



General Forensics

New light on old fingermarks: The detection of historic latent fingermarks on old paper documents using 1,2-indanedione/zinc



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ABSTRACT

This study explores trends in the effectiveness of 1,2-indanedione/zinc chloride (IND/Zn) for visualizing latent fingermarks on paper substrates of various ages. Preliminary investigation of contemporaneous documents showed that high quality fingermarks could be deposited through incidental handling, although smudging and overlapping were evident. IND/Zn was then applied to incidentally handled documents up to 80 years old and successfully developed potentially identifiable fingermarks, significantly increasing the established timescale for fingermark detection with amino acid sensitive reagents. The results indicate that IND/Zn remains effective over longer periods than has been previously demonstrated, although a comparison between documents of different ages suggest that progressive diffusion of the target amino acids occurs over time, affecting the proportion of potentially identifiable marks. The findings of this study reinforce the applicability of IND/Zn for the detection of historic latent fingermarks on old paper documents.

1. Introduction

Detection of latent fingermarks in operational cases is generally carried out days, months, or even years after deposition. Between deposition and processing, fingermarks undergo ageing through various chemical, physical and biological processes resulting in changes to their chemical composition [1–3]. The mechanism and rate of the ageing processes are influenced by environmental conditions, the nature of the substrate and the detection technique applied [1–5]. It is to be expected that longer ageing periods result in greater degradation of fingermark components, rendering detection of older fingermarks more challenging. This leads to the fundamental question; how long can latent fingermarks be realistically expected to survive on a depositing surface and remain detectable? Although the impact of time on the performance of the detection technique is well known over reasonably short timescales [6–8], there have been minimal studies that have attempted to answer this question, which has particular relevance for cold case reviews.

The nature of the deposition substrate plays an important role in the survivability of latent fingermarks. On non-porous surfaces (such as glass, metal, and plastics), both eccrine and sebaceous residues generally remain on the surface of the substrate and are vulnerable to environmental conditions. However, on porous surfaces such as paper,

cotton and wood, the eccrine residue can be absorbed into the substrate [2,9]. The absorption of these components within the substrate matrix provides better protection from the environment, which may enable them to persist longer compared to non-porous surfaces. For example, the ability for paper to absorb and retain fingermarks has been suggested as a mechanism of preservation [2,9,10] which influences the detection of old fingermarks. Paper substrates is one of the most abundant forms of fingermark substrates encountered in criminal investigations [11] and likely to be evidence of interest in cold case reviews.

Another factor in the ageing process of latent fingermarks is the environment. Whilst a target compound may be stable over time under certain conditions, exposure to unfavourable conditions can increase the rate of degradation or diffusion processes. It is known that humidity significantly affects the eccrine constituents; for example, chlorides have been determined to diffuse faster under high relative humidity. This has been suggested as the reason why silver nitrate, which targets the chloride ions, is less effective on older fingermarks [3,12,13]. In another study investigating the potential for degradation of selected amino acids present in eccrine residue, thermal degradation was observed after 3 min at 100 °C [14], although this was not performed on an actual fingermark residue. The combined cyclic exposure to high humidity and temperature may have an effect on the degradation and diffusion of eccrine

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components of fingermarks over time. This can have potential consequences on the detection of old fingermarks between jurisdictions with significantly different climatic conditions.

Against the backdrop of the nature of substrate and climate, the detectability of old fingermarks depends on the stability over time of the target compounds and sensitivity of the development technique. Physical developer (PD) is the best established method for developing significantly aged fingermarks on paper, presumably due to its interaction with what has been termed the robust component of the sebaceous constituents [15,16]. The ability of PD to develop old fingermarks was demonstrated in a recent study by Bleay et al. [17] where PD was found to develop fingermarks up to 90 years old, significantly extending the timescale of developing old fingermarks on paper. However, PD also involves the use of maleic acid to remove alkaline binders within the paper, rendering the paper extremely fragile [18] and can potentially damage the already delicate nature of historic documents. Furthermore, from an operational point of view, amino acid components would be targeted first, unless the paper surface is known to have been wetted.

Amino acid sensitive reagents such as 1,2-indandione/zinc chloride (IND/Zn), 1,8-Diazafluoren-9-one (DFO), and ninhydrin have long been proposed for developing older fingermarks on paper [2,19]. This has been practically demonstrated by Bleay et al. [17] where IND/Zn, DFO, and ninhydrin used in sequence were found effective in developing fingermarks on 32 year old cheques kept under controlled environments, while considerably less effective on older documents with an unknown history of environmental exposure. In contrast, Boudreault et al. [20] reported a significant decrease in the quality of developed fingermarks over time on white copy paper treated with IND/Zn, although this was not observed for fingermarks treated in sequence with ninhydrin. Taken together these studies suggest that while the binding of amino acids to cellulose is likely to provide some stability, amino acids are not completely stable.

In light of these limited studies [17,20], a natural progression is to investigate whether the trends found can be generally replicated under different climatic conditions. Such information would be beneficial to investigators by providing insight into the factors that may influence the effectiveness of the detection of old fingermarks. It is also important to consider variations in how the method is performed, particularly for techniques such as IND/Zn for which a variety of formulations have been reported [21–23].

A key challenge for studies in this area is that designing an experiment over an extended timescale is not practicable whilst accelerated ageing processes may introduce artefacts [24] and skew the results. In this context, the use of available aged documents of known handling history is a potential avenue of study. Additionally, a pseudo-operation trial has the advantage of using substrates of the type and condition likely to be encountered in cold cases thereby providing more accurate information.

This study built on previous work by Bleay et al. [17] by investigating trends in the effectiveness of IND/Zn for detecting historic latent fingermarks on old paper documents. Here, we explore a small subset of documents of various ages (75–90 years, 35–40 years and under 2 years) and extend the study on documents stored and processed under two different climatic conditions, using three different IND/Zn formulations [21–23]. Experiments were also conducted to provide an immediate comparison between contemporaneous and historic fingermarks for the

assessment of the effectiveness of IND/Zn on old paper surfaces. The findings of this research may provide information on the possible effect of climate on the detection and the comparative performance of different IND/Zn formulations of old fingermarks between the two different geographical locations.

2. Materials and methods

2.1. Substrates

Various types of paper documents representing different ageing periods, stored under the United Kingdom (UK) and Western Australian (WA) climatic conditions were donated by individuals (Table 1). These documents were categorised into three timescale groups; 75–90 years, 35–40 years, and contemporaneous (under 2 years). The known prior histories of each document set in terms of storage and handling are listed below.

2.1.1. 75-90 Year old documents

A set of 7 documents was taken from the same batch previously used by Bleay et al. [17] in the UK. This set consisted of correspondence and invoices from a ledger dated between 1927 and 1933. The documents had been stored in a house without central heating between 1933 and 1990, and subsequently in a centrally heated house. A separate set of 20 pages was taken from the middle part of a journal (autograph book) compiled in Holland and dated between 1940 and 1943. The journal was brought to WA in 1954 and had been kept in a suitcase until 2016 when the author passed away. During this time, it is unknown how many times the journal has been handled. Thereafter it was kept in a box and in a plastic bag for a year each before it was stored on a desk until 2019. During 2016–2019 some of the pages may have been handled at the edges, but to the best of the owner's knowledge, the middle of the pages had not been handled.

2.1.2. 35-40 Year old documents

A set of 21 Christmas cards and 13 pages were taken from two notebooks used for recording locomotive numbers, dated between 1978 and 1984. The notebook samples comprised 3 pages from a lined notebook and 10 pages from a pre-printed notebook of thicker and coarser paper. Both notebooks had been stored within an unheated attic space for at least 25 years in the UK. An additional set of 20 letters dated between 1980 and 1987 was also examined. These letters had been stored under WA climatic conditions in two houses until 2003 when it was transferred to a humidity and temperature-controlled storage until 2015, thereafter returned to storage in a house under ambient environmental conditions.

2.1.3. Contemporaneous documents

A set of 20 envelopes with a handling history ranging between 6 months to 2 years was collected from 3 different houses in Perth, WA. To a reasonable degree of certainty, none of the set of documents had been subsequently handled before being selected for chemical treatment unless otherwise stated. The environmental history (temperature and humidity) of these documents was unknown.

Incidental fingermarks deposited through the process of handwriting were also collected on notepad paper and envelopes. 10 donors (5 males

Table 1
Summary of the sources, ages, and the number of documents used in this study.

| Documents | Number of samples | Date(s) of Documents | Source | Timescale category |
|----------------------------|-------------------|----------------------|-----------|--------------------|
| Ledger documents | 7 | 1927–1933 | UK | 75-90 years |
| Christmas cards | 21 | 1979–1984 | UK | 35-40 years |
| Lined Notebook pages | 3 | 1979–1984 | UK | 35-40 years |
| Pre-printed notebook pages | 10 | 1978–1980 | UK | 35-40 years |
| Journal pages | 20 | 1940–1943 | Australia | 75-90 years |
| Letters | 20 | 1980s | Australia | 35-40 years |
| Envelopes | 20 | 6 months-2 years | Australia | Contemporaneous |

and 5 females aged between 23 and 48 years) were instructed to reproduce a standard portion of text through handwriting on a sheet of “Spirax” notepad paper, which was subsequently folded and placed in an envelope. These samples were left for an ageing period of at least 7 days before development.

2.2. Processing protocols

The IND/Zn formulation, treatment, and examination conditions were based on the geographical locations of each document set. Documents stored under UK conditions were processed and examined in the UK, using the formulation optimised by the Home Office Centre for Applied Science and Technology (CAST) for use in this jurisdiction [23,25,26]. The details of the processing protocols are provided in Appendix 1, with the chemicals and formulation listed in Tables A1 and A2.

Correspondingly, documents stored under Australian conditions were processed and examined in Australia using the formulation routinely used by the Australian Federal Police (AFP) on normal paper [21]. To investigate potential performance variations between formulations under particular environmental conditions, half of the documents stored under Australian conditions were processed using a formulation used by the German Federal Police [22]. The details of the processing protocols are provided in Appendix 2, with chemicals and formulation listed in Table A3 and A4.

For the purpose of this study, the different IND/Zn formulations will be referred to as the UK formulation [23,25,26], Australian formulation [21], and German [22] formulation respectively. Samples sourced in the UK were photographed under 445–510 nm excitation used in conjunction with a Schott glass OG550 filter and 480–560 nm excitation used in

conjunction with a Schott glass OG590 filter. Whereas those stored under Australian conditions were photographed under 505 nm excitation using a Schott glass OG550 nm filter.

2.3. Examination and interpretation

Post treatment, each document was examined back-to-back for developed fingermarks and characterised into three groups; no fingermarks, evidence of handling, and potentially identifiable fingermarks (Fig. 1), based on the best quality fingermarks developed across the document.

3. Results and discussion

3.1. Preliminary observation

Prior to examining the old documents, the expected distribution of fingermarks was evaluated by having donors deposit incidental fingermarks on handwritten notepad paper and envelopes. After an ageing period of at least 7 days, an equal number of samples were treated with the German and the Australian formulations.

The distribution of fingermarks on the handwritten note was found to vary across donors. Common trends found were a high number of fingermarks along the edge and smudged palm marks across the body of the document, likely due to the writing process (Fig. 2a). Similar fingermarks pattern associated with writing letters has been reported in a recent study [27] evaluating the location of fingermarks on letters to distinguish between the activity of writing and reading of letters. Randomly dispersed fingermarks observed on the envelope did not show

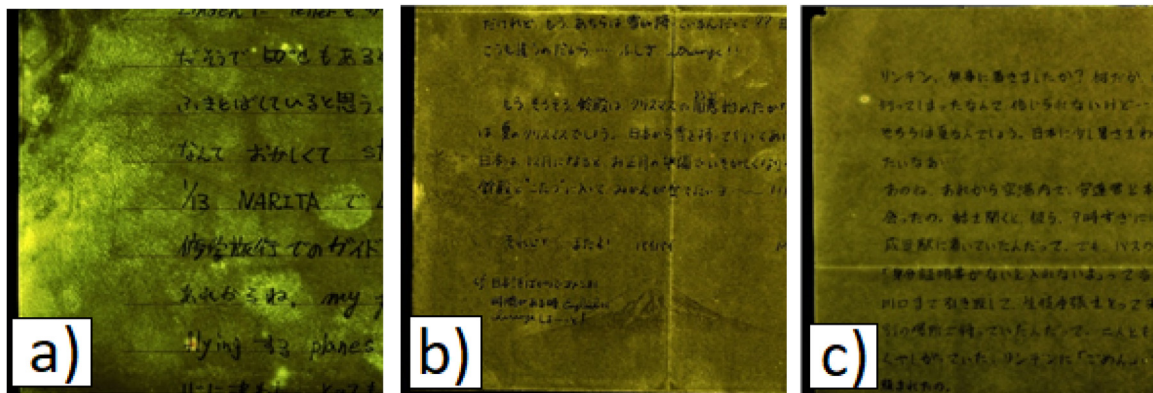


Fig. 1. Example of developed fingermarks on documents dating back to 1980, showing potentially identifiable fingermarks (a), evidence of handling (b) and no fingermarks (c).

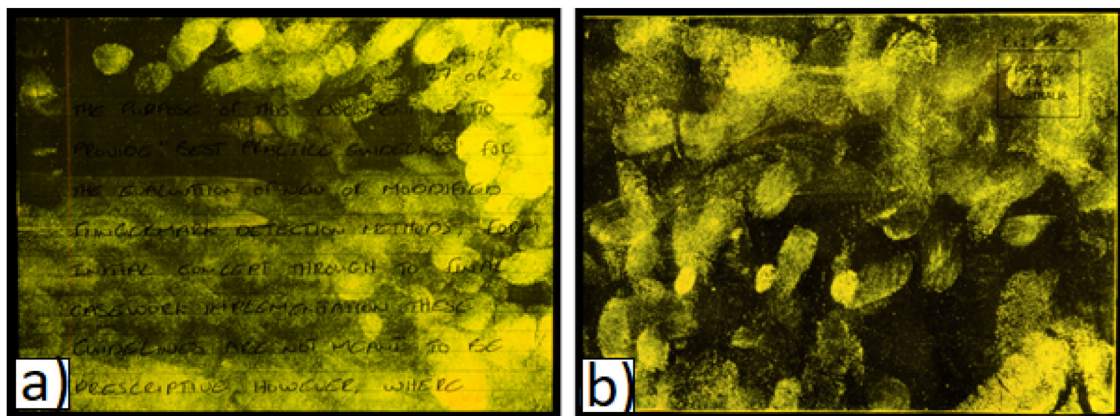


Fig. 2. Examples of the development of incidental fingermarks on handwritten notes (a) and envelopes (b) treated with IND/Zn, German formulation (a), and Australian formulation (b).

Table 2
Characterisation of documents of different ageing periods based on the quality of fingermarks developed after treatment with different IND/Zn.

| Ageing Time | Documents Type | Source | Number of Documents | | |
|------------------|----------------------------|-----------|---------------------|----------------------|--------------------------------------|
| | | | No fingermarks | Evidence of Handling | Potentially Identifiable Fingermarks |
| 75-90 years | Ledger Documents | UK | 7 | 0 | 0 |
| 75-90 years | Journal pages | Australia | 5 | 12 | 3 |
| 35-40 years | Christmas Card | UK | 10 | 8 | 3 |
| 35-40 years | Lined notebook pages | UK | 0 | 1 | 2 |
| 35-40 years | Pre-printed Notebook pages | UK | 0 | 9 | 1 |
| 35-40 years | Letters | Australia | 2 | 4 | 14 |
| 6 months-2 years | Envelopes | Australia | 0 | 1 | 19 |

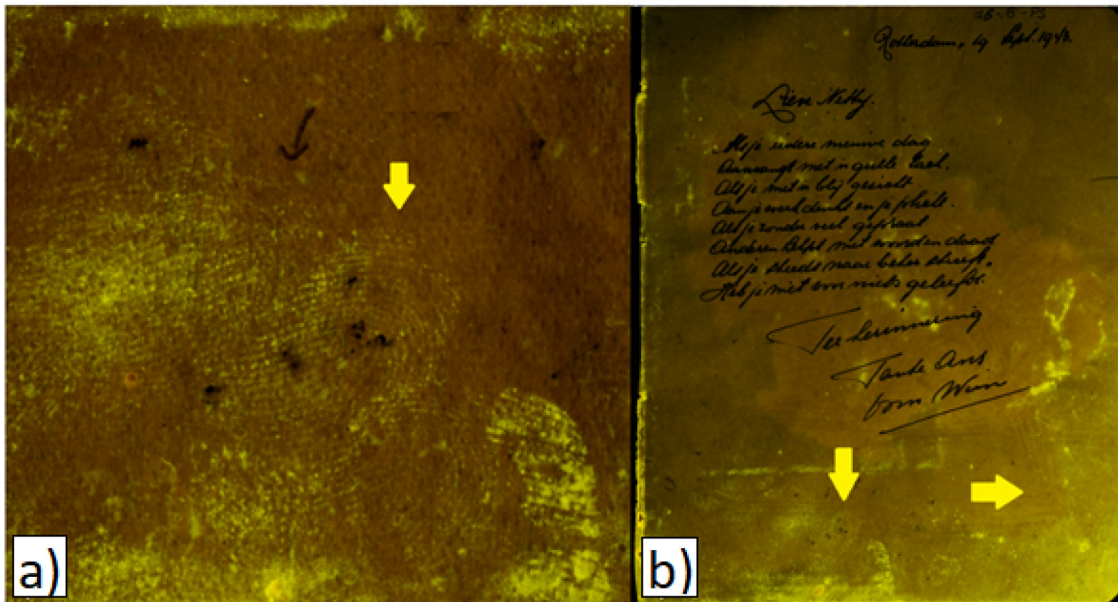


Fig. 3. Potentially identifiable fingermark (a), and diffuse features possibly associated with original areas of contact (b), developed with IND/Zn on a journal dating back to 1940s.

similarities with the model [27], likely due to the distinct activity of handling and placing the letters in the envelope compared to writing and reading of letters. The fingermarks were generally of high quality, but smudging and overlapping were evident (Fig. 2b). No observable differences in the effectiveness of the German and Australian formulation were observed. The result was used as a benchmark for comparing incidental fingermarks on the corresponding type of documents used in this study.

3.2. General observations

The three categories of documents represent a spectrum of fingermarks of potential forensic interest. The contemporaneous documents represent those likely to be encountered in routine casework; 35–40 year old documents align with cold-case reviews; whilst 75–90 years old with documents of historic significance. Following IND/Zn treatment, the three batches of documents were examined for fingermarks and the results summarised in Table 2. The results indicate that IND/Zn remains effective over longer periods than has been previously demonstrated, but there is a clear trend in which the proportion of potentially identifiable fingermarks decreases over time.

3.3. 75-90 Year old documents

IND/Zn was effective in developing fingermarks on 80 year old documents under Australian conditions. Potentially identifiable

fingermarks were found on three of the journal pages, with the best fingermark found shown in Fig. 3a. The developed fingermarks were faint, likely due to significant degradation of amino acids over the long ageing period. A high level of background luminescence was observed, predominantly at the edges of most documents (Fig. 3b), an area which is potentially more exposed to environmental conditions compared to the

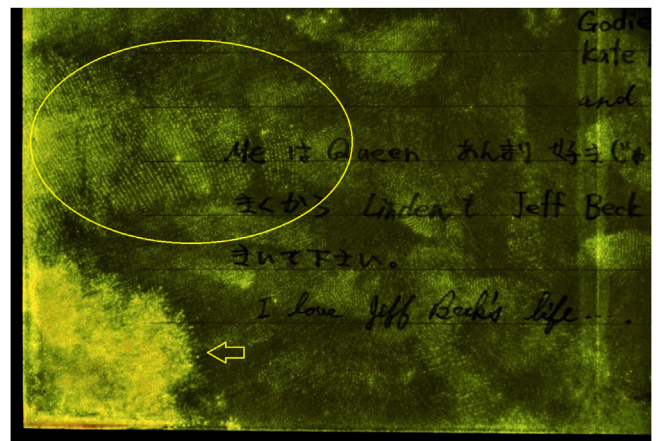


Fig. 4. Ridge detail (circled) and possible diffusion (indicated by arrow) developed with IND/Zn on a letter dating back to 1980s.

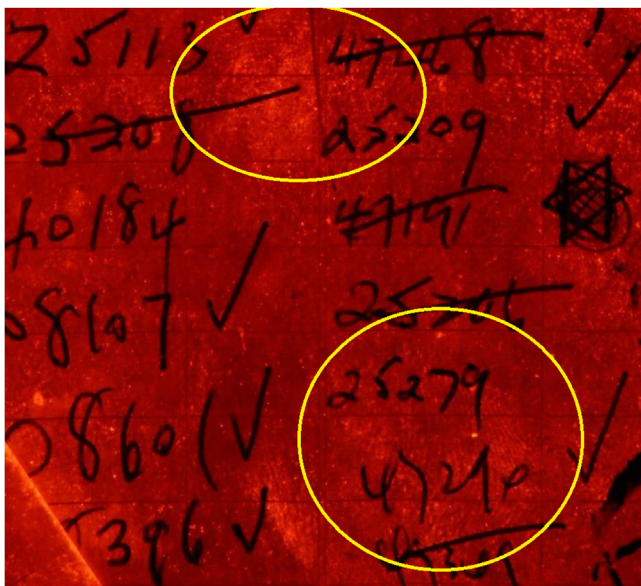


Fig. 5. Ridge detail (circled) developed with IND/Zn on a page of a lined notebook dating back to 1984.

centre of a closed journal. As indicated by preliminary investigations, the edge of a document is generally a high contact area. Diffusion of the deposited amino acids over time may result in background luminescence and loss of ridge detail, but the possibility of smudged fingermarks must also be considered. The position of these fingermarks away from the edges gives greater confidence that these marks were deposited at the time the documents were produced. Developments of marks that are over 40 years old have been recorded using ninhydrin [11], the results significantly increase the established timescale on fingermark detection with amino acid sensitive reagents.

Under UK conditions, IND/Zn was ineffective in developing ridge detail on the 75–90 year old documents. This is consistent with results obtained using ninhydrin and DFO in a previous study using documents from the same set [17]. Exposure of the documents to high humidity and temperature fluctuations over a prolonged period may have resulted in the dissipation of any amino acids that could have been present. The greater relative humidity (60–90%) in the UK compared to WA (40–70%) [28] are likely to have played a significant role in the differences observed.

3.4. 35-40 Year old documents

The success rate on the Australian 35–40 year old documents was higher in comparison to the 75–90 year old documents, with 70 % yielding potentially identifiable fingermarks. These were mostly located around the edges of the document as expected, with fewer across the main body (Fig. 4). In comparison to the 1940s journal, the quality of the fingermarks developed on the 1980s letters was significantly higher, possibly due to less diffusion having occurred over the shorter ageing duration. The result is in agreement with previous work by Bleay et al. [17] reporting the effectiveness of IND/Zn in developing 32 year old fingermarks stored under laboratory conditions. The present study extends this finding to documents of 40 years stored under non-laboratory conditions, which are more representative of the condition of documents likely to be encountered in casework.

Similar trends were observed for documents treated under UK conditions. The best results were obtained on the lined notebook pages, an example being shown in Fig. 5. These marks appeared to be more stable and yielded several regions of potentially identifiable ridge detail. The success rate on the thicker/coarser pre-printed notebook pages and Christmas cards from the same time period was much lower. The least effectiveness was observed on the Christmas cards with no marks found on 10 out of 21 cards. It has been shown previously [17] that ninhydrin, DFO, and IND/Zn remained effective on cheques stored under laboratory conditions for periods equivalent to the documents used in this study. However, the documents used here were of different paper types and had

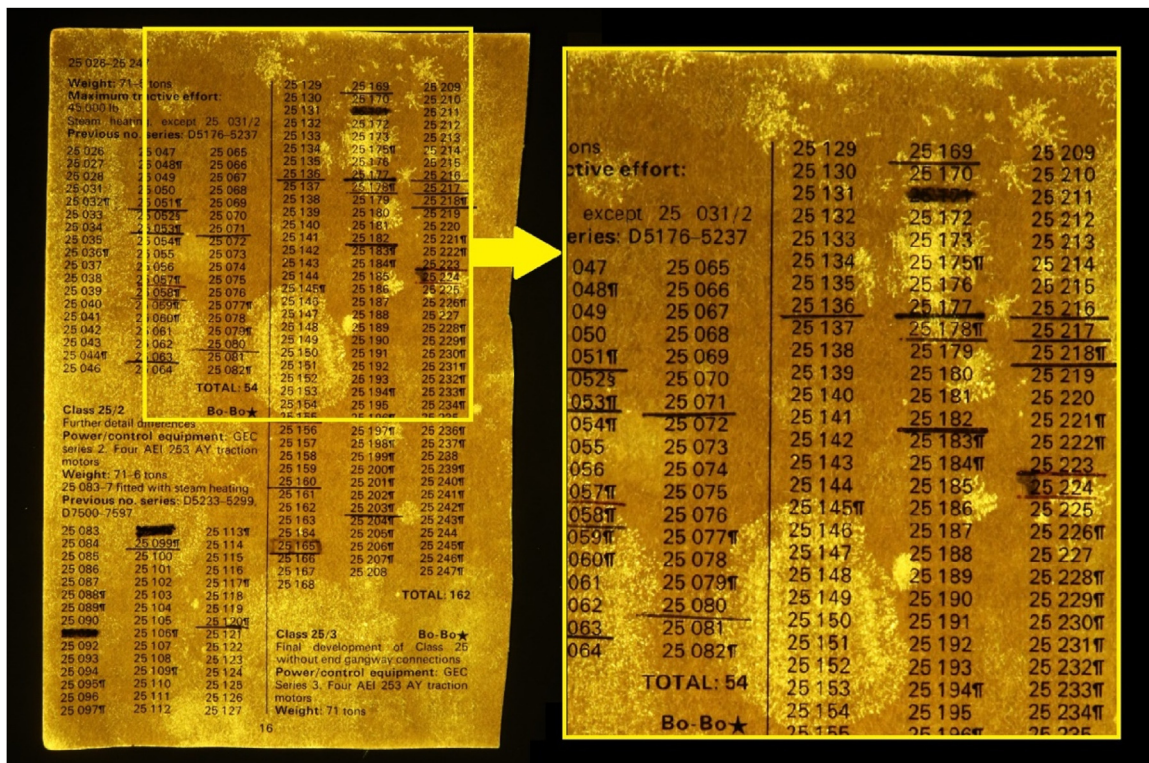


Fig. 6. Possible diffuse features possibly associated with original areas of contact, developed with IND/Zn on a pre-printed notebook page dating between 1978-1980.

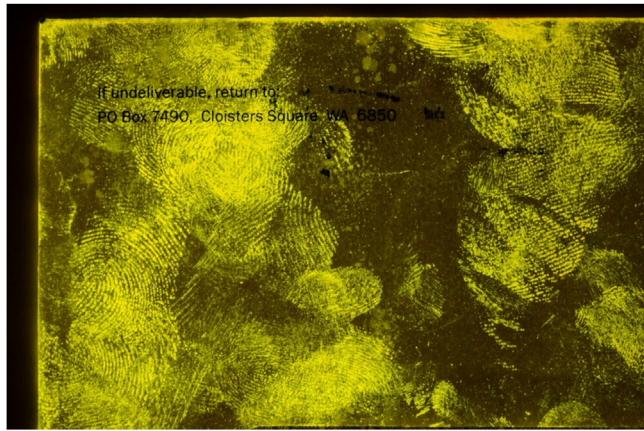


Fig. 7. Example of latent fingerprints developed with IND/Zn on a contemporaneous envelope.

been exposed to cyclic temperature and humidity conditions that were more extreme than those in the prior study. It is possible that these environmental conditions, in conjunction with differences in the paper type or porosity, make diffusion of amino acids more likely.

On several of the cards and thicker pre-printed notebook pages, it was observed that features which had possibly been ridge detail were developed as diffuse “dendrite-like” features without any clear ridge detail present (Fig. 6). Similar features were also been found on documents of similar age treated under Australian conditions indicated by the arrow in Fig. 4. It is known that fingerprint absorption into paper substrates is influenced by the smoothness of the paper, with smoother paper types exhibiting less absorption [9]. It is possible that the amino acids remaining closer to the surface are more susceptible to diffusion when exposed to cyclic conditions of temperature and humidity. The relatively lower effectiveness on Christmas cards may also be attributed to the cards being more vulnerable to environmental exposure compared to pages in a closed book, excluding cards in the middle of a bundle which potentially receive similar protection as the latter. Additionally, the natural moisture content and the natural ageing of the paper may have a combined effect on the diffusion mechanism. It has been demonstrated that various cycles condition of humidity and temperature changes the hydrogen bonding network in paper, as well as its capacity for moisture absorption and desorption [24]. These physicochemical changes over time are likely to affect the diffusion of water-soluble components of latent fingerprint residue, such as amino acids.

3.5. Contemporaneous documents

IND/Zn was highly effective in developing fingerprints on contemporaneous documents. All documents gave visible fingerprints, with 95 % showing potentially identifiable fingerprints. An example of the development obtained is shown in Fig. 7. Although some overlapping and smudging was evident, several marks with high intensity ridge details were obtained and no substantial diffusion features were observed.

3.6. General trends observed across the three ageing periods

As can be seen from the summary of the characterisation of documents in Fig. 8; fewer and lower quality marks were detected on increasingly older documents. The percentage of documents with potentially identifiable fingerprints decreased from 95 % to 70 % and 15 %. Conversely, the proportion of documents in which no fingerprints were observed increased from 5 % to 10 % and 25 % for contemporaneous, 35–40 year old and 75–90 year old documents respectively. It is suggested that whilst amino acid are well preserved in the paper matrix in contemporaneous documents, exposure to cyclic humidity and temperature over time can cause diffusion. It is known that other water-soluble components such as urea and chlorides migrate faster under high relative humidity [11], although this process would be expected to occur at a slower rate for amino acids. Other parameters such as the natural moisture content of paper may also affect the rate of diffusion. This warrants further study into factors that can influence diffusion, including the natural moisture content and the ageing mechanisms of the paper.

Several regions that may originally have shown identifiable ridge detail were developed as dendrite-like diffuse features on the 30–45 year old documents and background luminescence on the 75–90 year old documents; neither of which were observed on the contemporaneous documents. The possibility that these features indicate different stages of the diffusion process can be inferred. However, further study is needed to investigate any potential relationship between the features observed and the diffusion process.

Although similar general trends were observed between the two geographical locations, the success rate across the different ageing periods was higher for documents processed in Australia compared to the UK. The difference in the climatic conditions exposed by the documents was suggested as the likely reason. Additionally, the use of different IND/Zn formulations to treat documents based on its geographical location may also influence the results. However, no major differences were observed in the effectiveness of the German and Australian formulations in detecting latent fingerprints on documents exposed to similar environmental conditions across the three ageing periods (Fig. 9). The

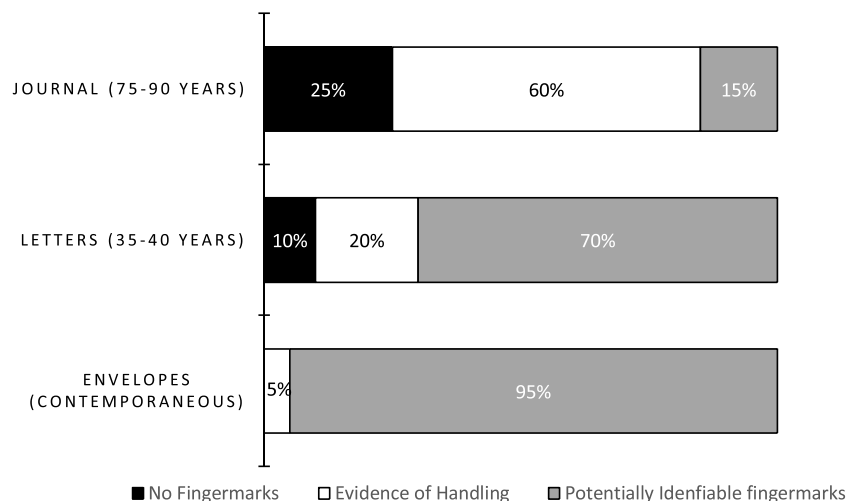


Fig. 8. Percentage results of the characterisation of documents of different ageing periods based on the quality of fingerprints developed after treatment with IND/Zn under Australian climatic conditions.

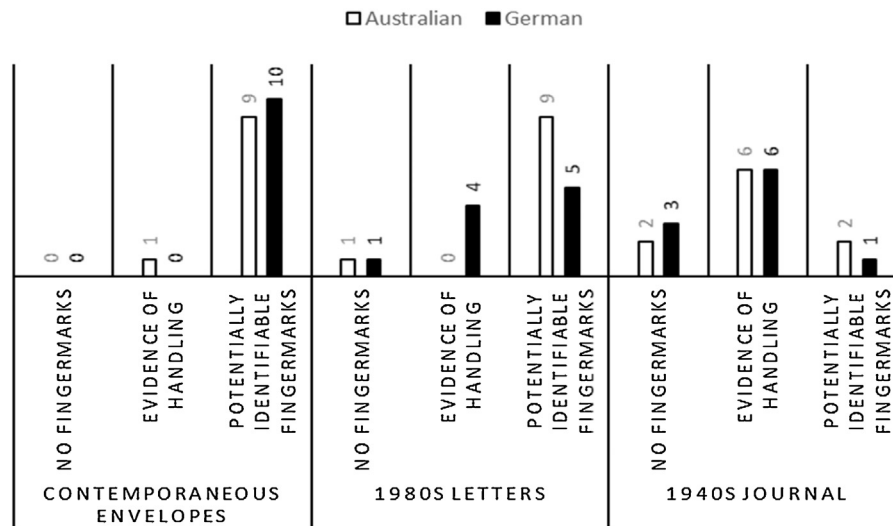


Fig. 9. Comparison between Australian and German IND/Zn formulations based on the best quality fingermarks developed on three sets of documents of different ageing periods.

result seems to suggest that the variation in IND/Zn formulations have a minor influence on the success rate. However, these findings were based on a limited sample size. A more comprehensive research on the effectiveness of the three IND/Zn formulations on various substrate is currently ongoing.

4. Conclusion

This study has shown that IND/Zn is a highly effective treatment for historic fingermarks on old documents. For the first time, we have shown the development of fingermarks on up to 80 year old documents, significantly increasing the established timescale on the detectable lifetime of fingermarks using amino acid sensitive reagents on paper. Its application has particular relevance in cold case reviews involving examination of historic fingermarks on porous evidence.

The current findings reinforce the suggestion of progressive diffusion of amino acids with time on paper surfaces. A greater proportion of potentially identifiable fingermarks were found on contemporaneous documents (95 %) compared to older documents (15 %). IND/Zn was also found to be less effective on documents or parts of documents that have been more exposed to cyclic temperature and humidity. Furthermore, the difference in climatic conditions appears to be more influential on fingermark development than the formulation used. Exposure to environmental factors is likely to accelerate progressive diffusion of amino acids, though the natural moisture and ageing of paper may also be influential. Therefore, the exposure and storage conditions and type of paper are some of the important parameters to be considered when assessing the success rate in developing historic fingermarks on old paper.

Altogether the results indicate that IND/Zn is highly effective in developing historic fingermarks but the quality is likely to be in part dependent on the extent of amino acid diffusion. This warrants further study into factors that can influence diffusion, including the environmental factors and the natural moisture content and the ageing mechanisms of the paper. The current investigation was limited to a small subset of documents. Further investigation needs to consider a larger number and type of paper substrate.

Declaration of Competing Interest

GS is a Section Editor of Forensic Science International: Reports. The authors declare no other competing interests.

CRediT authorship contribution statement

Jemmy T. Bouzin: Conceptualization, Investigation, Methodology, Visualization, Writing - original draft, Writing - review & editing. **Jason Merendino:** Investigation, Writing - review & editing. **Stephen M. Bleay:** Conceptualization, Investigation, Methodology, Writing - review & editing. **Georgina Sauzier:** Conceptualization, Supervision, Writing - review & editing. **Simon W. Lewis:** Conceptualization, Methodology, Project administration, Supervision, Writing - review & editing.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.fsir.2020.100145>.

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