing a multidisciplinary analytical approach to support and reinforce MALDI-MSI results and to look at other fingermark components such as amino acids and proteins to widen and deepen the applicability of the technology.

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