Application of MALDI MS Imaging after sequential processing of latent fingermarks

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Technical Note: Application of MALDI MS Imaging after sequential processing of latent fingermarks

Abstract

Latent fingermarks are routinely visualised by subjecting them to one or more CSI/crime lab processes to maximise the recovery of ridge flow and *minutiae* permitting an identification. In the last decade mass spectrometric imaging (MSI) techniques have been applied to fingermarks to provide information about a suspect and/or on the circumstances of the crime as well as yielding additional images of the ridge pattern. In some cases, these techniques have shown the ability to provide further ridge detail, "filling in the gaps" of the developed mark. Matrix Assisted Laser Desorption Ionisation Mass Spectrometry Imaging (MALDI MSI) is presently the most advanced of the so-called 'surface analysis' techniques, in terms of compatibility with a number of fingermark enhancement processes and implementation in operational casework. However, for the use of this technique in major crimes to become widespread, compatibility with sequential processing must be demonstrated. This short study has assessed compatibility with a number fingermark processing sequences applied to natural marks on the adhesive side of brown (parcel) and clear tapes. Within the study undertaken, the results confirm the possibility to use MALDI MSI in sequence with multiple processes offering in some instances, complementary ridge detail with respect to that recovered from marks developed by conventional sequence processing.

Keywords: latent; fingermarks; sequential processing; MALDI MSI; mass spectrometry

Introduction

Latent fingermarks are routinely visualised by subjecting them to one or more crime scene investigation (CSI)/crime lab processes to maximise recovery of ridge flow and *minutiae* permitting an identification. Sequential processing is possible as a single chemical development reagent can still leave a considerable proportion of fingermark constituents that can be targeted by an additional enhancement technique [1].

When these enhancement techniques are used in a sequence, an *optimum* order of application is recommended , starting from the least destructive when possible, to maximise the biometric detail that can emerge from the different fingermark constituents. This overarching strategy is complemented by a set of rules that form the basis for establishing the "Fingermark Evidence Recovery Plan" as detailed by the Fingermark Visualisation Manual (FVM) edited by the Home Office, UK. The FVM covers in detail both the enhancement processes (optical chemical and physical) and the application order [2].

Whilst the fingerprinting workflow has remained largely unchanged for over hundred years (a crime scene mark is enhanced, one image is obtained by photographing/scanning and a match with a reference fingerprint is sought), the analytical community has invested significant efforts in the last decade to develop and adapt technologies enabling the provision of additional "fingermark images" to support biometric identification. Such technologies have emerged particularly from spectroscopy and mass spectrometry (MS) which can enable both detection of a vast range of fingermark constituents (*profiling* capability) and their visualisation (*imaging* capability) onto the ridge pattern. These capabilities provide the opportunity to *potentially* narrow down the pool of suspects by providing lifestyle and personal information on the owner of the mark (such abuse/handling of drugs, medications, blood groups and sex [3-6]) whilst linking this intelligence to the biometric information retrieved from the reconstruction of molecular images of their fingermarks [7-8]. These techniques have been previously reviewed and advantages and drawbacks have been outlined [8-10]. However, it is important to highlight one significant difference between spectroscopic and MS techniques; spectroscopic techniques rely on the detection of molecular functional groups to infer the presence of specific classes of compounds. Very often compound identification is achieved through matching to a reference database in a probabilistic approach. For this reason, MS specificity is generally preferred, where possible, as compounds are detected and identified through their specific mass-to-charge ratio (*m/z*) and through structural elucidation/confirmation by tandem mass spectrometry. Therefore, mass spectrometry imaging (MSI) specificity yields higher confidence in the provision of associative evidence (identification of forensically relevant substance onto the identifying ridges of a fingermark). Examples of MSI techniques adapted to fingermark analysis are Desorption Electrospray Ionisation (DESI), Matrix Assisted Laser Desorption Ionisation (MALDI), Secondary Ion Mass Spectrometry (SIMS) and Silver Laser Desorption Ionisation (AgLDI). These techniques have been reviewed, together with other potentially interesting MSI methods by Francese et al [8]. All of these techniques have been tested for their compatibility with the prior application of CSI and crime labs techniques to various degrees. These studies are very important to assess and identify the potential operational role of these techniques within the fingerprinting workflow. DESI was the first technique to be reported for the imaging of fingermarks [11]), but there is only one report showing compatibility with a fingermark enhancement technique (FET), namely cyanoacrylate fuming (CAF) [12].

SIMS is a well published technique for fingermark *imaging*. It has the highest spatial resolution amongst all the MSI techniques (submicron) and as such can provide extremely high quality fingermark molecular images to the extent of showing pore shapes [13] (though this type of high resolution for full fingermarks comes at the expense of significant acquisition times of up to 12

hours). However, whilst there are many examples where SIMS could be used as an alternative to conventional methods [14], there is only one paper in the literature demonstrating SIMS capability to enhance the quality of ridge detail in marks only partially developed by CAF and CAF followed by basic yellow 40 (BY40) [15].

AgLDI MS has demonstrated profiling and imaging compatibility with the application of a number of FET including Oil Red O (ORO), ninhydrin, Indandione-Zinc chloride (Ind-Zn), Silver Physical Developer (PD), CAF, powders (green fluorescent, white and black) acid black 1 and leucocrystal violet [16-18]. Lauzon et al are one of the only two groups to report on the application of an MSI technique following sequential processing of a mark through the sequences Ind-Zn-ninydrin-ORO (seq. 1) and CAF-rhodamine (seq. 2) [17]. Although the AgLDI fingermark image did not show the same quality as exhibited by the optical image taken after seq. 1, it did rival the quality of that exhibited by the optical image taken after seq. 2.

MALDI MSI is the most published analytical technique for fingermark imaging [9] and has shown to be compatible with the prior application of single processes such as vacuum metal deposition (VMD), CAF [19], powders (carbon black, white, aluminium), ninhydrin, acid black 1 [20-22], acid yellow 7, leucocrystal violet (Kennedy et al 2020, *accepted,* Scientific Reports) and indandione (Fischer et al, *in preparation*) on a number of different surfaces, directly or following recovery using tape lifts. In a few cases, it has been possible to show that MALDI MSI provided additional ridge detail in partially developed or empty marks [19]. In line with previous reports on constituents availability after the application of a single FET [1], MALDI is able to detect and image constituents or classes of compounds even after being targeted by a prior FET [19,22] and even in specimens over 30 years old [9]. Additionally, four examples have been published in which MALDI MSI was shown to be compatible with fingermark sequential processing. Compatibility and the ability to obtain additional ridge detail was demonstrated after enhancement of a "condom contaminated mark" via irradiation with a laser shone at a wavelength of 532 nm and subsequent ATR-FTIR analyses [23]. Groeneveld et al also demonstrated MALDI MSI compatibility with CAF-BY40, CAF-VMD and CAF- BY40-VMD [20].

Amongst the aforementioned MSI techniques, only MALDI and SIMS have been reported in the FVM (latest version published in 2014) as Category C techniques. Category C indicates processes "*at a developmental stage exhibiting potential [...] an optional process for occasional operational use […] when Category A processes have been exhausted*". MALDI MS is now in the process to be promoted to Category B indicating *an "Established process [...] likely to offer benefits [...] for occasional operational use [...] and when all Category A options have been exhausted.* Additionally, amongst all the MSI techniques, to the best of the authors' knowledge, MALDI has been the only one that has been deployed operationally in casework in the UK and overseas, a few examples of which are reported by Bradshaw et al [24].

Due to the imminent promotion to Category B, the extensive body of knowledge acquired on the fingermark imaging and profiling capabilities of this technique, the deployment in casework and the compatibility with a significant number of FET, it becomes important to assess the wider potential of MALDI to be used in sequential processing.

In this proof of principle study, one donor has been employed with no selection criteria and a total of 10 marks were analysed. MALDI MSI has been conducted after four FET applied in sequence on fingermarks deposited on either brown or clear tape. Here, not only is compatibility of MALDI MSI application after up to four FET processes sequentially applied to latent fingermarks demonstrated, but MALDI appears to also provide additional ride detail in areas that are undeveloped or overdeveloped following the application of FET; in essence, the study demonstrates the ability of

MALDI "to fill in the gaps" of developed fingermarks. This capability is due to the generation of multiple molecular images of fingermarks in a single analysis, exploiting the constituents that have not been targeted or have "survived" the prior application of conventional processes. This forensic opportunity is of considerable importance to maximise the recovery of identifying details, just as the conventional processes, sequentially applied, aim to do.

Materials and Methods

Materials

Acetonitrile (ACN) and acetone were obtained from Fisher Scientific (Loughborough, UK). Trifluoroacetic acid (TFA) and α-cyano-4-hydroxycinnamic acid (α-CHCA) were purchased from Sigma Aldrich (Poole, UK). The materials used for fingermark development; cyanoacrylate (fuming) (CAF), basic yellow 40 (BY40), black powder suspension (BPS) and Basic Violet 3 (BV3) were provided by West Yorkshire Police (WYP) (Wakefield, UK). Brown (parcel) tape was purchased from Sainsbury's (Sheffield, UK) whereas clear tape was provided by the Fingerprint Enhancement Laboratory (FEL), Yorkshire and Humber Regional Scientific Support Services (YHRSSS) (Wakefield, UK).

Instrumentation and parameters

Optical images of all fingermarks were taken using a Foster and Freeman video spectral comparator (VSC 4CX) under reflected white light. MALDI-MSI was conducted using a modified Applied Biosystems Q-Star Pulsar *i* hybrid quadrupole time-of-flight (QTOF) instrument, equipped with a Nd:YVO_4 solid-state laser operating at 5000 Hz. All images were acquired at a spatial resolution of 150 x 150 μm using a 'slow' raster mode with ‘oMALDI Server 5.1’ software, supplied by MDS Sciex (Concord, Ontario, Canada). This mode enabled each image to be acquired in around 60-90 min, depending on size (although with modern instrumentation, only 10 minutes would be required for a full fingermark). The mass range analysed was between *m/z* 50-1000. The declustering potential 2 was set at 15 and the focusing potential at 20.

Data processing

All MALDI MS images were processed using Biomap (Novartis, Basel). Images of the most abundant ions were selected by importing MSI data and setting the bin size at 1; the "plot point tool" has been used to select ion signals. In generating the corresponding molecular images, the signals were baseline corrected using "maximum with BC" prior to being normalised against the total ion current (TIC) in the *m/z* range 100-1000 Th for each fingermark sample. Fingermark quality was assessed and graded based on the grading scheme published by Bandey (2004) [25]. According to this scheme, grade zero describes a mark yielding no evidence; evidence of contact but no ridge detail is described as a grade 1 mark; a grade 2 and 3 are associated to marks with about 1/3 of ridge detail, that probably cannot be used for identification, and to an identifiable mark (between 1/3 and 2/3 of ridge detail) respectively. Grade 4 is assigned to an identifiable mark with full ridge detail.

Fingermark deposition and development

Natural latent fingermarks (e.g. marks without any prior fingertip preparation) were deposited directly onto brown parcel tape and clear tape at FEL (YHRSSS) by the same male donor (totalling 10 investigated fingermarks). All fingermarks were developed by FEL personnel using the protocols outlined in the 'Fingermark Visualisation Manual' [2]. A series of fingermarks were developed in

duplicate using a number of processes such as cyanoacrylate fuming (CAF), basic yellow-40 (BY40), Black Powder suspension (BPS), Gold/Zinc vacuum metal deposition (VMD) and Basic Violet 3 (BV3). These processes were combined into different sequences such as: no development, CAF, CAF→VMD, CAF→BY40, CAF→BY40→BPS, CAF→BY40→BPS→BV3. Ten different fingertips were used, one for each of the sequences investigated. All the fingerprints were deposited in the same day, one after the other. Developed fingermarks were stored at room temperature within a sealed container up until the point of analysis.

Matrix deposition

All fingermarks were secured directly onto a MALDI target plate with double sided carbon tape before being sprayed with 5 mg/mL α-CHCA in 70:30 ACN:0.5% TFA using a SunCollect sample preparation device or 'auto-sprayer' (SunchromGmbH, Friedrichsdorf, Germany). A total of 4 layers were deposited onto each sample using a flow rate of 2 µL/min and a 'slow' raster setting. Fingermarks were immediately subjected to MALDI MSI analyses following matrix deposition.

Results and Discussion

Brown parcel tape and clear tapes are common surfaces of deposition on which marks are recovered at crime scenes; for example; in more violent cases where a victim might be bound or have their airways covered or in instances when it has been used on evidential items (such as letters). In this study, latent natural fingermarks were placed on either of the two tapes (adhesive side) and sequential processing was applied (three different sequences). At the end of each development process, MALDI MSI was performed to evaluate its compatibility at each stage of the sequential processing. This strategy also enabled investigation into possible incompatibility with any individual developing agent to enable the correct placing of MALDI MSI analysis within the operational workflow. The first FET of any of the sequences applied on clear and brown tape was CAF. In an initial investigation, CAF was followed by MALDI MSI, confirming prior studies demonstrating compatibility of MALDI MSI with this process (Fig 1).

CAF of a fingermark deposited on clear tape enabled visualisation of ridge detail with Grade 2 quality (according to the grading scheme of Bandey et al [25]) (Fig 1 (i)). For the latent mark deposited on brown tape, CAF development yielded a grade 4 mark as shown in Fig 1(iii). Analysis of the CAF developed marks by MALDI MSI revealed hundreds of ions enabling molecular images of the ridge pattern. Putatively identified lipids (fatty acids, diacylglycerols and triacylglycerols) provided generally the highest quality images. For the mark deposited onto clear tape, the selection of a species at *m/z* 230.2 (which was previously confirmed to be the endogenous 13-aminotridecanoic acid [26]), produced a grade 4 fingermark image (Fig 1 (ii)). For the mark deposited onto brown parcel tape, an ion at *m/z* 893.0 provided the most ridge coverage of the portion of the mark imaged and highlighted by the frame in Fig 1 (iv). It is important to note that the CAF developed mark on brown parcel tape shown in the optical image of Fig 1(iii) appears sharper than the image of the ion at *m/z* 893.0. However, higher resolution mass spectrometric imaging is available in advanced instrumentation and would provide a much "sharper" and "detailed" image. Notwithstanding even with the ‘low’ spatial resolution, the ridge flow appears clearer and to provide more contrast in the upper part of the mark and in places around its centre in the MALDI MS image over the optical image. The increased contrast may be beneficial particularly in the presence of surfaces with patterned/complex backgrounds as shown by Scotcher and Bradshaw [27].

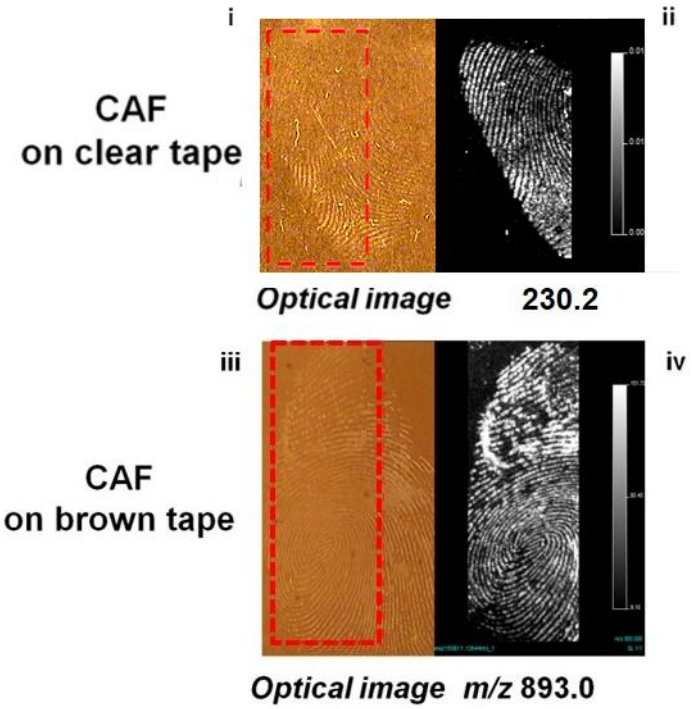

Fig 1. Application of MALDI MSI following CAF on a latent fingermark on clear and brown tape. The highest quality fingermark image was yielded through selection of ions at *m/z* 230.2 and *m/z* 893.0 for clear and brown tape, respectively.

"Dual" sequential processing of latent marks on clear tape and parcel (brown) tape

The sequential application of CAF and VMD followed by MALDI MSI was already investigated by Groeneveld et al [20]. However, this was in the context of detecting and mapping drugs of abuse within contaminated marks after being deposited on an ideal surface for MALDI, namely aluminium sheets. Therefore, the sequence CAF→VMD was investigated again and, this time, on both clear tape and brown tape. Similarly to the results obtained from the application of CAF alone, the CAF-VMD workflow yielded a grade 2 quality from a fingermark deposited onto clear tape (Fig 2 (i)). However, subsequent MALDI MSI analysis provided significantly more ridge detail (Grade 4) through the molecular image of benzalkonium chloride at *m/z* 304.2 (Fig 2 (ii)) firstly detected as an exogenous contaminant in fingermarks by Bradshaw et al [28]. Interestingly, CAF-VMD development of a mark deposited onto brown tape yielded a grade 3 fingermark image, with some regions of the mark that were either over or underdeveloped (Fig 2 (iii)); the corresponding MALDI image for an ion at *m/z* 240.2 (Fig 2 (iv) showed additional ridge flow and detail resolving some of the underdeveloped and overdeveloped areas.

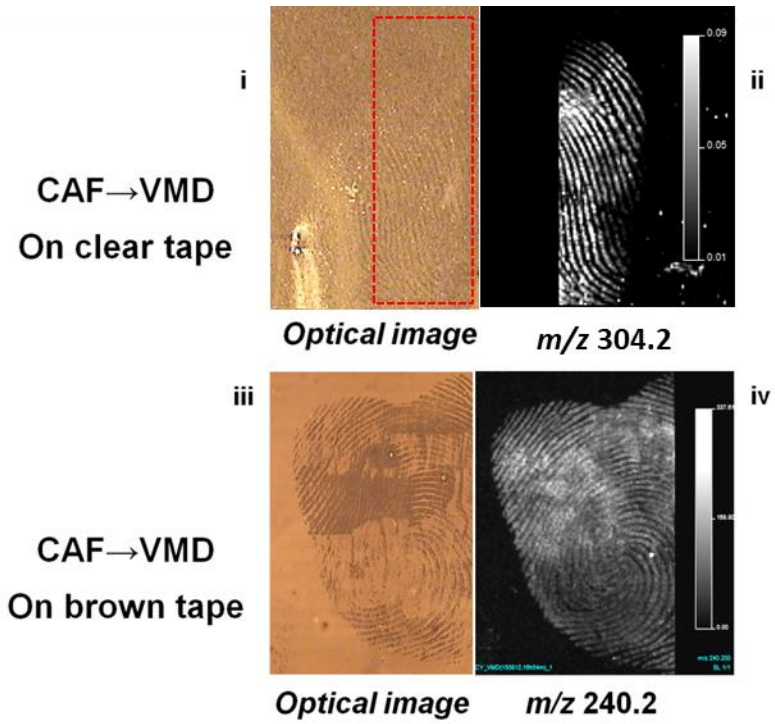

Fig 2. Application of MALDI MSI following CAF→VMD of a latent fingermark on clear and brown tape. The highest quality fingermark image was yielded by selection of ions at *m/z* 304.2 and *m/z* 240.2 for clear and brown tape, respectively.

Multi-step sequential processing of latent marks on clear tape

In cases where CAF developed marks require further enhancement, a dye such as BY40 is subsequently used. Therefore, another latent mark was deposited and developed sequentially by CAF→BY40 (Fig 3 (i)). It was found that viewing the developed mark by illumination at 365 nm improved the visualisation of ridge detail. However, as the marks were shipped to the mass spectrometric analysis location where no specialist light facilities were available, the mark could not undergo fluorescence examination using the excitation and emission wavelengths recommended by the FVM [2]. Subsequent MALDI MSI analysis did not allow for additional ridge detail to be acquired when selecting an unidentified species (*m/z* 643.8 (ii)), with the optical image appearing to show greater ridge coverage. The weak signal at *m/z* 643.8 only indicated the general region in which a mark was deposited (Grade 1). Indeed, the quality of the MALDI MS images could be due to the ionisation suppression effects of BY40 which have been observed previously [20].

Powder suspension (PS) may be used if the crime lab establishes the need to further enhance the mark. Therefore, MALDI MSI was performed after the sequential processing CAF→ BY 40→PS applied to an additional latent mark (Fig 3 (iv)). In this case, the same ion employed for MALDI MSI visualisation of the CAF→BY40 developed mark (*m/z* 643.8) provided the best ridge reconstruction, though predominantly in the top third of the image, with a grade 1 image being obtained.

A fourth FET was then added to the sequential processing, namely Basic Violet 3 (BV3) and the 4 processes were applied in sequence to another natural mark deposited on clear tape (Fig 3 (v)). MALDI MSI provided a fingermark with grade 3 ridge detail through molecular imaging of 13-aminotridecanoic acid (*m/z* 230.2) (Fig 3 (vi)), albeit possibly of overall lower quality than the corresponding optical image. The superiority of this fingermark quality over the two from the earlier steps in the workflow has been attributed to the quality of the fingermarks which were deposited in

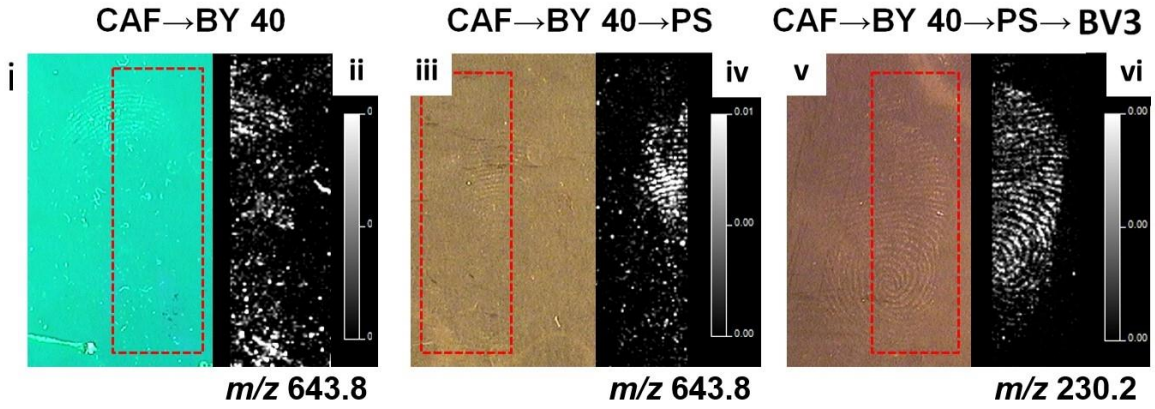

Fig 3. Application of MALDI MSI following sequential processing of a latent fingermark on clear tape. Panels i, iii and v indicate optical images of the mark after CAF→ BY 40, CAF→ BY 40→PS and CAF→ BY 40→PS→BV3 respectively. Panels ii, iv and vi indicate the corresponding MALDI MS images showing the most ridge flow/detail.

these examples. As natural fingermarks were employed throughout, there will be more variability in the molecular content of the fingermark residue, and henceforth the quality of the fingermark available for development.

Multi-step sequential processing of latent marks on parcel (brown) tape

Similarly to the optical images of marks developed on clear tape, best visualisation of ridge detail following the CAF→BY40 workflow was observed at 365 nm. The highest quality MALDI MS image (in terms of ridge detail) was obtained through the ion at *m/z* 326.2 (Fig 4 (iv)) which was previously identified as didecyl dimethyl ammonium ion (DDDMA) [19]. DDDMA was also confirmed in this study through MS/MS analysis (data not shown), though it is important to note that confirming the identity of these species was not within the aims of this study. Therefore, as also previously observed, MALDI MSI was compatible with the sequential process CAF→BY40, although in this case too, higher resolution imaging would have been desirable.

MALDI MSI analysis of a fingermark subjected to the CAF→ BY 40→PS workflow provided the most striking example of MALDI MSI ability to "fill in the gaps" as already shown in the CAF→ VMD sequence example on brown tape; whilst the sequential processing only yields a partial mark with poor ridge continuity, MALDI MSI yields a grade 4 image through the ion at *m/z* 362.0 and additional ridge detail, particularly in the area highlighted by the green frame (Fig 4 iv). This opportunity is due to the detection of hundreds to thousands of ions including those that the sequential processes may not target. Therefore, not only was MALDI MSI compatible with three fingermark enhancement processes applied in sequence but it also provided additional/complementary ridge detail.

MALDI MSI was shown to be compatible even after the application of 4 FET, that is, after the sequential processing CAF→ BY 40→PS→BV3 (Fig 4 v). Although in this instance too, higher resolution mass spectrometric imaging would have been desirable, MALDI MSI was able to "fill in the gaps" through the ion at *m/z* 559.0 as Fig 4(vi) shows.

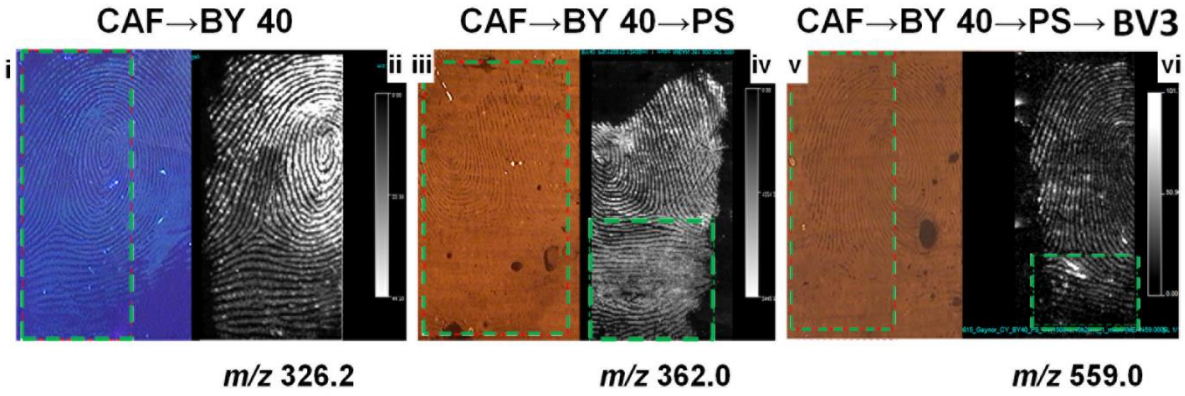

Fig 4. Application of MALDI MSI following sequential processing of a latent fingermark on brown tape. Panels i, iii and v indicate optical images of the mark after CAF→ BY 40, CAF→ BY 40→PS and CAF→ BY 40→PS→BV3 respectively. Panels ii, iv and vi indicate the corresponding MALDI MS images showing the most ridge flowing/detail.

For the sequence CAF→ BY 40, it is interesting to observe the possibility to combine two (or more) mass images from ions localised in different areas of the mark to maximise ridge pattern coverage. This opportunity was previously highlighted in undeveloped ungroomed marks [7] and it is shown for natural marks developed by two processes in sequence (Figure 5).

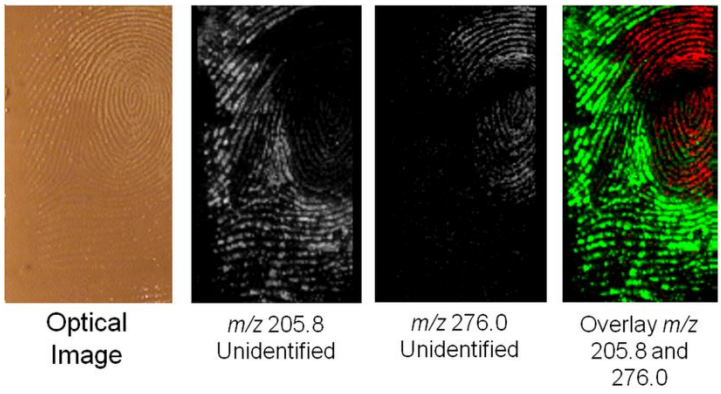

Fig 5. CAF→ BY 40→ MALDI MSI sequential processing of a natural mark on brown tape. The mass images for the unidentified ions at *m/z* 205.8 and 276.0 are shown as well as their superimposition maximising ridge pattern coverage.

Overall, MALDI MSI has been shown to be compatible with a number of sequential processes providing, in some cases, additional ridge detail, although image contrast and ridge pattern coverage varied across the two tapes examined, with brown tape yielding the highest grade fingermark images compared to clear tape.

Though the "ground truth" around the molecular composition of all the marks analysed is not known, this study does demonstrate a real potential for implementation of MALDI within the fingerprinting workflow, at every stage of the sequential processing investigated, thanks to the use of natural fingermarks, that is, fingermarks generated with no prior preparation of the fingertip prior to touching the surface. This is in contrast with many other studies using only ungroomed or groomed marks

(obtained through washing hands or enriching the fingertips by contact with face and scalp typically, respectively [26, 29-30]). This experimental choice is in line with the recommendations of the International Fingermark Research Group [31] whereby conclusions on performance of new visualisation reagents/techniques "*should be drawn – where possible – from natural fingermark sets, not from groomed samples or standard solutions*".

Conclusions

For an emerging technique to become operational within the fingerprinting workflow, compatibility with one and multiple fingermark enhancement techniques (FET) must be proven. MALDI MSI is one of the mass spectrometric imaging techniques most compatible with individual FET though only a few instances of application after more than one FET had been investigated.
In this work MALDI MSI has been successfully applied to latent natural marks on two tapes and following up to four FET applied in sequence, with the highest quality fingermark molecular images being obtained when brown tape was used as the surface of deposition. The results also confirm prior observations that the enhancement processes do not deplete all the fingermark molecular material which then becomes available for MALDI MSI analysis. As with other mass spectrometric imaging techniques, MALDI MSI enables the acquisition of hundreds to thousands of ions in a single analysis, thus it can work in an untargeted manner. However, during processing and generation of the images, molecules that have not been targeted by the enhancement technique can be used to generate images. For example, following the application of ninhydrin, molecules other than amino acids can be used to generate images. In addition, due to the non exhaustive molecular depletion observed for many processes, MALDI MSI is also able to generate images from the class of molecules targeted by the development process. An example of the latter instance is provided by the possibility to generate lipid images after VMD (targeting monolayers of fats in fingermark deposits) is applied to fingermarks, as previously shown. Additional work is needed to further demonstrate compatibility of MALDI with sequential processing; this includes the investigations of multiple donors of both sexes, a greater number of surfaces of deposition and marks of different age.

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