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# Biodegradable Plastics and Their Impact on Fingermark Detection Methods

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| **Abstract:** | The use of plastics is extremely prevalent in society, with most individuals likely to handle several plastic items per day. It is therefore not surprising that many exhibits recovered from the scene of a crime are plastics, which are processed and examined for traces such as fingermarks. Societal trends have been pushing towards more environmentally friendly products with alternatives to traditional disposable plastics becoming increasingly available. These alternate plastics have different chemical compositions and physical properties, which may impact fingermark development for these substrates. As most detection techniques are known to be substrate-dependent, it is crucial to review current methods and procedures to examine how effective they are on new materials.<br>The aim of this research was to assess a range of fingermark detection techniques on biodegradable plastics and provide recommendations for the preferred technique. First, the prevalence of these materials in the Australian market was evaluated. Over 40 different plastics obtained within the Sydney area were then divided into six broad categories using consumer information in combination with ATR-FTIR spectroscopy analysis. Following this, selected plastics from each category were used as substrates for the fingermark development study. In total, 6480 fingermark specimens were collected as split marks, to form 2160 fingermark comparisons. Each substrate was then developed with four fingermark detection techniques suitable for plastic substrates: cyanoacrylate (CA) fuming, vacuum metal deposition (VMD), powder suspensions (PS), and single metal deposition (SMD).<br>SMD resulted in the most consistent development method across all tested substrates. VMD was able to successfully develop fingermarks on polyethylene-based plastics, but led to poorer results on alternative plastics, while CA fuming and PS were notably more dependent on the surface texture.<br>This research was successful in confirming that biodegradable plastics do in fact have an impact on fingermark development techniques commonly applied on traditional plastics and recommendations have been formed to aid in operational contexts to improve the potential to recover latent fingermarks from biodegradable plastics. |
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*Proposition of: Original Research Article*

# Biodegradable Plastics and Their Impact on Fingermark Detection Methods

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# Highlights

- Biodegradable alternatives to traditional plastics are becoming more prevalent
- New materials create a gap in fingermark development research
- Conventional methods shown to be less effective on new plastics
- Single metal deposition is the preferred development method

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## Abstract:

The use of plastics is extremely prevalent in society, with most individuals likely to handle several plastic items per day. It is therefore not surprising that many exhibits recovered from the scene of a crime are plastics, which are processed and examined for traces such as fingermarks. Societal trends have been pushing towards more environmentally friendly products with alternatives to traditional disposable plastics becoming increasingly available. These alternate plastics have different chemical compositions and physical properties, which may impact fingermark development for these substrates. As most detection techniques are known to be substrate-dependent, it is crucial to review current methods and procedures to examine how effective they are on new materials.

The aim of this research was to assess a range of fingermark detection techniques on biodegradable plastics and provide recommendations for the preferred technique. First, the prevalence of these materials in the Australian market was evaluated. Over 40 different plastics obtained within the Sydney area were then divided into six broad categories using consumer information in combination with ATR-FTIR spectroscopy analysis. Following this, selected plastics from each category were used as substrates for the fingermark development study. In total, 6480 fingermark specimens were collected as split marks, to form 2160 fingermark comparisons. Each substrate was then developed with four fingermark detection techniques suitable for plastic substrates: cyanoacrylate (CA) fuming, vacuum metal deposition (VMD), powder suspensions (PS), and single metal deposition (SMD).

SMD resulted in the most consistent development method across all tested substrates. VMD was able to successfully develop fingermarks on polyethylene-based plastics, but led to poorer results on alternative plastics, while CA fuming and PS were notably more dependent on the surface texture.

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This research was successful in confirming that biodegradable plastics do in fact have an impact on fingermark development techniques commonly applied on traditional plastics and recommendations have been formed to aid in operational contexts to improve the potential to recover latent fingermarks from biodegradable plastics.


## Keywords

Compostable plastic, cyanoacrylate, powder suspension, vacuum metal deposition, single metal deposition, technique efficiency.


## Highlights

- Biodegradable alternatives to traditional plastics are becoming more prevalent
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## Introduction

Traditionally plastics derive from petrochemicals, and are often combined with functional additives such as plasticisers to increase flexibility and durability, as well as antioxidants to stabilise the polymer when exposed to light and heat, making the plastic less susceptible to degradation [1, 2]. However, with increasing global production of plastics [3] these long lasting and durable qualities have created the issue of managing pollution from the excess waste of disposable, single-use plastics [4].

To address this problem, society has trended towards an uptake of more environmentally friendly alternatives such as newly developed biodegradable plastics to lessen the impact on our environment. However, from a forensic context, new materials create a void in knowledge for the potential impact on fingermark development. It is well known that the substrate plays a major role in fingermark development, with variations in method required for samples within the same broad category of base-polymers [5]. With new environmentally friendly plastics becoming more common, it is crucial to research the impact these materials have on fingermark development to ensure optimal processing and reduce the risk of not detecting or losing valuable forensic traces.

Plastic terminology and the meaning of biodegradability is an area for debate, with terms such as biodegradable, compostable, bio-based being used interchangeably. With region dependant standards for biodegradability, the term used in Australia refers to anything designed to break down under certain conditions of light, heat, and oxygen exposure within a year according AS 4736 [6]. The term "compostable plastics" refers to polymers which can be broken down and decay to produce water, carbon dioxide, inorganic compounds, and biomass when consumed by organisms such as worms, while importantly having no toxic impact [7]. "Bio-based" refers to polymers, which are produced primarily from organic materials, but the resulting polymer may have no enhanced ability to degrade compared to a traditional plastic. All these plastics are considered as "alternative plastics" to traditional petrochemical-based ones, and as such are of value for this study. The plastics highlighted in green, or blue in **Figure 1** are of interest for further assessment – regardless of their ability to degrade [8].

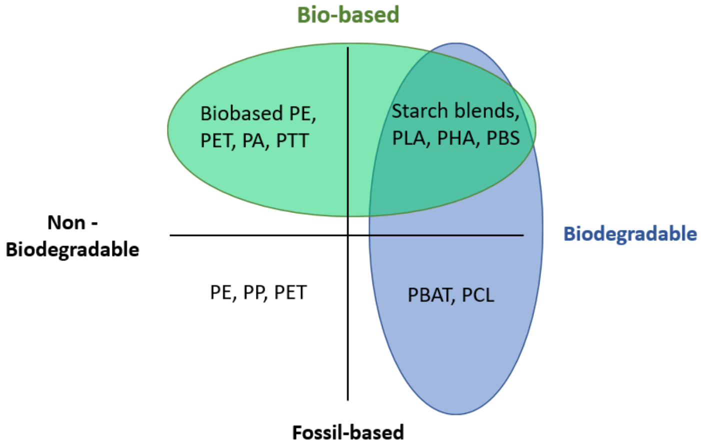


***Figure 1**: Common Plastics by Type, adapted from Australasian Bioplastics Association 2019 [8], full list of abbreviations can be found in table 1.*

**Table 1** lists the abbreviations of each polymer referred to from **Figure 1**. The most common plastic manufactured is Polyethylene (PE). It exists in a high- and low-density form and can be made into rigid or flexible films. Polypropylene (PP) and Polyethylene Terephthalate (PET) are common plastics, although their physical and chemical structures are different, each of these traditional plastics can be extremely visually similar [9].

Biobased variations of these traditional polymers are also becoming more prevalent. The most common is biobased PE (Bio-PE). Where traditional PE is produced using ethylene sourced from non-renewable petrochemicals, Bio-PE exists with identical physical properties to traditional PE. The ethylene required for manufacture is instead sourced by dehydration of ethanol from sugar cane [10]. With an indistinguishable chemical structure, this biobased alternative shares an endless list of uses, but like traditional PE is not degradable.

Some petrochemical-based polymers are degradable. The most common of which is Polybutylene Adipate Terephthalate (PBAT), which is highly degradable and can be used to produce flexible films, but due to its chemical structure is not suitable for making rigid containers or bottles [11]. Instead, it is often blended with other more suitable materials to enhance the overall degradation.

The area of most interest for this study are the biobased biodegradables highlighted in green and blue (**Figure 1**). The most common polymer manufactured from this category is Polylactic Acid (PLA). As it is highly transparent, strong, and can be made into rigid or flexible sheets, PLA is often considered as the most suitable replacement for PE and PET. In flexible form, it makes packaging and films, while rigid PLA is used for disposable containers, cups, and utensils. It also highly used as a 3D printing filament [12].

**Table 1**: Abbreviations of common types of polymers

| | |
|---|---|
| **PE** | Polyethylene |
| **PP** | Polypropylene |
| **PET** | Polyethylene Terephthalate |
| **PA** | Polyamide (nylon) |
| **PTT** | Polytrimethylene Terephthalate |
| **PLA** | Polylactic Acid |
| **PHA** | Polyhydroxyalanoate |
| **PBS** | Polybutylene Succinate |
| **PBAT** | Polybutylene Adipate Terephthalate |
| **PCL** | Polycaprolactone |

The surface of a substrate has been shown in previous studies to be a significant contributing factor in fingermark development quality [13, 14], however such studies have often been limited to a single development method such as powder suspensions. When introduced to new materials with different surface characteristics and chemical compositions, it is important to perform a comparative study of the most common development methods to explore if and how these materials impact the successful development of fingermarks.

Although biodegradable polymers have been produced for several decades it is only recently that they have become more widely available and commonly encountered, and as such can be considered a novel material in fingermark detection research. Two recent publications were the first to specifically investigate these materials, and both are important first steps in the exploration of eco-friendly materials.

Zampa *et al* [15] performed a collaborative exercise exploring a particular biodegradable brand of plastic (Mater-Bi®) with 40 laboratories taking part to develop pre-deposited fingermarks using their own processes. It was found that multi metal deposition was the more effective and consistent technique, but an important note was made that other biodegradable materials may behave very differently, while this study was limited to one particular type. Illston-Baggs *et al* [16] in a different study suggested that the current recommended sequence for development for soft plastic packaging was able to develop the most fingermarks compared to three other methods [17]. However, this study also identified significant differences between substrates, aging periods, and donor performance, it was identified that considerably more research was needed.

The aim of this study was to explore the impact of these materials on fingermark detection specifically within an Australian context to make recommendations for the preferred development method. Where the previous research was limited to materials and methods recommended in United Kingdom, there is an obvious need to further test local materials and consider additional detection methods. By performing a comparative assessment of each method, where all techniques are compared to each other allowing for a direct representation of the impact and relationship between substrate and methods of fingermark detection.

# Materials and Methods

The study compared the impact of different biodegradable plastic types on fingermark development quality with four different methods. Cyanoacrylate fuming (CA), powder suspensions (PS), and vacuum metal deposition (VMD) were chosen as more commonly employed operational techniques [17], while single metal deposition (SMD) [18] was chosen as an alternative technique which has shown to be effective on non-porous substrates [19]. Each of these fingermark detection techniques were compared directly to each other across split fingermarks over several substrates, while also monitoring for variations in donor, depletions, and age of the fingermarks since deposition. The collection of latent fingermarks were performed in accordance with the guidelines published by the International Fingerprint Research Group (IFRG) [20].

## Substrates

Plastic samples were obtained from the local Sydney area with consideration for easily available samples such as shopping bags, vegetable grocery bags, dog waste bags, and take-away food and drink containers, as well as those available for purchase from retail and commercial suppliers.

A total of 42 different plastic samples were obtained, 17 of which were excluded as being readily identified as non-biodegradable traditional plastics such as PE or PET. Any plastic that was of unknown composition or was indicated as being of an environmentally friendly alternative was then analysed further. A Nicolet™ FTIR (Fourier transform infrared) 6700 spectrometer with ATR (attenuated total reflectance) attachment (diamond crystal) was used for further analysis. Each plastic had three small sections removed from different areas to be processed with the ATR-FTIR, forming a transmittance spectrum. These spectra were then combined to form an average using the software Omnic™ by Thermo Fisher Scientific™.

Following analysis of the FTIR spectra in combination with manufacture information, six categories were formed, with the total number of each plastic provided in the **Table 2**.

***Table 2***: *Type of plastics obtained with total count value*

| TYPE | ABREVIATED | COUNT |
|---|---|---|
| Polyethylene | PE | 3 |
| Polyethylene with degradable additives | PE+ | 7 |
| Bio-polyethylene | Bio-PE | 3 |
| Starch based compostable | Starch | 6 |
| Rigid polylactic acid | PLA | 3 |
| Mix blends containing starch, PLA, and PBAT | Mix Blend | 3 |
| | **Total** | **25** |

Provisional tests were undertaken to ensure consistent fingermark development quality within plastics of the same category, before choosing a single sample to be representative of the broader category.

In choosing the final substrates, considerations were made for colour contrast, thickness, availability, as well as including a rigid PLA sample to contrast the flexible polymers. This ensured a broad range of qualities were considered for comparison. The final plastic chosen of each category is represented in **Table 3**.

***Table 3***: *Final selection of substrates for comparison stage*

| Brand | Type | Colour | Manufacturer claim | Category |
|---|---|---|---|---|
| Coles | Rubbish bag | White | n/a | PE |
| Sugar Wrap | Rubbish bag | White | 100% plant based | Bio-PE |
| Bio-Gone | Resealable bag | Transparent | Landfill biodegradable | LD-PE with degradable additive |
| Maze | Compost bag | Green | 100% compostable vegetable material | Compostable starch |
| Better Packaging Co | Postage satchel | Black | Home compostable | Starch / PLA / PBAT blend |
| Vegware | Hard container | Transparent | Commercially compostable | PLA |

[Type here]

## Fingermark Deposition

Fingermarks were deposited in sets of three depletions and were then treated as split marks where each sample was cut in half and developed using a different technique on each side. For example, CA fuming on the left, and PS on the right. This allowed for direct comparison of the techniques from the same fingermark deposition. This was repeated between each detection method forming six sets (**Table 4**).

***Table 4**: List of direct comparisons between all development methods, where method A and B were used on either side of each sample that was cut in half.*

| Development Method A | Development Method B |
| --- | --- |
| CA | PS |
| CA | VMD |
| CA | SMD |
| PS | VMD |
| PS | SMD |
| VMD | SMD |

Fingermarks were donated by five individuals, three female, and two male. Hands were required to be washed and dried five minutes prior to deposition. The fingermarks were deposited naturally and were not deliberately enriched in anyway. Each donor deposited using the three middle fingers in a sequence of three depletions, using either hand, then waited three minutes between additional depositions. No instruction was given in regards deposition pressure. Each sample was aged 1 day, 1 week, 2 weeks, or 4 weeks before development. After deposition, the samples were pinned to cork boards and kept out of direct sun in a laboratory environment with mean temperature of 19.5 ± 1 °C and mean relative humidity of 54.3 ± 15%. In total, each donor deposited nine individual marks for a single sample, in sets of three depletions per sample. With six direct comparisons of methods, across six different substrates, each of which being aged four different times, making a total of 1296 fingermarks per donor. With five donors this was 6480 individual fingermarks deposited.

## Development methods

As the focus of the research was to assess the impact that biodegradable materials have on fingermark detection methods, the following four detection techniques were completed with direct comparisons to each other, and no sequential techniques applied. Rather it was the single methods only that were compared, without subsequent enhancement by any other method. Although further enhancement may have improved the fingermark quality in some cases, it was beyond the scope of this research, and subsequent work may be conducted to examine the preferred methods in sequence.

### Cyanoacrylate Fuming

A MVC 1000 Cyanoacrylate Fuming Cabinet (Foster + Freeman) was used for CA fuming, with the following parameters chosen for fingermark development. 0.5 g of Loctite™ 406 super glue, with the chamber set to fume for 20 minutes at 120°C, at 80% relative humidity. No more than 20 samples were processed in a single run, and all samples were hung vertically from the top rail, as it was noted in the trial period that using the lower rail or laying samples flat resulted in inconsistent fingermark development. Samples from each category of plastic were processed at the same time, as variations in the parameters chosen did not appear to favour any sample.

### Powder Suspensions

Optimum Technology™ supplied WetPowder™ by Kjell Carlsson Innovation™, in both a carbon-based black powder suspension, and a titanium dioxide white powder suspension. These solutions were used as supplied.

As most of the chosen substrates were a light colour, the carbon-based solution was appropriate for use in all cases except for the black postage satchel, where the white PS was used to maximise contrast.

The most optimal application method was found to be using a squirrel hair fingermark brush (also supplied by Optimum Technology™). This was applied onto the substrate and left for 10-30 seconds before rinsing with water under a tap. Each sample was then pinned to a board and allowed to dry before imaging.

### Vacuum Metal Deposition

A VMD360 vacuum metal deposition chamber (West Technology Forensics®) was used, with consumable 0.15 g zinc pellets and 0.25 mm gold wire in 4mm lengths supplied Ezzi Vision™. The recommended procedure was followed as listed in the operating instructions [21]. The chamber was pumped until a vacuum pressure of 2.0 x $10^{-4}$ mBar was reached, before sequentially evaporating two pieces of the gold wire then one zinc pellet. The zinc evaporation was not fully completed but was stopped once visually satisfactory results were achieved. This was determined using two test pieces with charged fingermarks placed on either side of the chamber, which required slightly less zinc to fully develop, and as such was an indicator of when the process was almost complete. Different plastics were able to be processed at the same time provided enough gold was placed in the chamber for evaporation. Only a single length of gold wire was necessary for the polyethylene-based plastics, but doubling the wire was required for the other samples. The excess gold had no observable negative impact on the polyethylene plastics. The zinc evaporation required in this case was observed to be consistent for each of the materials chosen, allowing a mixture of samples to be processed at one time. It is important to note that this is not always the case when processing different materials with VMD, and tests should be carried out prior to processing different materials at the same time to determine the required metal evaporation. A maximum of 18 samples were processed at one time in the chamber.

## Single Metal Deposition

The SMD II protocol proposed by Moret and Bécue in 2015 [18] was followed without variation. Gold (III) chloride trihydrate, sodium citrate, sodium hydroxide, Tween®20 (Sigma-Aldrich), citric acid monohydrate (Chem-Supply), L-aspartic acid (Tokyo Chemical Industry), hydroxylamine hydrochloride (Acros Organics), were all of high purity and were used without further purification. Reverse osmosis deionised water (18.2 Ω·cm) was used in the preparation of colloidal gold solution, with deionised water used in the rinsing baths.

Contamination of dirty glassware was observed to negatively impact the development process, so care was taken to thoroughly wash equipment before use. Care was also taken to monitor the agitation process as floating plastic samples were inclined to stick together which also prevented successful fingermark development. This was largely avoided by reducing the number of samples to no more than 24 per run. Each sample was then pinned to a board and allowed to dry before imaging.

## Imaging Methods

After each half sample was developed, the plastics were paired again for imaging. This was completed with a Canon 750D DSLR, fitted with a Canon EF-S 60mm macro lens. Each sample was carefully aligned prior to capture. Some samples were not perfectly flat after exposure to water, and some were more reflective than others. To account for this a combination of overhead fluorescent lighting was used in combination with a Rofin Polilight PL500 to achieve optimal photography of each fingermark.

## Fingermark Analysis

The University of Canberra (UC) scale [22] was used to assess the comparative quality of the developed fingermarks. This was slightly modified to include a 00 score for cases of no fingermark detection [23] on either side as seen below in **Table 5**. If the development method used on the left-hand side of the split mark was more effective, it was given a negative score. If the right-hand side was more effective, it was given a positive score. With a score of 0 used in cases of equivalent development.

***Table 5:*** *Modified comparative UC scale including 00 score [24]*

| Score | Definition |
|---|---|
| **-2** | Development of technique A is substantially less effective compared to technique B |
| **-1** | Development of technique A is slightly less effective compared to technique B |
| **0** | Both methods are indistinguishable in quality |
| **+1** | Development on technique A is slightly more effective compared to technique B |
| **+2** | Development on technique A is substantially more effective compared to technique B |
| **00** | No detection on either half of the fingermark sample |

Three independent assessors rated each of the 2160 images in a random order using this scale in **Table 5**, with the median of their scores being taken as the result. To minimise errors the variance between assessors was checked and investigated further if the results appeared highly inconsistent. Of the 2160 assessments there were 35 cases of notable inconsistencies that were reviewed and corrected. These cases appeared to be the result of human error and not genuine disagreements. From these results, trends were able to be observed between the variables.

Following analysis, to aid in interpretation of the data, the following classifications were applied to produce the graphs. Depending on the comparison, scores were combined to give an indication of the preferred method for that particular comparison. For instance, in a comparison between CA (technique A) and VMD (technique B), any comparison that was a positive value (+1 or +2) was classified to be CA preferred, indicating that CA was the preferred method of development. Similarly, any comparison that was a negative value (-1 or -2) would be classified as VMD preferred indicated that VMD was the preferred method of development. Through this classification of preferred method, it allowed for a clearer understanding of the impact the substrate was having on development, this has been applied to all graphs in this manuscript.

# Results

## General Results

Although this research can be considered as preliminary, with a total of 2160 comparisons made some meaningful trends can be drawn. While the individual donor, the age of the fingermark, and the depletion number of each mark was monitored throughout, the most important variables were the substrates and the effectiveness of development methods. These are discussed individually below for clarity.

It is important to note that the individual quality of fingermarks was not monitored as only a comparative scale was used, the cases of no detection were tracked with a total of 7.5%. Almost half of these no detections can be attributed to a single donor. Overall, in the vast majority of comparisons some ridge detail was able to be developed successfully on at least one side of the comparison.

## Cyanoacrylate

As CA fuming is one of the most common techniques for fingermark detection the results of these comparisons are quite interesting. The general performance of each method across all substrates compared to CA fuming is displayed in **Figure 2**, where better performance by CA is indicated in dark blue, while the alternative method is indicated in orange for PS, green for SMD or pale blue for VMD. Zero values suggest an equivalent development between both methods shown in yellow and grey indicates cases where no ridge detail was detected. VMD was the preferred development in only 2% of cases, however when compared to PS and SMD, CA is quite evidently outperformed. Considering the CA-SMD comparison, and including equivalent 0 scores, SMD was at least as effective or better than CA fuming in 87% of cases. Although this is clearly a substantial preference for SMD, it is important to note that dye staining and sequential developments were not applied which may have improved the performance of each method in some cases. In general, however, the poor results of CA fuming were not an issue of visualisation but were due to poor development overall. In these cases, dye staining would not have had any major effect as there was little ridge detail to enhance.

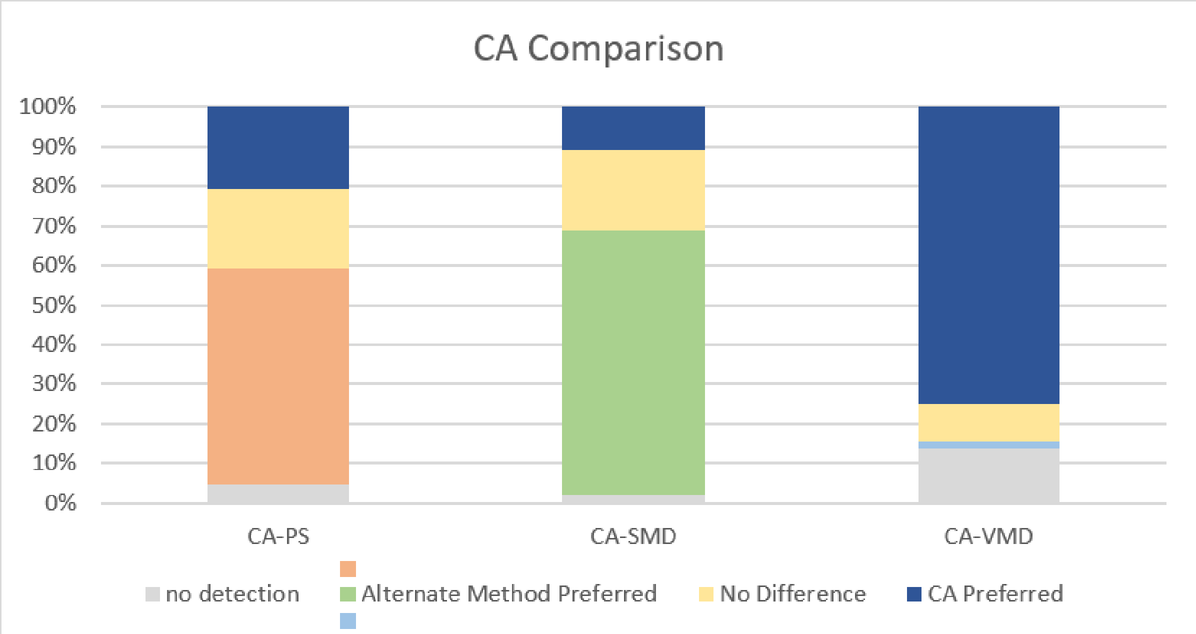


***Figure 2**: Comparison of CA vs all other development methods (orange for PS, green for SMD or pale blue for VMD)*

The following images are representative examples of the results obtained. With **Figure 3** indicating the comparisons with CA and each other method, across a range of the polyethylene-based substrates.

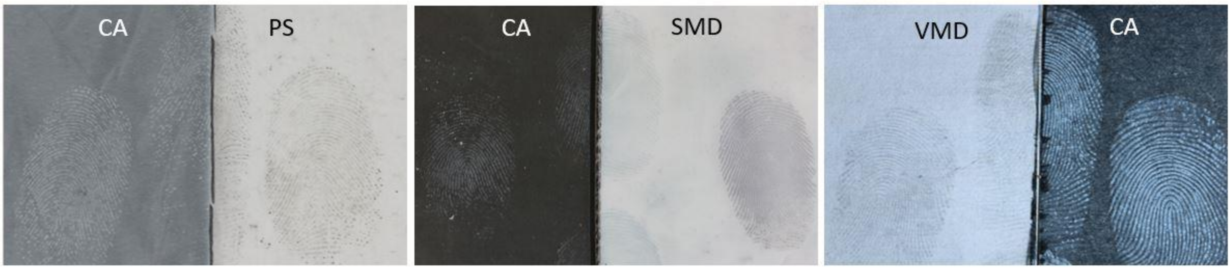


***Figure 3:** Left – CA vs PS on PE. Middle – CA vs SMD on BioPE. Right – VMD vs CA on degradable PE+. Images taken from 1 week aged samples.*

These results can then be expanded to examine the impact of the substrate on the detection method. The most evident variance between substrates appears with the PE+ (with degradable additives) and starch-based samples. PE+ is an exception to the previous observations where PS and SMD were favoured, particularly with PS, which was largely ineffective on these plastic samples, as seen in **Figure 4.** To a lesser extent the PLA samples follow this trend with more equivalent results when compared to PS**.**

Of most importance is the comparison on starch-based plastics as seen in column four of **Figure 4, 5, and 6**. For this surface, CA fuming resulted in ridge development in very few cases regardless of the parameters used. This unexpected result is concerning as CA fuming is often the primary method of fingermark processing for plastic surfaces, not only this, but on regular PE surfaces it appears that CA is only preferred to VMD, so may need to be revaluated as the technique of choice. If a starch-based sample is processed with CA the underlying fingermark may at bet escape detection, or at worse may be irreversibly damaged – although further research is necessary to experiment with sequential developments to explore the possibility of success if a subsequent technique is applied.

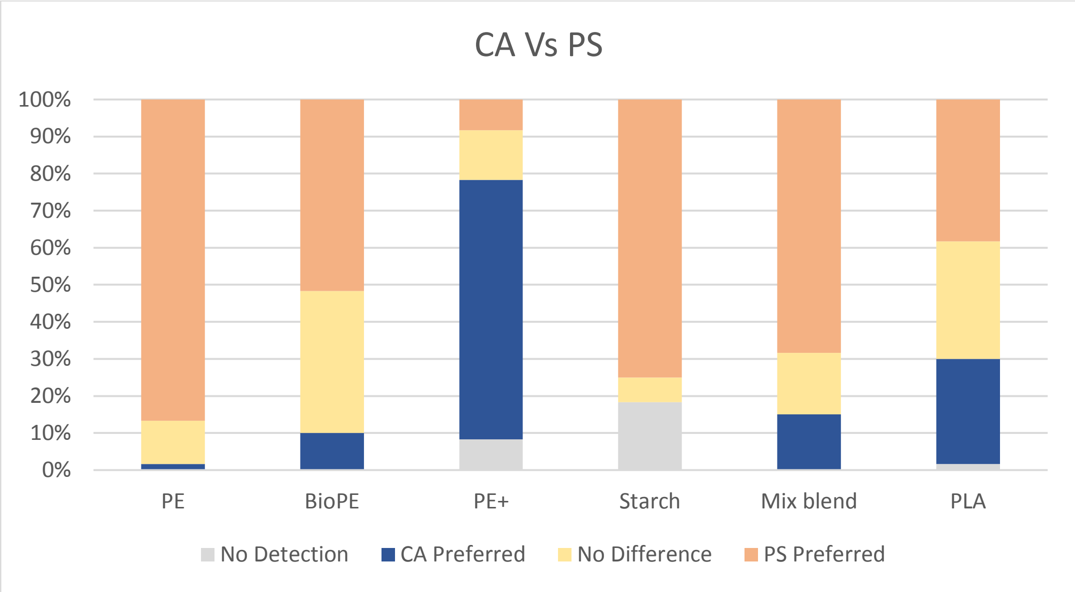


***Figure 4:*** *CA vs PS by substrate, indicating an exception in performance with PE+ samples, and zero cases of preferential development for starch samples*

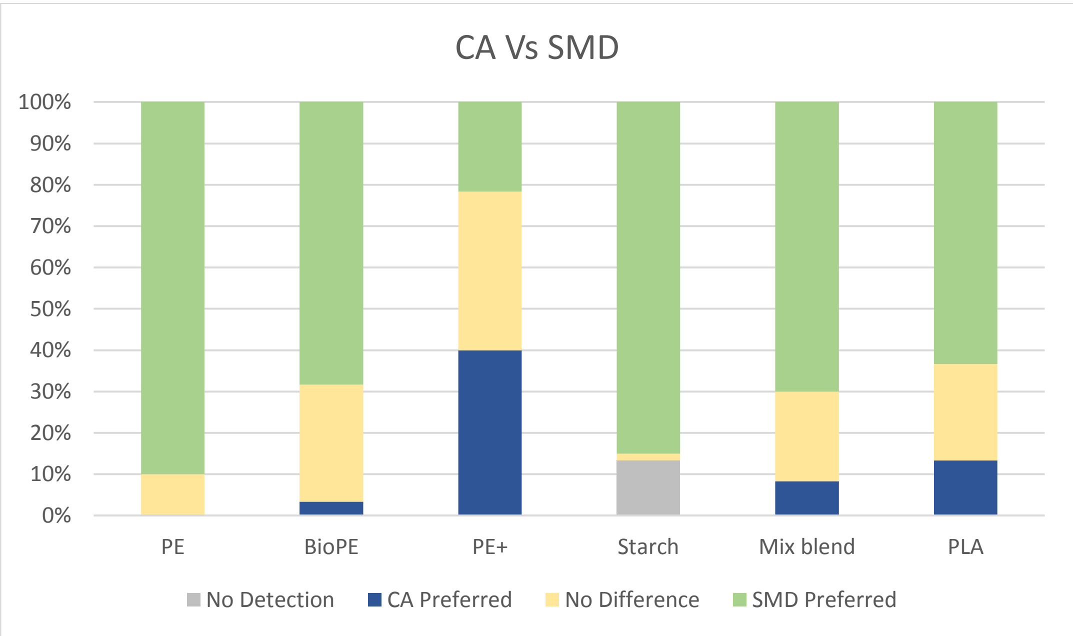


***Figure 5:*** *CA vs SMD by substrate*

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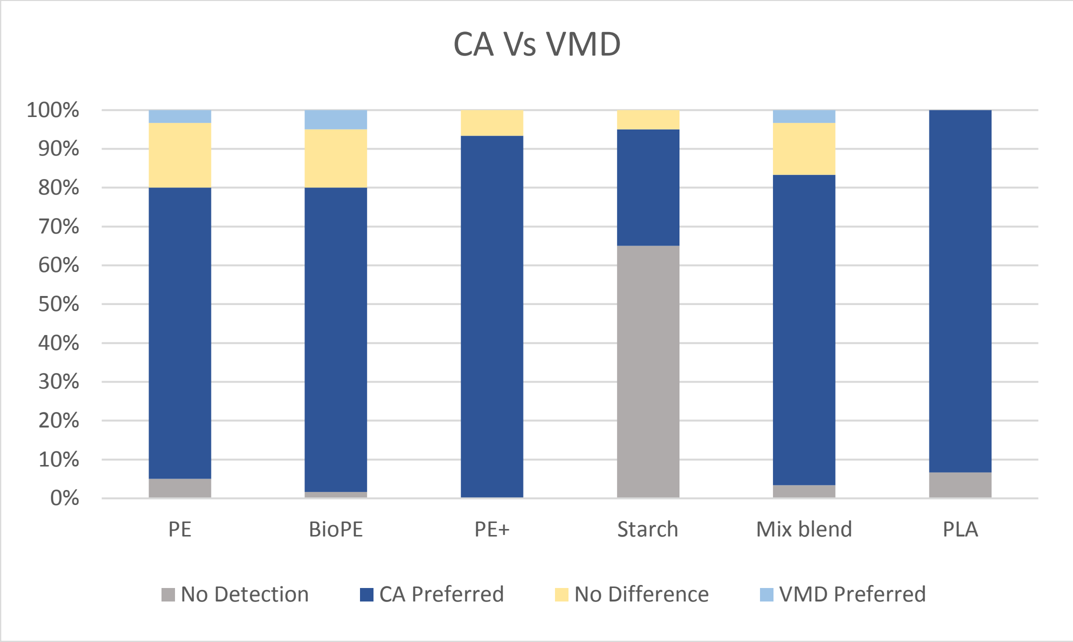


***Figure 6**: CA vs VMD by substrate*

## Powder Suspensions

The direct comparison of CA and PS previously discussed in **Figure 4** highlights the different results based on the surface of the substrates. Although PS was in general better than CA, the exceptions appear related to the surface qualities of the plastic. Of note is the PE+ sample, which was a thick, smooth, and shiny surface. For this substrate, the CA fuming was notably more effective. To a lesser extent this can be seen in the PLA sample which also appears relatively smooth and shiny. For this surface CA was equivalent or better in 60% of cases. Alternatively, PS was more favourable on the other plastics which were visually less smooth and reflective.

## Vacuum Metal Deposition

VMD was the least effective method overall, as shown in **Figure 2 and 7**, where each other development method performed notably better across all comparisons. Observationally, VMD was able to develop ridge details on PE based substrates, however in the vast majority of cases the alternative developed a fingermark of equivalent or better quality.

[Type here]

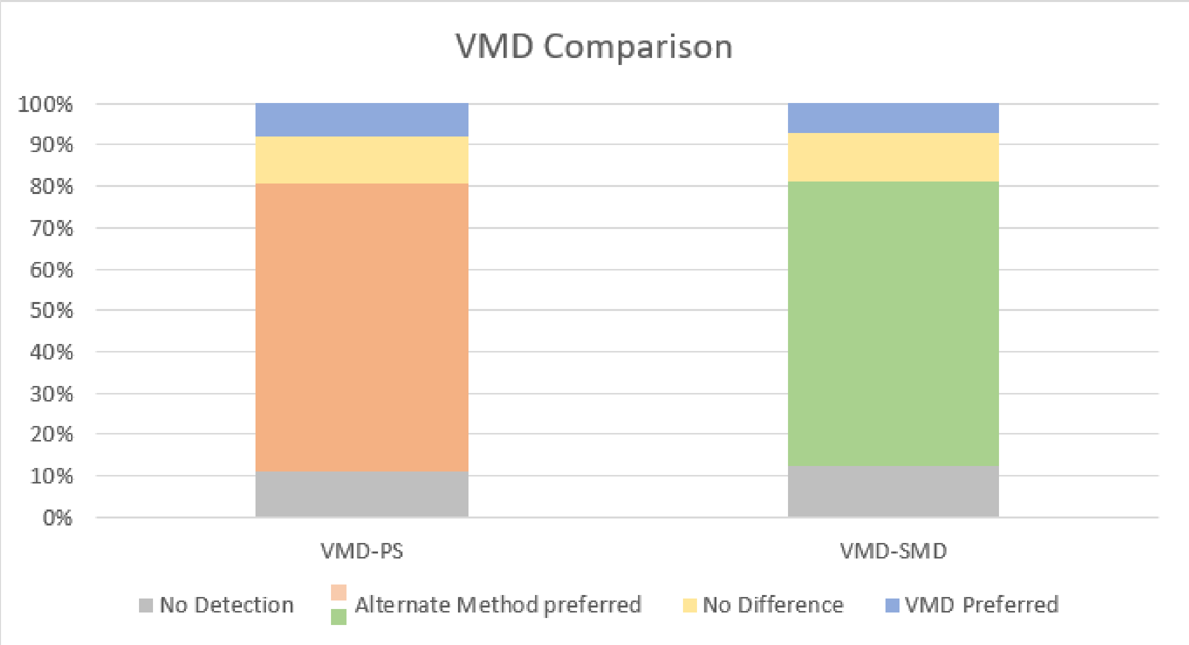


***Figure 7***: *Comparison of VMD vs PS and SMD.*

## Single Metal Deposition

SMD was originally considered as an alternative technique, which has shown to be effective on non-porous samples. However, it is generally not used operationally. Despite this, SMD was overall the most consistent and effective method when compared to the other fingermark detection techniques. **Figures 2, 7 and 8** indicate the results of all comparisons to SMD. From those results, SMD was the preferred method. SMD tends to provide the most consistent results across all substrates and was less affected by the donors and the age of the mark. As visible on **Figure 2**, it also led to the smallest rate of no detection.

The time taken for complete processing with SMD was approximately about 45 minutes. This was similar to CA when accounting for humidity and purge cycles but was much quicker than VMD, which often took over an hour to reach the required vacuum level before evaporation could take place. PS was the quickest with at most a 30 second delay between application and rinsing – but was limited to processing each sample individually while each other method could process batches of samples. SMD required more hands-on processing than VMD and CA, but a notable advantage is that no over development occurred with SMD, which is a common risk to both VMD and CA unless carefully monitored. The times discussed above do not account for drying the samples after processing with SMD and PS. Although initial observations can be made while still wet, for optimal photography of each fingermark enough time must be accounted for to dry the samples completely.

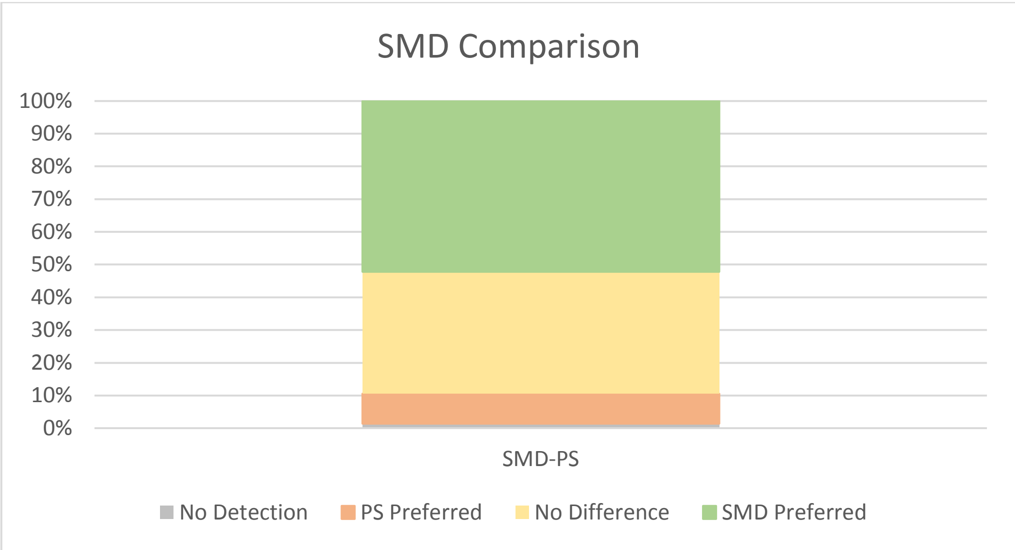


***Figure 8**: Comparison of SMD vs PS*

The following images in **Figure 9** are representative of the comparisons between SMD and PS, across PLA, starch, and mix blend compostable plastics. Of note is the green starch sample which was consistently difficult to develop clear ridge detail on, but of the methods used SMD was most effective. While on the right of the image is the black mix blend plastic. This image highlights the colour changing ability of an SMD developed fingermark in certain circumstances. When developed on a dark surface, with careful lighting control during photography, the fingermark can appear as a bright gold colour, allowing for increased contrast.

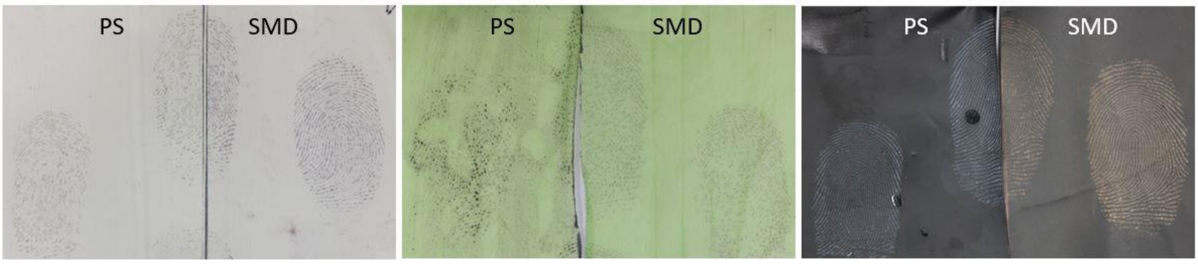


***Figure 9**: Left – PS vs SMD on rigid PLA. Middle – PS vs SMD on starch based compostable . Right – PS vs SMD on mix blend. Images taken from 1 week aged samples.*

## Other variables

In this study variables were minimised where possible to simplify gaining an initial grasp of the impacts of biodegradable plastics on fingermark development. Although the age of fingermarks, the donor, and depletions were monitored throughout – as the marks were only assessed with a comparative scale, few observations can be made based on these variables. Using the cases of no detection, we

[Type here]

can observe an expected but minor raise in cases of no detection as the depletion number increased. Similarly, with the aged fingermark samples the lowest incidence of no detection occurred with the freshest fingermark samples at 27 occurrences – this raised to 36, 55, and 45 occurrences for one, two, and four-week-old fingermarks respectively. Interestingly there was one donor of the five who had more than double the incidences of no detection attributed to their fingermarks, highlighting the significance of donor variability. The variables surrounding fingermarks are complex, and without quality assessments further conclusions are beyond the scope of this research.

### Results Summary

Overall, the results can be effectively summarised in **Table 6** which displays a ranking from 1 to 4 in order of performance across each substrate. SMD outranks each method on all surfaces except for PE+ (with a degradable additive). CA and PS favoured particular surfaces, while VMD was generally ranked the least effective, with only limited success on Bio-PE surfaces when compared to the alternative development methods. Equal rankings were given in cases of similar effectiveness for that surface.

***Table 6****: Ranking of each development method by performance across each substrate.*

| Development | PE | Bio-PE | PE+ | Starch | Mix blend | PLA |
|---|---|---|---|---|---|---|
| **CA** | 3 | =3 | 1 | 3 | 2 | =2 |
| **PS** | 2 | 2 | 3 | 2 | =1 | =2 |
| **VMD** | 4 | =3 | 4 | 4 | 4 | 4 |
| **SMD** | 1 | 1 | 2 | 1 | =1 | 1 |

## Discussion

To be able to continue to effectively process items for fingermark development in operational contexts, it is important to constantly research and explore newly developed materials to ensure the accepted and currently employed techniques are optimised, and in this case still effective at all. As previously mentioned, CA and VMD are two of the most employed development methods for plastics samples. However, this study has determined that both methods have limited effectiveness for recovering fingermarks from biodegradable plastics and may not be an appropriate first choice method for even traditional PE-based plastics. Particularly as alternative plastic materials become more common, it is paramount to be aware of the limitations of these methods to ensure that valuable forensic traces are not lost.

The primary variable was the substrates themselves. Initial efforts were made to broadly categorise each plastic based on chemical composition, yet it was observed, particularly in the case of CA and PS, that the surface texture had a larger impact on the success of fingermark development. This is an area of much needed further research to explore texture and composition within a category of materials to determine the interactions taking place with fingermark residues.

The previous research on biodegradable plastics by Illston-Baggs *et al* [16], concluded the most effective sequence of CA fuming, followed by staining with basic yellow 40 (BY40), then VMD, while powder suspensions were the least effective. In contrast to this study, where powder suspensions were generally the second most effective development method. This is likely due to the previous research using an iron oxide-based suspension which is observed to cause background staining on some surfaces, compared to the more appropriate choice of a carbon or titanium dioxide-based powder suspension. The most obvious difference in results is the inclusion of SMD as an alternative method, which was shown to be far more effective than the other recommended methods. However as sequential development was not examined it cannot be conclusively determined as the best available method. This previous study also observed surface texture to have a notable impact on the detection methods where relatively rough surfaces were seen to negatively impact detection methods, however in this case the powder suspension method was most drastically affected.

As this study considered direct comparisons between multiple development methods, the individual quality of fingermarks was not assessed, rather only the comparative performance was monitored per the scale in **table 5.** It is important to be careful when interpreting these results, while also being aware of the limited number of depletions and donors. This does not necessarily mean the preferred development method had extremely high-quality fingermark developments, it could suggest instead that the alternative method had no developed ridge detail, and so any clear ridge detail would make it the preferred method. Further research is necessary to assess fingermarks by quality to make any statements of being suitable for use in comparison for identification.

Several results were obtained from this study, which could have a large impact on operational processing recommendations, for example not using CA on starch-based plastics. However, it is important to note that each method was considered in isolation. For casework application, fingermark development can require a sequence of development methods to achieve optimal results. In this case further research is necessary to explore if fingermarks can be recovered post CA fuming using sequential developments, to then determine if CA fuming is still an appropriate method to consider for first processing.

Post fuming CA with a fluorescent dye (such as BY40) was also not undertaken for this study. Although it has been shown to visually enhance the fingermark and maximise contrast, in this case it was considered unnecessary as the contrast against the surfaces was not an issue during imaging. As staining is a sequential technique, it would be considered as another major variable that would need to be compared to all other methods, while also including sequential developments of each other technique, hence the added complexity outweighs the benefit for this preliminary research.

## Conclusions

The aim of this research was to determine if biodegradable plastics have an impact on fingermark development. This was ultimately confirmed by the results which indicate a clear favourability towards SMD development across all substrates, while more conventional methods such as CA fuming and PS were inconsistent and depended heavily on the texture of the surface, while VMD favoured more traditional PE based polymers and was relatively unsuccessful with the biodegradable plastics. The significant difference of some bioplastics when compared to PE is evident in these results, and importantly should be considered a separate substrate category. Initial recommendations can already be made based on these results to avoid CA fuming starch-based plastics, and more broadly to consider SMD as a reliable development method across a range of surfaces.

Operationally it may not be possible to profile an unknown plastic sample prior to development. However, as this research has shown, considerations should be made to allow for such sample analysis where practical. In the absence of profiling the material, awareness of these plastics can be sufficient to consider alternate development methods.

With a constantly changing landscape of materials being developed and becoming more common, this in turn creates a constant challenge of keeping up to date with how these changes and new materials can impact forensic science in general. This highlights the need for continual research into the nature of the impact of different surfaces and how they interact with fingermark residues and development methods to try to understand at a more fundamental why these results take place.

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The authors truly appreciate the time taken to provide detailed feedback from each reviewer. Below are the responses to each question, comment, and suggestions. All minor grammatical and typographical corrections have been accepted and corrected in the manuscript, as well as minor changes to figure numbers and captions as required.

Reviewer #1: This study presents an insight into the development of latent fingermarks on biodegradable plastics from an Australian perspective. This study will be relevant to the forensic science community as these substrates are set to rise in the general circulation due to environmental and governmental policies. Although the authors address the limitations of the study by stating the work is preliminary, caution is required with some of the statements made.

Title: biodegradable and compostable is not the same. Can the authors clarify which type of material was used? A particular material can be degradable and even biodegradable without being compostable. Compostable polymers have additional requirements when compared to biodegradable polymers.

> **Authors Response:** Of the six materials chosen, four are biodegradable, and of these four, three are compostable according to manufacturing claims. As the other two materials (polyethylene and a non-degradable plant-based polyethylene) were chosen as a reference point to compare to the four biodegradable materials, it was deemed more appropriate to simplify the title to “Biodegradable Plastics…”, with the further explanation of the difference in terminology explained in the introduction.

Introduction
Good explanation of the terminology and an insight into what we are dealing with. Also good set of references. The explanation here does partly address my comment above in relation to the title.

> **Authors Response:** The previous authors response referring to the title choice applies to this comment. No changes were made to the manuscript title.

"As it is highly transparent, strong, and can be made into? rigid or flexible sheets, PLA is often considered as the most suitable replacement for PE and PET." Remove ?.

> **Authors Response:** Correction accepted.

Methodology:
The methodology is well presented and explains how the number of samples was selected and why.

How were the hands washed? Was it with soap and if so what type of soap? Can the soap can an effect on the fingermark constituents when deposited with a different result when in contact with the enhancement technique? Is 5 minutes prior to deposition enough time to wait - it seems like really short. Although the IFRG guidelines do not stipulate a time, CAST guidelines state "Testing in this phase of the work should be the most extensive, using deliberately deposited finger marks throughout. These should be 'natural' finger marks, deposited by donors who have not washed their hands in the past 30 min, have not applied cosmetics in the same time period, and have not deliberately wiped their hands across regions of the face rich in sebaceous products."

**Authors Response:** Hands were required to be thoroughly washed using soap and water. Although a specific soap was not supplied, donors were instructed to thoroughly rinse hands to remove any potential soap residue before drying. This was to minimise contamination allowing for the most consistent fingermark deposition possible.

As the IFRG guidelines did not stipulate a required time, a pilot study was completed to determine an appropriate time between washing hands and deposition that compromised for donor logistics. Due to the large number of fingermarks deposited per donor, multiple sessions were required throughout the study, and shortening preparation time allowed for less of a time commitment and minimised the length of time for potential hand contamination (through sub consciously touching a surface or their face). 5 minutes was chosen as times beyond this showed little improvement if any at all, and the extra donor availability allowed for the examination of more variables which far outweighed the small benefit of delaying deposition.

The methodology states "three minutes between additional depositions" - is this enough time for the secretions to charge again. Is there a reference for this? And is 3 depletions enough to assess sensitivity?

**Authors Response:** The pilot study previously discussed in the above authors response also examined the time between depositions. With clean washed hands, three minutes was enough time to allow the secretions to charge again if donors were careful to not touch any other surface. Waiting any longer than three minutes did not have an observable difference on fingermark quality.

Three to five depletions is the standard practice for this level of exploratory fingermark research. With the focus of the research being on the materials themselves, additional depletions were not necessary.

Cyanoacrylate - was there a dye stain procedure?

**Authors Response:** Dye staining was not used in this case, as it was considered to be a sequential development method, and if included, all other sequential development methods would need to be considered for each comparison to make fair assessments. There is little doubt that sequential developments would have improved the quality of the fingermarks, however as this was a preliminary comparison study it was beyond the scope of this research and is an opportunity for future work.

Please see the below response to the question "*Is there a justification why sequences were not considered? Improvements would have been observed*" to see the addition to the manuscript which clarifies this point.

VMD - when different types of material were tested, did the evaporation and visualisation by zinc not have a different rate for each material? Is this standard practice that different materials are processed at the same time?

**Authors Response:** In this case the materials were able to be processed at the same time as the zinc evaporation required was observed to be consistent between the polymers in prior testing.

The following was added the manuscript (page 10, line 223) "*The zinc evaporation required in this case was observed to be consistent for each of the materials chosen, allowing a mixture of samples to be processed at one time. It is important to note that this is not always the case when processing different materials with VMD, and tests should be carried out prior to processing different materials at the same time to determine the required metal evaporation."*

Is there a justification why sequences were not considered? Improvements would have been observed

**Authors Response:** The following has been added to the manuscript (page 8, line 186) to explain why sequences were not considered

*"As the focus of the research was to assess the impact that biodegradable materials have on fingermark detection methods, the following four detection techniques were completed with direct comparisons to each other, and no sequential techniques applied. Rather it was the single methods only that were compared, without subsequent enhancement by any other method. Although further enhancement may have improved the fingermark quality in some cases, it was beyond the scope of this research, and subsequent work may be conducted to examine the preferred methods in sequence."*

Results and discussion

The authors do state this work is preliminary but suggest that "SMD was at least as effective or better than CA fuming in 87% of cases." Having said that, the results can be very different if the reflected long-wave UV and dye staining with CA fuming are applied. Also, sequences were not considered.

**Authors Response:** This is correct, and is potentially not clear enough in the results section, although further sequencing improvements are briefly explained in the discussion section.

The following has been added to the manuscript (page 12, line 292) "*Although this is clearly a substantial preference for SMD, it is important to note that dye staining and sequential developments were not applied which may have improved the performance of each method. In general, however, the poor results of CA fuming were not an issue of visualisation but were due to poor development overall. In these cases, dye staining would not have had any major effect as there was little ridge detail to enhance."*

Did the authors make any observations with regards to the texture of the surface when rinsing with water after PS? Did they go wrinkly or bubble up? If water was used as a rinse - does this not justify the use of dye stain after CA even if it is water-based rather than solvent-based?

**Authors Response:** No observable changes occurred to the surface or texture of the materials when rinsed with water. The starch containing materials sometimes curled after drying, but as this did not appear to impact the fingermark development it is not discussed in the manuscript. Also, as the sample halves were separated, the CA fumed materials were not exposed to water for the comparison.

The graphs are certainly useful but I would recommend a few images of developed fingermarks comparing across substrates, techniques, donors etc.

**Authors Response:** This is a good suggestion. Images have been included in the results of the manuscript (page 13 – Figure 3. Page 17 – Figure 9), as well as text above each image to give slightly more context. Brief captions have been included, as well as an adjustment to each figure number to account for this change.

The images included cover each method, each substrate, and come from a range of different donors. For consistency each image included is from the 1 week age period.

When discussing the treatment times, consideration has to be given for the drying time. No observations can be made until thoroughly dry for PS and SMD.

**Authors Response:** Good point, although initial observations can be made, optimal photography was completed when dry.

The following has been added to the manuscript (Page 16, line 363) "*The times discussed above do not account for drying the samples after processing with SMD and PS. Although initial observations can be made while still wet, for optimal photography of each fingermark enough time must be accounted for to dry the samples completely.*

For SMD, can the authors discuss the fact that this technique is a category C process in the Fingermark Visualisation Manual. Why is that the case? And why is the MMD process a Category A. What is the difference? What more can be done to use this process more in operational casework?

**Authors Response:** As the authors have no affiliation with the Fingermark Visualisation Manual, our comments are limited to speculation in this matter.

The manual acknowledges SMD as being in Category C instead of A because "This process has not been compared to Multi-Metal Deposition and currently produces marks of lower contrast with the surface. The Single Metal Deposition formulation has not been fully optimised." While this was true at the time of publication in 2014, the SMD (II) protocol was published in the following year, which is simpler and more optimised than the previous SMD method, while requiring less steps and chemicals than MMD, and also being more stable.

Before being used more in operational casework a thorough comparison between the two methods would need to be undertaken. As the SMD (II) protocol is still relatively recent, more research will need to be completed before operational validation is likely to occur.

I would recommend results and discussion together.

**Authors Response:** The discussion has been intentionally included as a separate section instead of combining with results. The authors feel that as the results section is already heavy with different comparisons, it is valuable to clearly frame the overall results within the contexts of previous research and operational contexts. To include these statements within the results may create a cluttered manuscript and would require repetition of similar statements within each result sub-heading.

In the discussion, based on the results - care must be taken about certain conclusions as sequences were not considered, depletions were only down to three and CA fuming was not followed by dye staining. Although the authors acknowledge the dye stain - further down the depletion sequence, the use of a dye stain can be beneficial. I would not consider this to be a minor benefit. Were other processes considered such as one-step fluorescent cyanoacrylate processes and amino acid staining techniques.

**Authors Response:** The following has been added to the manuscript (page 19, line 430) in regard to conclusions drawn, *"It is important to be careful when interpreting these results, while also being aware of the limited number of depletions and donors."*

The final sentence of the discussion section (page 19, line 445) has been slightly rephrased to reflect the significance of dye staining (removed "minor benefit"), and clarify why it was not included: *"As staining is a sequential technique, it would be considered as another major variable that would need to be compared to all other methods, while also including sequential developments of each other technique, hence the added complexity outweighs the benefit for this preliminary research."*

Regarding one-step fluorescent cyanoacrylate processes, studies conducted by the authors have indicated that one-step cyanoacrylates have a poorer performance compared to traditional cyanoacrylate fuming so were not considered.

Amino acid techniques would be more suited for porous substrates, so were not initially considered. However, after observing the poor performance of CA fuming on some materials, tests were conducted using indanedione zinc. This was entirely unsuccessful, with no fingermark development at all, and as such is not discussed in the manuscript.

Conclusions

The authors should discuss how will this reflect operational work? Do the authors envisage that laboratory officers will check what material they exactly have before processing?

**Authors Response:** The authors acknowledge that with operational constraints of time and budget, it is unlikely that samples will regularly be examined to determine what the material is. We envisage that in high profile cases such an examination should take place, while in more routine case work this may not be practical. However, the two starch containing plastics that were examined had quite a distinct look and feel, which is purely observational and by no means conclusive, but awareness of these types of materials would be sufficient to rethink processing of unknown plastics.

The following has been added to the manuscript (page 20, line 459): *"Operationally it may not be possible to profile an unknown plastic sample prior to development. However, as this research has shown, considerations should be made to allow for such sample analysis where practical. In the absence of profiling the material, awareness of these plastics can be sufficient to consider alternate development methods."*

Reviewer #2: Interesting research in the current context of the increasing use of biodegradable materials.

1) The first and most important remark I have to make is related to the section "Materials and Methods" and more precisely to "Fingermark Deposition". In my opinion, this section lacks a little clarity. At the end of the reading, I am not sure I really understood how the fingermarks were deposited.
The total number of fingermarks deposited is not indicated outside the Abstract, and it would be nice to recall it in this part, and perhaps to specify how many fingermarks are left by a donor for a single comparison of techniques and aging time (3, 9, or more ?)

**Authors Response:** The authors acknowledge that this section could be more clear, and have included the following statement to the manuscript (page 8, line 180) to clarify by restating the specific variables and include the total number of fingermarks as mentioned in the abstract.

*"In total, each donor deposited nine individual marks for a single sample, in sets of three depletions per sample. With six direct comparisons of methods, across six different substrates, each of which being aged four different times, making a total of 1296 fingermarks per donor. With five donors this was 6480 individual fingermarks deposited."*

2) Some relevant images of fingermarks for the different comparisons would be appreciated to illustrate the results in addition to the graphs. In particular, you say in the "Discussion" section, that results favouring one technique over another do not mean that the fingermarks produced are necessarily of high quality. Having images in this case would support your point.

**Authors Response:** Images have been included in the results of the manuscript (page 13 – Figure 3. Page 17 – Figure 9), as well as text above each image to give slightly more context. Brief captions have been included, as well as an adjustment to each figure number to account for this change.

The images included cover each method, each substrate, and come from a range of different donors. For consistency each image included is from the 1-week age period.

3) Modification of the titles of figures 6 and 7.

**Authors Response:** The captions of figures 6 and 7 have been corrected.

4) Nowhere in the article is there a reference to Table A-1 in the Annex.

**Authors Response:** Appendix removed as deemed not required for the article

Credit Author Statement

Harrison Woodward: Methodology, Investigation, Writing – Original draft, Writing – Review and editing, Visualisation

Sebastien Moret: Conceptualisation, Supervision, Writing – Review and editing, Project administration

Scott Chadwick: Conceptualisation, Supervision, Writing – Review and editing, Visualisation, Project administration