Establishing evidence bases for crime reconstructions: Experimental studies on the recovery, transfer, and persistence of forensic evidence

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Declaration

I, Mark Andrew Amaral confirm that the work presented in this thesis is my own. Where information has been derived from other sources, I confirm that this has been indicated in the thesis.

Signed: ……

21st December 2021

Acknowledgments

"Success is falling nine times
and getting up ten" – Jon Bon Jovi

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Abstract

The focus of this research lies within the context of the forensic process and addresses a current debate within the literature for the importance and necessity of a growing body of empirical research to inform each stage of that process.

This thesis presents three experimental studies addressing the recovery, transfer, and persistence of forensic traces. First, a novel gelatine-based collection medium was created, and a sampling method validated, for recovering explosive and drug residues from a wide range of porous and non-porous surfaces. Second, the first reported use of Instron's ElectroPuls for application to forensic science is also presented, employing a reductionist approach to evaluate the individual impact of force, time, and rotation on the transfer of explosive and drug particulates. Third, a comparison of the dynamics of drug particulates on paper and polymer banknotes are presented, assessing the implications this might have on crime reconstruction approaches as more countries adopt polymer banknotes as legal tender.

Based on the results, this thesis presents an effective method for inclusion in the tool kit which investigators can rely upon when tasked with the forensic collection and recovery of trace particulates. Additionally, the findings indicate that there is value for broader crime reconstruction endeavours in taking a reductionist approach when seeking to understand the mechanics of trace transfers. This can assist in creating simulation models where specific parameters can be adjusted for a given case in which the transfer of forensic materials may have occurred. Such datasets are valuable for modelling the movements of traces to enable more transparent and reproducible interpretations of pertinent trace materials in crime reconstructions.

Impact Statement

The work presented herein supports and contributes to a research culture in forensic science that is grounded in reality and provides solutions which are implementable in practice. Since 2015, the Forensic Science Regulator in the United Kingdom has consistently highlighted the need for empirical studies addressing the transfer and persistence of trace materials. As such, the experimental studies presented in this thesis were designed to contribute to an evidence base needed to underpin the interpretation of forensic materials. This is fundamental in enabling robust reconstructions of crime events. Before the behaviour of materials can be assessed, traces must first be identified and successfully recovered. Accordingly, the creation of a novel gelatine-based sampling medium formed the starting point for this thesis. This media provides a highly cost effective and easily available tool for the recovery of trace particles from a wide range of surfaces. The second part of this thesis explored the value of utilising a reductionist methodology not typically employed in forensic science research to address the challenge of incorporating an evidence-based understanding of trace evidence dynamics in crime reconstruction approaches. Therefore, this work set out to investigate this complementary approach to understand the mechanics of evidence transfer and persistence. It is anticipated that the findings of this work will be relevant to academics by providing avenues for further research and exploration as well as by practitioners who are tasked with the collection and interpretation of forensic materials. Additionally, aspects of this work have been disseminated in peer-reviewed journals and presented at a number of international forensic science conferences.

Abbreviations

%T	percentage transferred
ABS	acrylonitrile butadiene styrene
ACN	acetonitrile
ANOVA	analysis of variance
Avg.	average (arithmetic mean)
BOPP	biaxially oriented polypropylene
CAI	case assessment and interpretation
Conc.	concentration
CV	coefficient of variation
DNA	deoxyribonucleic acid
EPA	Environmental Protection Agency
FID	flame ionization detector
FSR	Forensic Science Regulator
GC	gas chromatograph
HCl	hydrochloride
HPLC	high-performance liquid chromatography
LC	liquid chromatography
MDF	medium-density fibreboard
MeOH	methanol
NAS	National Academy of Science
PETN	pentaerythritol tetranitrate
RT	retention time
SD	standard deviation
SPE	solid-phase extraction
TNT	2,4,6-trinitrotoluene
UFLC	ultra-fast liquid chromatograph
UK	United Kingdom
US	United States
USD	United States dollar
UV	ultraviolet
UV-Vis	ultraviolet/visible light detector
VND	Vietnamese đồng

Units

°	degree
µL	microlitre
µm	micrometre
Å	ångström
C	Celsius
cm	centimetre
cm^2	square centimetre
cm^3	cubic centimetre
g	gram
kN	kilonewton
kPa	kilopascal
L	litre
m	metre
mg	milligram
min	minute
mL	millilitre
mm	millimetre
mV	millivolt
N	newton
nm	nanometre
rpm	revolutions per minute
s	second

Glossary

CALIBRATION CURVE

A linear regression model used to predict an unknown concentration of a given analyte based upon the response of the instrument to a series of known standards. The measured signals (***y***) of known standards are plotted against the corresponding concentration (***x***) of the standard and a line of best fit (least square method) fitted to the data. The model equation is: $\boldsymbol{y} = m\boldsymbol{x} + b$, where ***m*** is the slope and ***b*** is the *y*-intercept. The concentration of an unknown sample (***x***) is determined by: $\boldsymbol{x} = \frac{(\boldsymbol{y} - b)}{m}$, where ***y*** is the measured signal obtained for an unknown sample.

CHROMATOGRAPHY

A technique employed to separate a given mixture into its individual components. This is achieved through the differential partitioning of individual components of a mixture between a mobile and stationary phase. The mixture to be separated is dissolved in a mobile phase, such as an inert gas (gas chromatography) or a solvent (liquid chromatography) and carried through a stationary phase (column). The time it takes for each component to pass through the column is used to determine its identity and can also be used to quantify how much of that component is present in the mixture.

ELECTROPULS™ E3000

An electrodynamic test instrument produced by Instron® and designed for dynamic and static testing of a wide array of materials. Operation is controlled through a series of programmable wave functions that allow for both linear and rotary motion. With common application to biomedical and biomechanical research, it allows for the fatigue testing of materials (establishing the lifespan that may be expected from a material under certain conditions). Specifically, the impact of tension, compression, bending, torsion, or combinations of these stresses on material fatigue can be assessed. This thesis presents the first reported use of this instrumentation for application to forensic science.

FLAME IONIZATION DETECTOR (FID)

A sensitive gas chromatographic detector used to measure the concentration of organic species, compounds that contain carbon atoms, in a gas stream. The detector creates a hydrogen flame which produces electrically charged particles (ions) as the sample is burned. These ions are collected and the resultant electrical signal measured. The output is a graph where time (min) is on the x-axis and signal intensity (mV) is on the y-axis.

HYDROGEL

Hydrogels are comprised of three-dimensional (3D) cross-linked polymeric networks. Due to the presence of hydrophilic functional groups attached to the polymer backbone, hydrogels can absorb large quantities of water within the spaces available among the polymeric chains. Water absorbed by the hydrogel allows for the free diffusion of solute molecules. The amount of water absorbed by the hydrogel, or total water holding capacity, is dictated by the cross-linking density. As cross-linking density increases, there is a decrease in equilibrium swelling as a result of a decrease in the hydrophilic groups.

SOLID-PHASE EXTRACTION (SPE)

A technique used to extract compounds from sample matrices prior to chromatographic analysis. In solid-phase extraction, one or more analytes from a liquid sample are isolated by extracting, partitioning, and/or adsorbing onto a solid stationary phase. The stationary phase (a sorbent or resin) binds either the analyte, or impurity, through strong but reversible interactions to extract the analyte of interest reliably and rapidly from a complex matrix.

ULTRAVIOLET/VISIBLE LIGHT DETECTOR (UV-VIS)

A liquid chromatographic light absorbing detector which focuses on the ultraviolet (UV) and visible regions of the spectrum; within the 190 – 900 nanometre (nm) wavelength range. The detector is first set to a wavelength that is absorbed by the analyte(s) of interest. As the sample passes through the detector, the absorbance is measured, producing a continuous signal that can be used to quantify the amount of chromophoric compounds (fragments of a molecule that interact with light) present.

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Chapter 1

Introduction

The simple reality is that the interpretation of forensic evidence is not always based on scientific studies to determine its validity. This is a serious problem. Although research has been done in some disciplines, there is a notable dearth of peer-reviewed, published studies establishing the scientific bases and validity of many forensic methods. (National Research Council, 2009; p. 8)

1.1 Establishing Scientific Bases in Forensic Research

Forensic science has found itself "in a state of crisis, presenting a threat of undermining trust in the criminal justice system" (House of Lords Science and Technology Select Committee, 2019; p. 9). The spark that ignited the proverbial 'powder keg' was a 2009 report issued by the National Academy of Science (NAS) which highlighted a lack of empirical evidence underpinning the foundation of many forensic methods (National Research Council, 2009). This report came to fruition after the United States (US) Congress instructed the NAS to review the status of forensic techniques used in criminal prosecutions. Upon review, instances of malpractice, failures of quality standards, and lack of training were raised as key issues that needed to be addressed (National Research Council, 2009). When forensic science fails, it can have serious ramifications resulting in wrongful convictions and miscarriages of justices. This has been exemplified by the Innocence Project, a non-profit legal organisation, who found that the misapplication of forensic science contributed to 45% of DNA exoneration cases (Innocence Project, 2017). Furthermore, the Federal Bureau of Investigation in the US identified that hair evidence had been either overstated, or misinterpreted, in over 90% of the cases reviewed (Federal Bureau of Investigation, 2015). In the United Kingdom (UK), it was found that one in every five cases upheld by the Court of Appeal, had evidence that was misleading, largely due to misinterpretation (Smit *et al.*, 2018).

The "[m]isinterpretation of forensic evidence is the biggest challenge facing forensic science" (TED Archives, 2018; 3:30) and an understanding of what the evidence means is pivotal in the delivery of justice. It is not enough to be able to detect trace particulates left behind during the commission of a crime, but rather it is crucial to be able to interpret what those traces mean in the context of a crime

reconstruction (Morgan, 2019). To be successful, there needs to be a research culture in forensic science, one that is “grounded in the values of empiricism, transparency, and a commitment to an ongoing critical perspective” (Mnookin *et al*., 2011; p. 726). As such, the interpretation of forensic science evidence should be supported by empirical research rather than conclusions being based solely upon craft knowledge, experience, and longstanding use (Saks & Koehler, 2005; Mnookin *et al.*, 2011; Koehler & Meixner, 2016). The annual report issued by the Forensic Science Regulator (FSR) has consistently highlighted the need to understand trace evidence dynamics to assist in answering how and when a given particulate transferred and under what conditions and timeframes they persist (Forensic Science Regulator, 2015, 2017, 2018, 2019). To address this, structured empirical studies are required which establish scientific bases to underpin the evidence dynamics of trace materials (Morgan *et al.*, 2020). This therefore informed the research aims of the work presented herein.

1.2 Research Aims

This thesis focuses on the recovery, transfer, and persistence of forensic materials on porous and non-porous surfaces. The aims set out were threefold:

- first, to address the poor recovery of forensic materials from porous surfaces by exploring the use of a novel hydrogel formulation (Chapters 3 & 5);
- second, to expand upon an empirical evidence base through implementing a reductionist approach to understand the mechanics of how a particular form of trace may have been transferred (Chapters 4 & 5);
- third, to offer insights into how a certain activity can impact upon the persistence of a given trace (Chapter 5).

To address these aims, three experimental studies were undertaken (Chapters 3–5). Explosive and drug residues were the focus of these studies due to a notable dearth of published literature addressing the evidence dynamics of these particulates. The results from these studies can indicate the best approaches for the collection of such trace material and the importance of the context that such data provide for effective inferences to be made. The wider implications of these results for the interpretation and presentation of expert evidence in court can also be assessed.

1.3 Thesis Outline

This thesis is comprised of seven chapters (**Figure 1.1**). The first two chapters provide the context for work carried out. This is followed by three chapters pertaining to the experimental work conducted. The last two chapters discuss the ramifications of the findings.

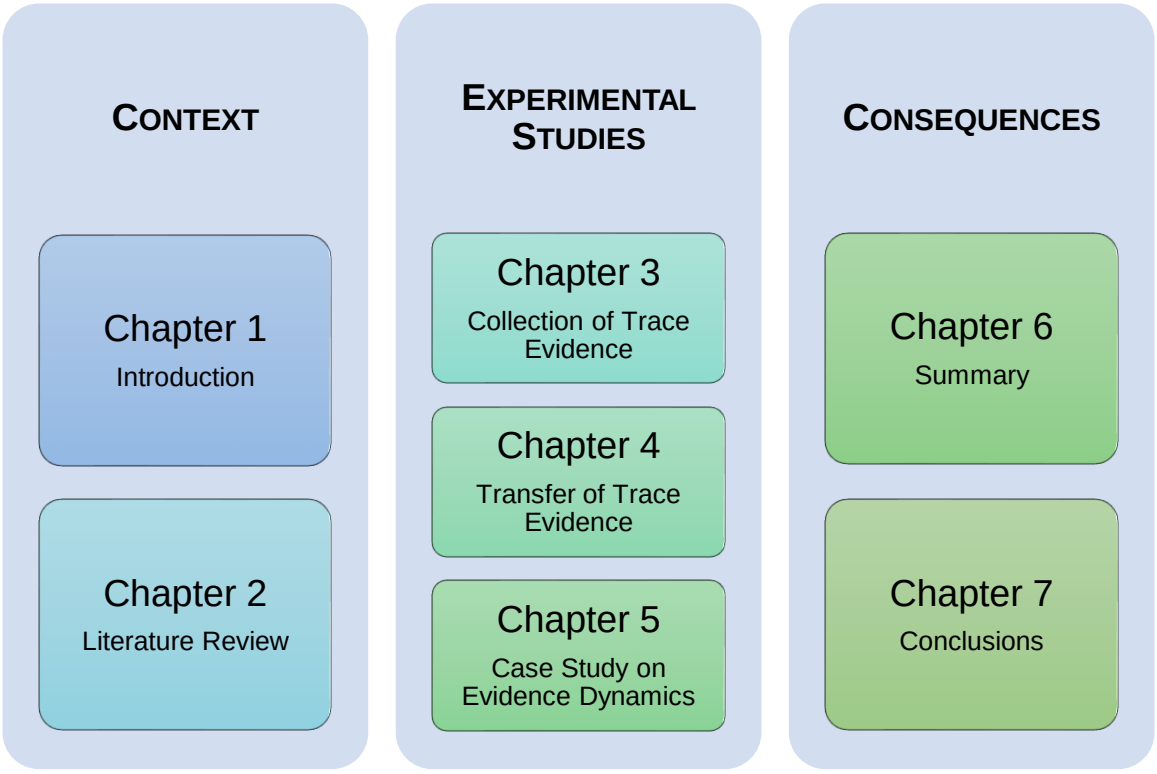


Figure 1.1. Thesis outline.

Chapter 1 – Introduction

This introductory chapter provides the context for the empirical research presented in this thesis by highlighting the necessity of scientific bases to underpin the evidence dynamics of forensic materials, a key requirement for enabling robust crime reconstructions. Fundamental to this is the successful recovery of forensic materials along with understanding the mechanics of how these materials transfer and under what conditions and timescales they persist. The aims are also presented which are formalised into research questions in Chapter 2 and informed the development of the three experimental studies (Chapter 3–5). Finally, a list of publications and presentations are included where aspects of this thesis have been disseminated.

Chapter 2 – Literature Review

The literature review expands upon the context outlined in Chapter 1 by: (1) defining the scope of forensic science; (2) providing an overview of the current landscape of forensic science research; (3) highlighting the importance of situating evidence within a framework for interpretation; (4) outlining the role of trace particulates in crime reconstructions; and (5) identifying the current gaps in the knowledge base. Application of a reductionist approach to understanding the mechanics of transfer and persistence at the activity level is discussed, along with the potential use of hydrogels as a method of trace particulate recovery. This section concludes with a summary and presentation of the research questions which form the basis of the three experimental studies (Chapters 3–5).

Chapter 3 – Collection of Trace Evidence: A Novel Gel Formulation

This chapter presents the first experimental study on the collection of ammonium nitrate, an explosive compound, from a variety of porous and non-porous surfaces using a novel gelatine-based hydrogel formulation. Within this chapter, the criteria for a recovery medium for evidence collection is outlined and the preparation of the hydrogel detailed. Following the recovery of particulates from a given surface, an extraction method to liberate ammonium nitrate from the hydrogel matrix is also presented. The recovery efficiency of the hydrogel is assessed with respect to the different surface types and the avenues for future work discussed. Chapter 5 expands upon the application of this technique as a recovery medium for other forensic materials, namely, drug residues.

Chapter 4 – Transfer of Trace Evidence: A Reductionist Approach

The adoption of a reductionist approach to understand the transfer of explosive residues is explored in this chapter. A bespoke application of an Instron ElectroPuls E3000 for forensic research is presented, affording independent control over the force of contact, duration of contact, and amount of rotation applied. Additionally, the repeatability and reproducibility of the contact parameters is demonstrated with respect to the method employed. The of amount of ammonium nitrate transferred from a porous and non-porous surface onto a human skin analogue as a function of each of the three variables is provided and

discussed. This approach and method are developed further in Chapter 5 to assess the impact of these variables on the transfer of cocaine residues.

Chapter 5 – Cocaine on Banknotes: Evidence Dynamics Case Study

Recent years have seen the adoption of polymer banknotes as legal tender to replace older paper notes in circulation. In response, this chapter evaluates the impact this will have on the transfer and persistence of cocaine residues, a known contaminate present on paper banknotes. Application of the novel hydrogel formulation presented in Chapter 3 is expanded upon, assessing the efficiency of the method in recovering cocaine residues from banknotes. As in Chapter 4, a reductionist approach was adopted to understand the mechanics of transfer, utilising the ElectroPuls E3000 to control the contact parameters (time, force, and rotation). Additionally, the persistence of cocaine residues on both banknote types following handling is presented. Finally, the evaluative interpretation in crime reconstruction is discussed.

Chapter 6 – Summary

This chapter synthesises the findings from the three experimental studies and outlines a methodological framework for framing and supporting a research culture moving forward. Two key themes are addressed with respect to solving problems and addressing pertinent issues in forensic science. First, solutions need to be implementable in practice and second, solutions need to be context focused. The work presented in this thesis posits that a reductionist and holistic approach are complementary, with both being required to understand the behaviour of forensic materials. Finally, avenues for further research are considered.

Chapter 7 – Conclusions

The final chapter of this thesis summarises the key research findings and conclusions with respect to the aims put forth in Chapter 1 and research questions outlined in Chapter 2. The implications of the findings with respect to the interpretation of evidence dynamics at the activity level and crime reconstruction are also provided.

1.4 Publications

Aspects of this PhD thesis have been published in the following:

Amaral, M.A., Gibson, A.P., & Morgan, R.M. (2022). Trace evidence dynamics of cocaine on banknotes: A comparison study of paper and polymer banknotes. *Science and Justice, 62*(2), pp. 221–228. DOI: 10.1016/j.scijus.2022.02.008

Amaral, M.A., Hatten, E., Gibson, A.P., & Morgan, R.M. (2022). The impact of force, time, and rotation on the transfer of ammonium nitrate: A reductionist approach to understanding evidence dynamics. *Science and Justice, 62*(2), pp. 129–136. DOI: 10.1016/j.scijus.2021.12.006

Amaral, M.A., Yasin, S., Gibson, A.P., & Morgan, R.M. (2020). Sampling of explosive residues: The use of gelatine-based medium for the recovery of ammonium nitrate. *Science and Justice*, *60*(6), pp. 531–537. DOI: 10.1016/j.scijus.2020.07.007

Additional publications undertaken at the same time as this PhD research:

Moorat G., Reed., J., **Amaral, M.A.**, Chappell, B., Pamment, N., Plowman, C., & Smith, P.A. (2020). The visualisation of fingermarks on Pangolin scales using gelatine lifters. *Forensic Science International*, *313*, pp. 1–10. DOI: 10.1016/j.forsciint.2020.110221

1.5 Conference Presentations

Facets of this PhD thesis have been presented at the following conferences:

- The Australian and New Zealand Forensic Science Society (ANZFSS) 23rd International Symposium on the Forensic Sciences. Auckland, NZ. Keynote presentation: *The persistence of trace explosive residues on fabrics*. 21st September 2016.
- Forensics in Defence and Security Symposium, Cranfield Forensics Institute. Defence Academy, Shrivenham, UK. Poster presentation: *The persistence of trace explosive residues on fabrics*. 14th September 2016.
- 10th International Crime Science Conference, UCL Jill Dando Institute of Security and Crime Science. British Library, London, UK. Poster presentation: *Explosive residues on fabrics*. 12th July 2016.

Chapter 2

Literature Review

Trace evidence is highly complex, and this complexity needs to be incorporated into any reconstruction approach. Forensic evidence is complex due to the nature of traces (its different forms, capabilities, ability to infer source/activity levels, and the interaction of different forms of trace with one another), and the integral part human decision-making plays in evaluating trace materials.
(Morgan, 2017a; p. 456)

Introduction

'Every contact leaves a trace'. During the commission of a crime, a perpetrator will transfer some material, through contact, onto the surfaces and objects they encounter in the environment (Locard, 1920). The analysis and characterisation of this material can therefore provide valuable insights that can aid in the reconstruction of a given crime event (Morgan *et al*., 2020). As such, understanding the trace evidence dynamics of forensic materials has been highlighted as a priority for research (Forensic Science Regulator, 2015, 2017, 2018, 2019). Specifically, understanding the transfer and persistence of trace materials, as well as the successful recovery of these materials, is of great importance in understanding what happened at the activity level. Although the empirical evidence base on trace evidence dynamics has grown in recent years, there is a gap in the knowledge base concerning the behaviour of drug and explosive residues. Additionally, understanding the mechanics influencing the behaviour of forensic materials through a reductionist approach has not been fully explored. Reductionism seeks to explain complex phenomena using the most basic principles, the adoption of which allows for the separation of individual variables within a specific scenario to establish the mechanics of what is being observed.

2.1 Forensic Science

Forensic science is an applied science; that is, it implements the scientific method and processes to aid in the investigation and prosecution of crime (Chisum & Turvey, 2011a). In essence, it is an interdisciplinary approach, drawing from a diverse range of disciplines to provide insights which can be applied to questions relating to the law (Morgan, 2017a). Forensic science is therefore commonly defined as the application of science to the law and criminal justice system.

Deviating from this ‘traditional’ law-oriented definition, Ribaux *et al.* (2010), defined forensic science as the “study of traces, which themselves are present as remnants of an activity, most often a criminal activity” (p. 10). More recently, the term traceology has been formalised, defined as the “study of event traces created during an event, which encompasses the detection, recognition, identification, process of individualization toward source attribution, and evaluation of the physical record created” (Ristenbatt *et al*., 2021; p. 2). Irrespective of the verbiage used to define forensic science, the goal of forensic science is to gain a better understanding of a criminal event through the scientific examination and analysis of physical evidence, as well as providing insight into offender characteristics and criminal behaviour (Julian *et al.*, 2011; Roux *et al.*, 2015). As such, forensic science is an integral component of the criminal justice system which is increasingly relied upon by law enforcement to solve crime and by the judicial system to prosecute offenders and exonerate the innocent (Julian *et al.*, 2011; Sapir, 2020).

2.2 A Paradigm Shift in the Forensic Sciences

With the advent of DNA profiling in forensic science, underpinned by reproducible methodological research in DNA typing theory, Saks and Koehler (2005), predicted, “a paradigm shift in the traditional forensic identification sciences in which untested assumptions and semi-informed guesswork are replaced by a sound scientific foundation and justifiable protocols” (p. 895). The necessity of this shift was further highlighted in February 2009 when the NAS in the US issued a report stating that the forensic science community had failed to deliver on the standards necessary to meet its responsibilities. The crux of the problem was identified to be a “dearth of peer-reviewed, published studies establishing the scientific basis and validity of many forensic methods” (National Research Council, 2009; p. 8). Giannelli (2010), postulated that “the reason for the lack of empirical research was simply a stubborn refusal to reconsider beliefs in light of credible challenges” (p. 517). One could argue that the legal system also shoulders some of the blame, previously having accepted forensic testimony based on these methods. As such, the forensic community may not have perceived they needed to change the way things operated. Regardless, this is the antithesis of how science should be conducted and a major cause for concern, substantiating that the *status quo* needs to be challenged.

In addition to the NAS's report, the Law Commission (2011), raised issues of admissibility based on validity, noting that "…special rules are required for assessing the reliability of expert evidence as a factor bearing on admissibility, and that opinion evidence with insufficient indicia of reliability (that is, pointers to reliability) ought not to be admitted in criminal proceedings (paragraph 1.11)." This was echoed in the US by the President's Council of Advisors on Science and Technology (2016), who concluded that there are two important gaps that need to be addressed. The first is "the need for clarity about the scientific standards for the validity and reliability of forensic methods (p. 1)" and the second, is "the need to evaluate specific methods to determine whether they have been scientifically established to be valid and reliable (p. 1)". As such, forensic science has found itself in a precarious state and therefore must embrace a paradigm shift if it is to be fit for purpose and meet its responsibilities.

Despite the criticisms raised in these authoritative reports, quality standards for forensic laboratories have remained largely inconsistent (Widener & Drahl, 2014). Funding to implement improvements has also been scarce, with funding cuts seen across universities, the justice system, and police (Forensic Science Regulator, 2018; Cookson, 2019; House of Lords Science and Technology Select Committee, 2019). This has made addressing these issues more challenging. In addition, forensic science has also encountered setbacks with high-profile miscarriages of justice (both in the UK and overseas) (Federal Bureau of Investigation, 2015; Smit *et al*., 2018), along with reports of malpractice in accredited labs (Trager, 2014, 2018). At a time when arguably forensic science needed the most support, governments withdrew their support, as exemplified by the closures of the Forensic Science Service in the UK in 2012 (Ludwig, 2016) and the termination of the National Commission for Forensic Science in the US in 2017 (Bell *et al.*, 2018).

In 2015, The Royal Society hosted a two-day discussion meeting entitled 'The paradigm shift for UK forensic science', which brought members of the forensic science community together to discuss and outline a plan moving forward (Black & Nic Daéid, 2015). While in agreement that empirical research is needed across all forensic science domains, the implementation of this is plagued by challenges due to significant funding cuts (Forensic Science Regulator, 2018). These

problems have persisted and in 2019, The House of Lords' Science and Technology Committee's reported that forensic science is in a state of crisis.

Forensic science has found itself at a juncture, conflicted between the practices of science and the practices of law (Bell *et al.*, 2018). On the one hand, there exists a legal system which has accepted methods based on historical precedent. On the other hand, there is science which requires empirical validation of its methods. To be successful moving forward, holistic reform is required, merely strengthening the existing system is not sufficient. "[I]ncorporating an awareness of the requirements of the law in its broadest sense, and embedding research into both practice and policy within forensic science, is arguably critical to achieving such an endeavour" (Morgan, 2017a; p. 455). An important aspect of this is to ensure that evidence is situated within the forensic science process and is bolstered by an empirical evidence base.

2.3 The Forensic Process (Crime Scene to Court)

Irrespective of the specific area of focus, a broad general conceptual framework underpinning forensic science emerged, structured around five basic concepts: transfer (Locard, 1920), identification (De Forest *et al*., 1983), individualisation (Kirk, 1963), association (Osterburg, 1968), and reconstruction (De Forest *et al*., 1983). Arguably, this framework served practitioners well for several decades, despite failing to provide a complete picture of the fundamentals involved, such as the origin and generation of evidence (Inman & Rudin, 2002). This conceptual framework was later formalised and expanded upon by Inman and Rudin (2002), incorporating the 'division of matter' as a sixth fundamental tenet. This tenant precedes the transfer of evidence to account for the fact that "matter must divide before it can be transferred" (Inman & Rudin, 2002; p. 12). The work of Inman and Rudin has since been built upon and this process reiterated in several ways (Morgan & Bull, 2007; Ribaux *et al*., 2010; Ribaux & Talbot Wright, 2014).

As it currently stands, the forensic process can be divided into six core tenets (**Figure 2.1**). These tenets outline a logical progression which begins with the origin of evidence and finishes at the presentation of evidence in court; from crime scene to court. This process is integral to the practice of forensic science in the attempt to answer the investigative questions: who, what, where, when, and how? (Inman & Rudin, 2001). Each step is reliant on the previous one being 'successful'

and therefore understanding where to look for evidence in a particular scenario is critical for generating evidence/intelligence for interpretation or presentation (Morgan & Bull, 2007).

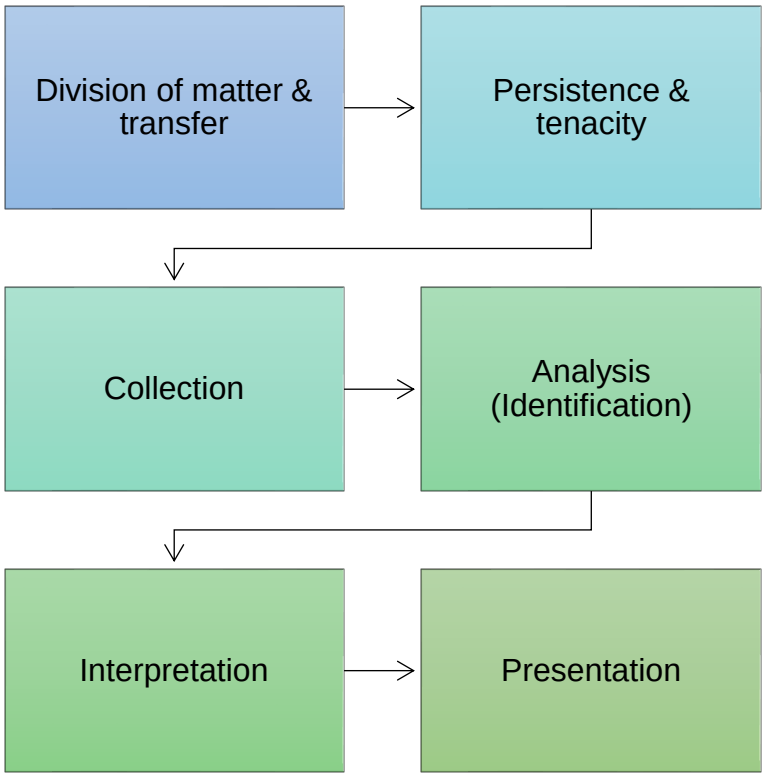


Figure 2.1. The six stages of the forensic science process as outlined by Morgan & Bull (2007).

The process begins with the division of matter, a fundamental principle in forensic science whereby the generation of physical evidence occurs when a sufficient force is applied to an object resulting in the fractionation of sediments from the parent body. According to Inman and Rudin (2002), “the component parts will acquire characteristics created by the process of division itself and retain physico-chemical properties of the larger piece” (p. 12). Once the physical evidence has been generated, it can then be transferred to other surfaces and objects. The transfer of evidence is founded upon the principle put forth by Edmond Locard which states that:

> [w]henever two objects come into contact, there is always a transfer of material. The methods of detection may not be sensitive enough to demonstrate this, or the decay rate may be so rapid that all evidence of transfer has vanished after a given time. Nonetheless, the transfer has taken place (Murray & Tedrow, 1991; p. 7).

Once transfer has occurred onto a surface, or object, the retention of the physical evidence becomes an important property. The works of Pounds and Smalldon (1975a,1975b, & 1975c), along with Robertson *et al*. (1982), were influential in highlighting the importance of understanding the persistence and tenacity of evidence. How likely the transferred material will be retained, as well as the rate at which it is lost from the surface, is important to understand for forensic inferences to be made.

Although the division and transfer of matter are not limited to forensic scenarios, the latter half of the process is unique to forensics; “the practice of forensic science begins after the crime event with the recognition of evidence” (Inman & Rudin, 2002; p. 16). Once a crime occurrence is suspected, investigators begin to search for pertinent evidence at the scene. For evidence to be collected, it must first be observed and recognised as potentially having evidentiary value. As such, effective detection methods and techniques are of paramount importance. This is especially true for trace evidence, which is not readily visible without some form of detection or enhancement (Roux *et al.*, 2015; Trejos *et al.*, 2020).

The next stage of the process involves the analysis of the collected materials. Forensic analysis can be considered as the process of identification and individualisation, with the former answering the investigative question of “what is it?” and the later “which one is it?” or “whose is it” (Inman & Rudin, 2001). For sound methodological research, forensic methods need to be validated, with each method assessed on a number of parameters including accuracy, precision, measurement uncertainty, matrix effects, interference, limit of detection, limit of quantification, linearity range, stability of measured compounds, specificity and selectivity, repeatability, reproducibility, and robustness (International Laboratory Accreditation Cooperation, 2014).

Following analysis of the collected material, the results must then be interpreted correctly in order for robust crime reconstructions to take place and to ensure that the evidence presented accurately in court (Morgan & Bull, 2007; Morgan, 2017a, 2017b). The interpretation of forensic evidence should be based on empirical data (Mnookin *et al.*, 2011; Government Chief Scientific Advisor, 2015; Koehler & Meixner, 2016), not supposition or hearsay (Innocence Project, 2019). The final step in the forensic process is the presentation of the results in the courtroom.

This thesis primarily focuses on the first three tenets, namely the transfer, persistence, and recovery (collection) of evidence, but also contributes to the other tenets succeeding this step as each tenant is reliant on the previous one being 'successful'. Understanding the transfer and persistence of forensic materials is paramount in crime reconstruction and operating within an interpretation framework is central to this process. In forensic science, the Case Assessment and Interpretation (CAI) model (Cook *et al.*, 1998a) provides such a framework for structuring examinations and evaluating evidence.

2.4 Interpretation Framework (Hierarchy of Propositions)

"[T]he essence of forensic science is the drawing of rational and balanced inferences from observations, test results and measurements" (Cook *et al.*, 1998a; p. 152). In order to assist with the interpretation of evidence, Cook *et al*. (1998a) proposed a CAI model. The CAI model provides an operating framework for interpreting forensic evidence through the inclusion and integration of a likelihood ratio in the assessment of the weight of evidence (Fenton *et al*., 2016; Williams & Maskell, 2021). This framework can assist the scientist in decision-making and establishing a strategy for examination and analysis as well as assessing the probative value of evidence in relation to a given case (Jackson *et al*., 2013).

To assess the weight of evidence, a scientist must frame at least two competing propositions to weigh against each other (Cook *et al*., 1998b; Evett *et al.*, 2000; Schaapveld *et al*., 2019). In the adversarial system (practiced in countries such as England and the US), these competing propositions represent the positions of prosecution and the defence (Fenton *et al*., 2014). The propositions that are addressed depend upon a number of factors, including the circumstances of the case, the availability of empirical data, observations that have been made, and the knowledge and expertise of the scientist (Morgan *et al*., 2020). By framing competing propositions to assess the weight of evidence, the scientist offers rational conclusions to aid the jury in deciding the ultimate issue of guilt or innocence. As such, a 'hierarchy of propositions' was proposed by Cook *et al.* (1998b). This 'hierarchy' outlines a logical structure for CAI to develop realistic, logical propositions to assist the court.

The hierarchy of propositions is comprised of three levels: source, activity, and offence (**Figure 2.2**) (Cook *et al*., 1998b; Evett *et al.*, 2000). Although the evaluation of forensic evidence can provide support for propositions at any level within the hierarchy (Taylor *et al*., 2016), from a judicial standpoint, the offence level is the definitive matter that needs to be proven (Cook *et al*., 1998b). The offence level is seen as the ultimate issue and, in an adversarial system, is for the jury to ultimately decide; forensic scientists do not operate at this level (Kokshoorn *et al*., 2017). As the offence level incorporates both source and activity level questions, "[a] clear understanding of … [forensic findings] within the hierarchy is a first step toward understanding their potential impact on the ultimate issue" (Schiffer & Champod, 2008; p. 47).

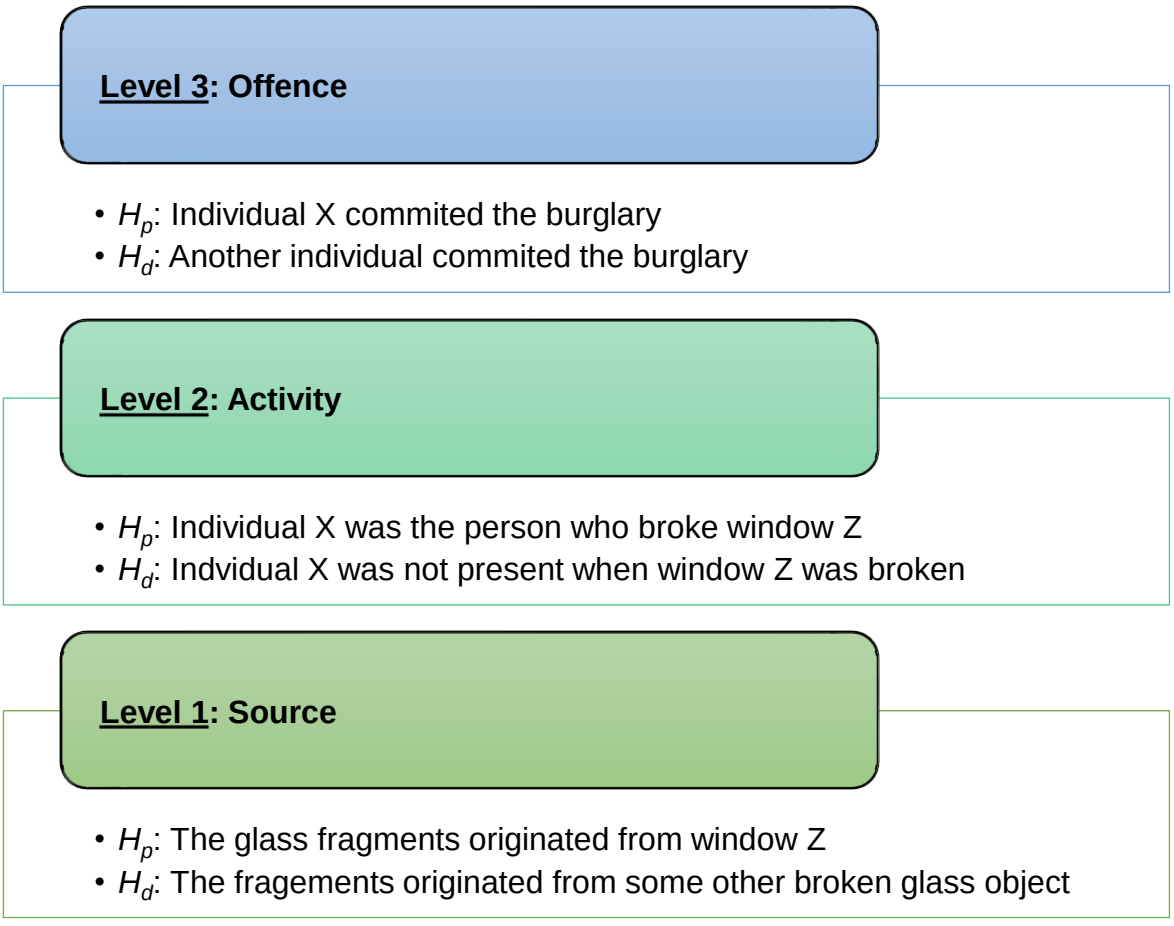


Figure 2.2. Hierarchy of propositions with exemplar prosecution (H_p) and defence (H_d) propositions for glass evidence at each level.

In many instances, such as DNA analysis, there is a focus on the source of the recovered material (Biedermann *et al*., 2016); however, "questions in court are shifting from identity to transfer mechanism" (Volgin, 2019; p. 10). To assist the

jury in reaching a decision, forensic evidence supporting higher-level propositions, the activity level questions, are of greater assistance to the court when deliberating over someone's guilt or innocence (National Institute of Forensic Science, 2017; Gill *et al.*, 2020).

Smit *et al*. (2018), analysed a sample of successful appeals and reported that 22% of these cases involved the misinterpretation of evidence, with "the majority (66%) of misleading evidence types relating to their interpretation at the activity level" (p. 128). This illustrates the necessity to understand trace evidence dynamics of forensic materials, including their value and limitations, when addressing activity level propositions. As such, the two experimental studies outlined in Chapters 4 and 5 contribute to an empirical evidence base which can be used to address activity level propositions. Through an understanding of the transfer and persistence properties of different forensic materials, we can assist the court with answering 'how' and 'when' a given trace may have been deposited. Additionally, unlike source level propositions, activity level propositions can also be used to assess the probative value of the absence of evidence.

The interpretation of forensic evidence should be conducted using reference to data from published studies (Mnookin *et al.*, 2011). According to the International Laboratory Accreditation Cooperation (2014), interpretation should be based on findings that are both reproducible and robust, caveats the findings based on the limitations of the study, and considers other scientific literature. As such, it is paramount that research-based knowledge generated from experimental studies dictates forensic practices.

2.5 The Role of Experimental Studies within Forensic Science

Establishing the significance of forensic materials relies upon having an evidence base, compiled from experimental studies, that can support source, activity, and offence level propositions (Evett, 2015; Biedermann & Hicks, 2016; Morgan *et al.*, 2020). Traditional forensic identification sciences are rooted in the unfounded assumption of discernible uniqueness which asserts that "markings produced by different people or objects are observably different" (Saks & Koehler, 2005; p. 892). The lack of empirical and theoretical foundations used to base such assumptions on has drawn numerous criticisms (National Research Council,

2009; Law Commission, 2011; Centre for Forensic Science and Medicine, 2013; President's Council of Advisors on Science and Technology, 2016), emphasising the need for experimental studies across all forensic science domains.

To overcome these criticisms and establish itself as a credible academic discipline, forensic science must be rooted in a theoretical framework that is both intellectually defensible and validated through rigorous research. Forensic questions should be answered using reference to data from published studies, incorporating pertinent empirical data and statistics, rather than primarily by reference to experience or craft knowledge (Saks & Koehler, 2005; Mnookin *et al.*, 2011; Koehler & Meixner, 2016; Morgan, 2017a, 2017b). As such, it is paramount that research-based knowledge generated from empirical studies dictates forensic practices and informs evidence interpretation.

Experimental studies in forensic science typically take two approaches: primary level experiments which provide a fundamental knowledge base around the generation, transfer, and persistence of forensic materials and secondary level experiments which incorporate variables pertinent to a given crime event or scene (Morgan *et al*., 2009a). Many of these studies have adopted holistic experimental designs that seek to mimic forensically relevant scenarios. However, such approaches are unable to separate individual variables within a specific scenario and establish the mechanics of what is being observed. As such, the work presented in this thesis employed a reductionist approach, one that seeks to assess each variable independently in a controlled and repeatable manner. Through understanding the mechanics of individual variables in a complex system, the behaviour of forensic materials can be better understood. Although often seen as disparate, reductionism and holism (**Figure 2.3**) are interdependent and complementary, with both approaches required to strengthen the scientific evidence base upon which we make inferences and conclusions.

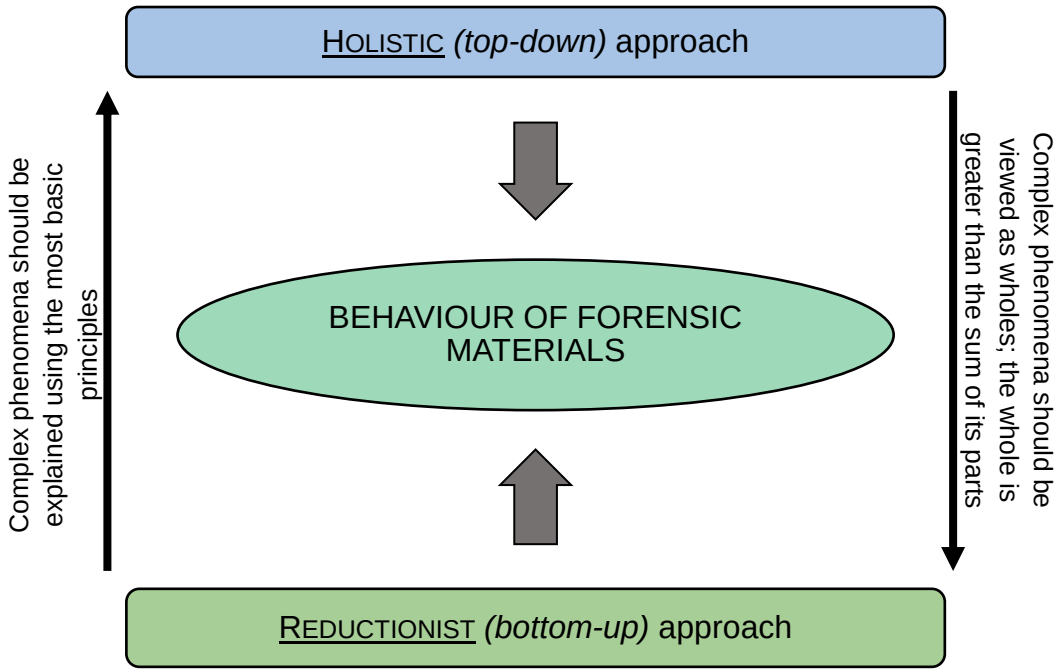


Figure 2.3. Holistic and reductionist approaches to understanding the behaviour of forensic materials.

The establishment of empirical evidence bases is also important for the admissibility of expert evidence in court (The Law Commission, 2011; Morgan *et al.*, 2014). "(J)udges and juries, who impliedly are incapable of understanding the technical aspects of such a case, are at the mercy of the experts" (Owen, 2002; p. 346). Although no former standards exist for the admissibility of forensic evidence in the UK, there have been calls from the government, practitioners, and the Law Commission to develop a 'gate-keeping' test for the validity of scientific expert testimony. This is in the contrast to the US, where standards for the admissibility of scientific evidence have existed since 1923. Due to the lack of a formal structure in the UK, the standards for the US will be highlighted.

In 1923, *Frye v. United States* discussed the admissibility of a polygraph test as evidence. From this case, the Frye standard was developed which was used to determine the admissibility of scientific evidence. Using this standard, expert opinion is only admissible when the technique is generally accepted as reliable in the relevant scientific community. This standard was superseded in 1993, following the US Supreme Court case *Daubert v. Merrell Dow Pharmaceuticals*. Under the Daubert standard, five statutory criteria must be fulfilled, referred to as

reliability factors, to assess the validity of an expert's proffered testimony: testability, peer review, error rate, control standards, and general acceptance (Owen, 2002). Since Daubert, the US Supreme Court has revisited the issue of expert testimony numerous times, including *General Electric Company v. Joiner* (1997), *Kumho Tire Company v. Carmichael* (1999), and *Weisgram v. Marley Company* (1999).

Amendments to the Federal Rules of Evidence regarding opinion evidence and expert testimony were approved in 2000 by the Supreme Court. Following these amendments, expert testimony is permitted only if it is grounded on sufficient facts, supported by data, and is the result of reliable principles and methods that are themselves reliably applied to the facts of the case (Owen, 2002). As such, it is important that forensic science has an empirical evidence base, bolstered by sound methodological research, which experts can draw on in order to interpret forensic evidence effectively (House of Lords Science and Technology Select Committee, 2019).

With respect to experimental studies within the forensic sciences, targeted research which addresses trace evidence dynamics has been highlighted as a priority (Forensic Science Regulator, 2019). As such, the experimental work conducted in this thesis set out to develop a greater understanding of the trace evidence dynamics of forensic materials under controlled conditions through the adoption and implementation of a reductionist approach (Chapters 4 and 5).

2.6 Trace Evidence

The necessity of data to underpin the understanding and effective interpretation of trace materials has been highlighted in a series of high-profile reports in the UK (Forensic Science Regulator, 2015, 2017, 2018, 2019; House of Lords Science and Technology Select Committee, 2019), Canada (Centre for Forensic Science and Medicine, 2013), and the US (President's Council of Advisors on Science and Technology, 2016). The significance of such intelligence, or evidence, is reliant upon having an evidence base that can provide the context for a particular form of trace and offer insight into how that trace may have been transferred, and under what conditions it may persist and over what timeframes (Forensic Science Regulator, 2015; Government Chief Scientific Adviser, 2015).

Conceptually, trace evidence is regarded as a minuscule quantity of a substance that is too small to be measured. A more practical definition of trace evidence, as proposed by Roux and Robertson (2016), is “the analysis of materials that, because of their size or texture, transfer from one location to another and persist there for some period of time” (p. 20). The range of materials covered by the terminology is broad and can include, but is not limited to, soil, pollen, fibres, hairs, glass fragments, paint chips, gunshot residue, fingermarks, drugs of abuse, and explosive residues (Roux & Robertson, 2016). The information obtained from these materials can help reconstruct events and their sequence by associating people, places, and items of interest as well as inferring circumstantial information when a crime scene is unknown (Petraco, 1986; Roux *et al*., 2015).

The foundation of trace evidence is contact between people, places, and objects. Edmond Locard, a pioneering French criminologist, was interested in microscopic evidence and believed that such trace evidence was crucial in linking people to places (Bell, 2004). Locard noted that “[*i*]*l est impossible au malfaiteur d'agir avec l'intensité que suppose l'action criminelle sans laisser des traces de son passage*” (it is impossible for the criminal to act, with the intensity that criminal action requires, without leaving traces of his presence) (Locard, 1920, p. 139). This later became a fundamental tenet of forensic science known as Locard’s Exchange Principle, summarised as “any action of an individual … cannot occur without leaving a trace” (Stauffer, 2006; p. 109). As such, during the commission of a crime event, there will be a two-way transfer of trace evidence between: (1) a perpetrator and their victim; (2) a perpetrator and the crime scene; and (3) the victim and the crime scene (**Figure 2.4**).

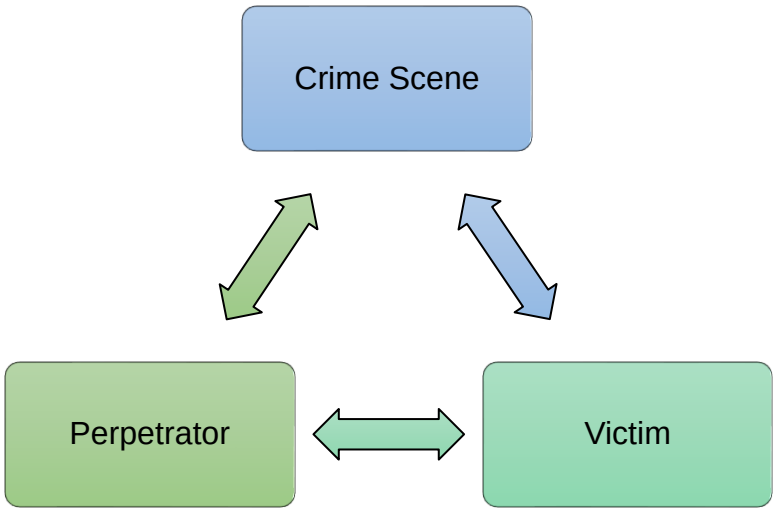


Figure 2.4. Locard's principle of exchange. Arrows represent two-way transfer of trace evidence between a perpetrator, victim, and crime scene.

As a form of evidence, particle traces should be a major problem-solving tool. Particle traces are always present, and they have the potential to address a wide range of questions facing the forensic investigator. Despite this, "the value of trace evidence has traditionally been underestimated, and its detection and collection are often neglected due to lack of knowledge, time, resources, and challenges in interpretation" (Roux *et al*., 2015; p. 2). Empirical studies which address the dynamics of how material traces may have been transferred, under what conditions they may persist, and over what timescales they can persist, can provide contextual information, and enable robust crime reconstructions (Morgan *et al.*, 2020; Trejos *et al.*, 2020).

2.7 Transfer and Persistence of Forensic Materials

"A consideration of evidence dynamics is axiomatic to the process of crime reconstruction" (French *et al.*, 2012; p. 33). The transfer of evidence is founded upon Locard's exchange principle which states that a transfer of material always occurs whenever two objects come into contact with one another. As such, whenever a crime has occurred, the perpetrator(s) of that crime will both bring something into the scene and take something from the scene when they leave. This transfer of trace evidence, both to and from a scene, can be used as forensic evidence in enabling crime reconstructions.

Particulate matter can be transferred in a number of ways. The first mechanism is through primary transfer, whereby materials are transferred onto an item

through direct contact (van Oorschot *et al.*, 2019). For example, a perpetrator may leave their DNA on a surface they touch in the commission of a criminal event (Daly *et al*., 2012). Primary transfer of materials can also occur without direct contact, such as through coughing, sneezing, and speaking (Meakin & Jamieson, 2013). The second mechanism is the indirect transfer of materials through an intermediary item (Taupin, 2016). This can occur via one intermediatory item (secondary transfer) or multiple intermediaries (such as tertiary and quaternary transfers) (van Oorschot *et al.*, 2019). For example, the secondary transfer of gunshot residue can occur when an individual either handles a previously discharged firearm or through shaking hands with someone who has discharged a firearm (French *et al*., 2014). Although particulate matter transfers resulting from a crime event (syn-event transfers) are important, it is also prudent to consider transfers that occurred prior to a crime event (pre-event transfers) as well as after a crime event (post-event transfers) and those transfers that might have occurred through the collection, storage, and analysis (**Figure 2.5**) (Morgan *et al.*, 2020).

Although the transfer of particulate matter readily occurs, the amount of material that is transferred, as well as the distribution of the transferred material, depends upon a number of factors. Firstly, the nature and properties of the donor and recipient surfaces will influence how easily a material is lost from the donor surface and to what extent that material is then able to adhere to the recipient surface (Deedrick, 2000). The propensity for transfer will also be influenced by the physical and surface properties of material being transferred (Brewster *et al.*, 1985; Chisum & Turvey, 2011b). Studies have also shown that the force (Pounds & Smalldon, 1975a; Goray *et al*., 2010a; Tobias *et al*., 2017), duration (Roux *et al*., 1999; Deedrick, 2001; Gherghel *et al*., 2016), and conditions of contact can also have an impact on the transfer of trace materials. Lastly, environmental conditions, such as wind activity and moisture availability, can also affect the transfer of forensic materials (Ruffell & McKinley, 2008).

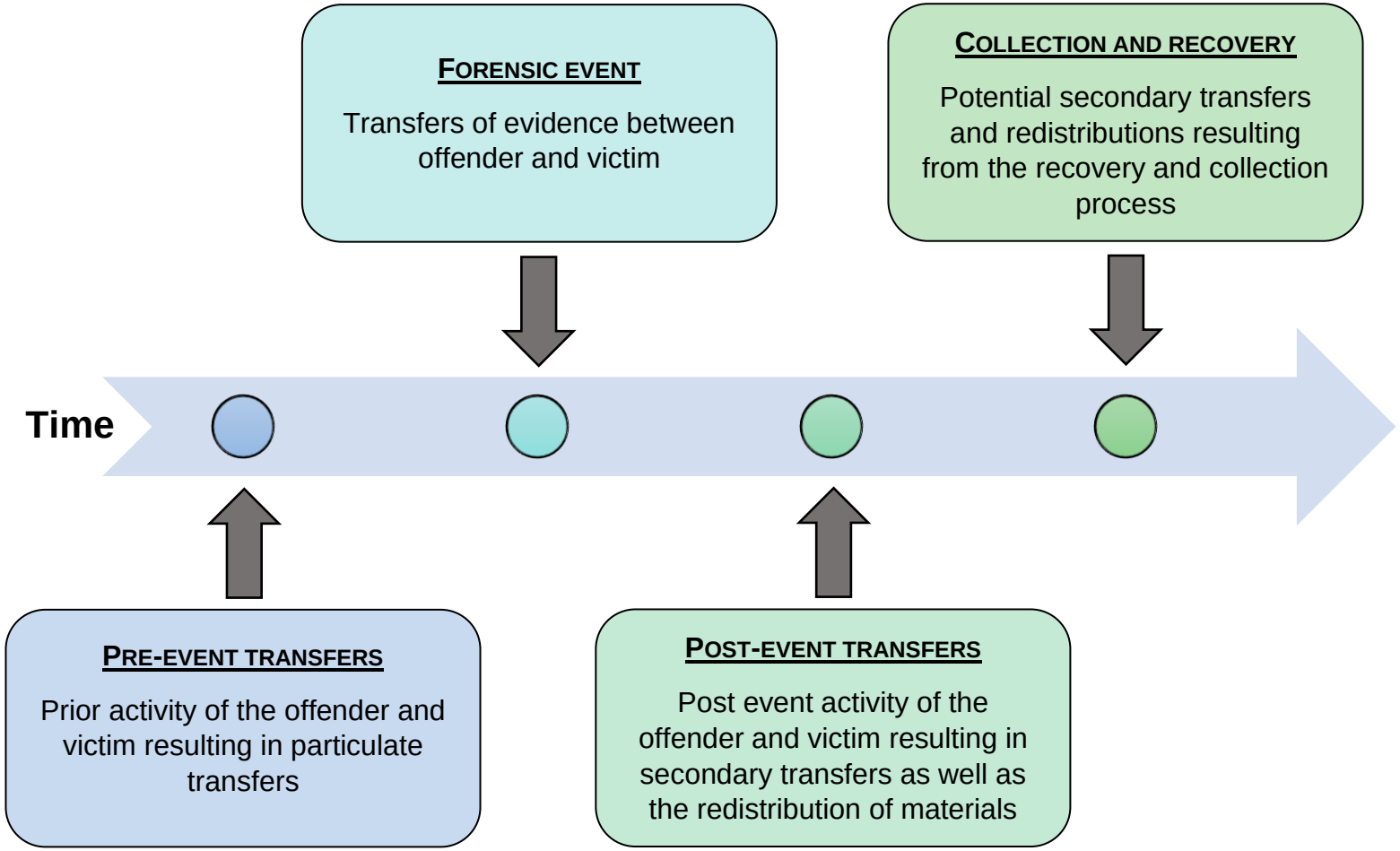


Figure 2.5. Timeline of particulate matter transfers (pre-, syn-, and post- crime event).

Once physical evidence has been generated and transfer onto a surface or object has occurred, the retention of the physical evidence becomes an important property; particulate matter will not remain on an item indefinitely but will be lost over time. Studies have shown that there are four main factors that can affect the persistence of trace materials including the retentive properties of the host surface, the tenacity of the trace material, the nature of the post-forensic event activity and the extent of the redistribution/reincorporation mechanisms (Akulova *et al.*, 2002; Wiggins *et al.*, 2002; Dachs *et al.*, 2003; Bull *et al.*, 2006). The longer a trace persists on a given item, the greater the chance that once that material is recovered it will comprise of particulates from pre-, syn-, and post-forensic event transfers (Morgan & Bull, 2007).

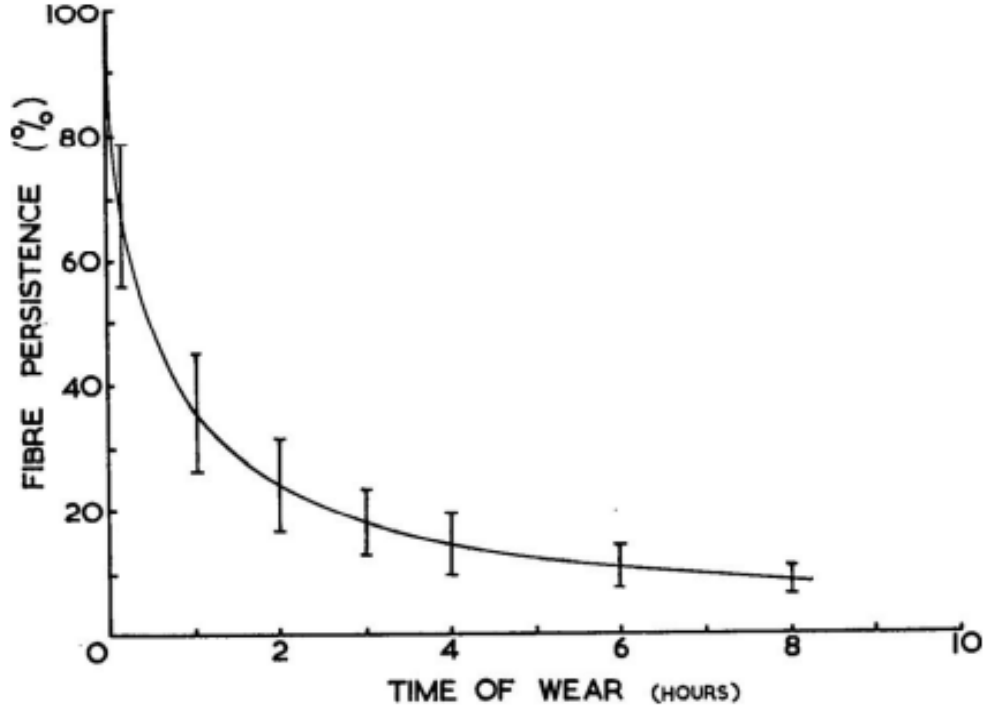


Figure 2.6. Two-stage mechanism of decay. From “The transfer of fibres between clothing materials during simulated contacts and their persistence during wear: Part II – Fibre persistence” by C.A. Pounds & K.W. Smalldon, 1975, *Journal of the Forensic Science Society, 15*(1), p. 34. Copyright 1975 by Forensic Science Society.

As trace particulates are lost over time, “the absence of evidence is not evidence of absence” (Grieve & Houck, 2003; p. 3). Thus, not finding trace evidence does not necessarily exclude a transfer from having taken place. Pounds and Smalldon (1975b), were the first to report upon a two-stage mechanism of decay that was

exhibited for fibre evidence (**Figure 2.6**). This was subsequently observed for other evidence types such as glass (Hicks *et al.*, 1996), polyurethane foam (Wiggins *et al.*, 2002), hair (Dachs *et al.*, 2003), pollen (Bull *et al.*, 2006), fragrances (Gherghel *et al*., 2020), and diatoms (Scott *et al.*, 2021). In this mechanism of decay, there is a rapid initial loss of the transferred material, which is followed by a more gradual loss, tending towards zero, but never reaching zero. Those particulates that are weakly bound (loosely adhered) are more readily lost from a given item once transferred whereas those which are more strongly bound exhibit a slower rate of loss (Roux *et al.*, 2013). Although intermolecular forces, such as electrostatic interactions, are considered to impact upon the adhesion of particulates during initial transfer (Roux *et al*., 1999), persistence is likely to be largely influenced by the combined effects of gravitational forces and movement (Pounds & Smalldon, 1975c).

A summary of the literature surrounding the transfer and persistence of different evidence types is presented in **Table 2.1**. Although a wide array of forensic materials has been examined, there is a notable dearth of literature surrounding the trace evidence dynamics of explosive and drug traces. As such, these particulates formed the focus of the experimental studies conducted in this thesis. Additionally, a large proportion of the previous literature have assessed clothing as the recipient surface. This thesis therefore sought to expand upon the knowledge base through assessing the impact of porous and non-porous building materials typically encountered in the environment as recipient surfaces. The term “porosity” being used to characterise surfaces aligns with the broad body of published work in forensic science.

Table 2.1. Examples of previously published literature surrounding the transfer and persistence of different forensic materials.

Evidence Type	Recipient Surface	Citation(s)
Diatoms	Clothing textiles	Scott *et al.*, 2014
	Footwear	Levin *et al*., 2017
DNA	Knife handles	Meakin *et al*., 2017
Fibres *(automobile)*	Footwear	Roux *et al*., 1999
Fibres *(clothing)*	Clothing textiles	Pounds & Smalldon, 1975a Pounds & Smalldon, 1975b Robertson *et al.*, 1982 Lowrie & Jackson, 1991 Akulova *et al*., 2002 Lepot *et al*., 2015
	Hair	Ashcroft *et al*., 1988 Salter & Cook, 1996
	Linens	Palmer & Banks, 2005
	Skin	Palmer & Burch, 2009 DeBattista *et al*., 2014
Foam (*polyurethane)*	Clothing textiles	Wiggins *et al*., 2002
Fragrances	Clothing textiles	Gherghel *et al*., 2019 Gherghel *et al*., 2020
Glass fragments	Clothing textiles	Brewster *et al*., 1985 Hicks *et al*., 1996
Gunshot residue	Hair	Zeichner & Levin, 1993
	Skin	Jalanti *et al*., 1999 Lindsay *et al*., 2011 French *et al*., 2014 Lindström *et al*., 2015
Hair *(human and animal)*	Clothing textiles	Dachs *et al*., 2003 Boehme *et al*., 2009
Lighter flint	Clothing textiles	Bull *et al*., 2006
Paint fragments	Clothing textiles	Pearson *et al.*¸1971
Pollen grains	Clothing textiles	Bull *et al.*, 2006 Schield *et al*., 2016
	Footwear	Morgan *et al*., 2009b
Soils	Clothing textiles	Murray *et al.*, 2016
	Footwear	Stoney *et al*., 2016

2.8 Recovery of Forensic Materials

Although understanding transfer and persistence is fundamental in enabling robust reconstructions, to have evidential value, these traces have to first be identified and successfully recovered. A number of methods are utilised by examiners to recover trace particulates. Such methods include tape lifting, stubbing, combing, swabbing (dry and wet), scraping, and vacuum sweeping (Petraco, 1987; Bisbing, 2001; Song-im *et al*., 2012a; Jones *et al*., 2019; Aberle *et al*., 2021). The method employed to recover these particulates is dependent upon several factors including the trace that is being collected, where that trace is situated (its location), and the presence of other evidence along with the nature and condition of this evidence (Scientific Working Group on Materials Analysis, 1998; National Institute of Standards and Technology, 2020). As with the transfer of evidence (Goray *et al.*, 2010a, 2010b), the nature of the surface to which the trace is adhering has been shown to impact the recovery. With respect to trace particulates, studies have shown that a lower recovery is obtained from porous surfaces (for example paper, cloth, and wood) than from non-porous surfaces (such as glass, metal, and plastic). This has been observed for a number of different forensically relevant materials: bacterial endospores (Valentine *et al*., 2008), anthrax and ricin (Frawley *et al*., 2008), fibres (Jones *et al*., 2019), DNA (Verdon *et al.*, 2014), drug residues (Sisco *et al*., 2018), and explosive traces (Yu *et al*., 2016). The lower recovery from these surfaces can be partly attributed to the particulates being ensconced within the pores of the surface or enmeshed within the tightly woven fibres of fabrics (Yu *et al*., 2016) (**Figure 2.7**).

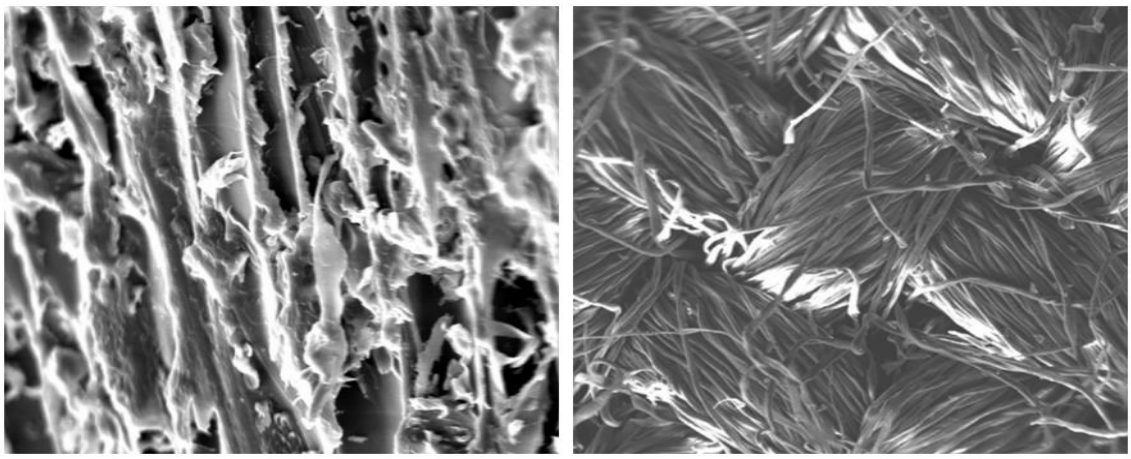


Figure 2.7. SEM micrographs of two porous surfaces: unfinished wood (left) and a cotton polyester blend fabric (right).

To overcome the problems associated with the lower recovery of trace particulates from porous surfaces, there may be advantages in exploring and developing more effective methods for inclusion in the tool kit for forensic collection. The application of hydrogels to evidence recovery may offer a solution through their capacity to permeate into the pores of such surfaces and encapsulate particulates for recovery as has been highlighted in other applications (VanHorne-Sealy, 2008). Hydrogels are comprised of a three-dimensional cross-linked polymeric network. Depending on the nature of the cross-linking reaction, either covalent bonds or ionic interactions are formed (Singh *et al.*, 2010). Those hydrogels containing covalent bonds are referred to as permanent hydrogels whereas those containing ionic interactions, hydrogen bonding, or hydrophobic interactions, are classified as physical hydrogels (Pal *et al.*, 2009). Regardless of the bonding interactions, hydrogels exist in a state that is neither completely solid nor liquid (Okay, 2009). A key property of hydrogels is that they can absorb large volumes of water, or biological fluid, within the spaces available among the polymeric chains as a result of the hydrophilic functional groups attached to the polymer backbone (Okay, 2009; Pal *et al*., 2009; Ahmed, 2015). The total water holding capacity of a given hydrogel is predicated on the number of hydrophilic groups and cross-linking density (Miyata, 2002). As the number of hydrophilic groups increases, the capacity of a hydrogel to absorb water also increases (Pal *et al.*, 2009). Conversely, increasing the cross-linking density results in a lower rate of water absorption (Kowalski *et al*., 2019). Any volume of water that is absorbed by the hydrogel then allows for the encapsulation and free diffusion of solute molecules which can be exploited for various applications (Okay, 2009). The physical properties and characteristics of a hydrogel, such as diffusivity of the entrapped molecules and mechanical strength, are determined by the mesh size of the polymeric network as well as the chemical and physical crosslinking present (Singh *et al*., 2010) (**Figure 2.8**).

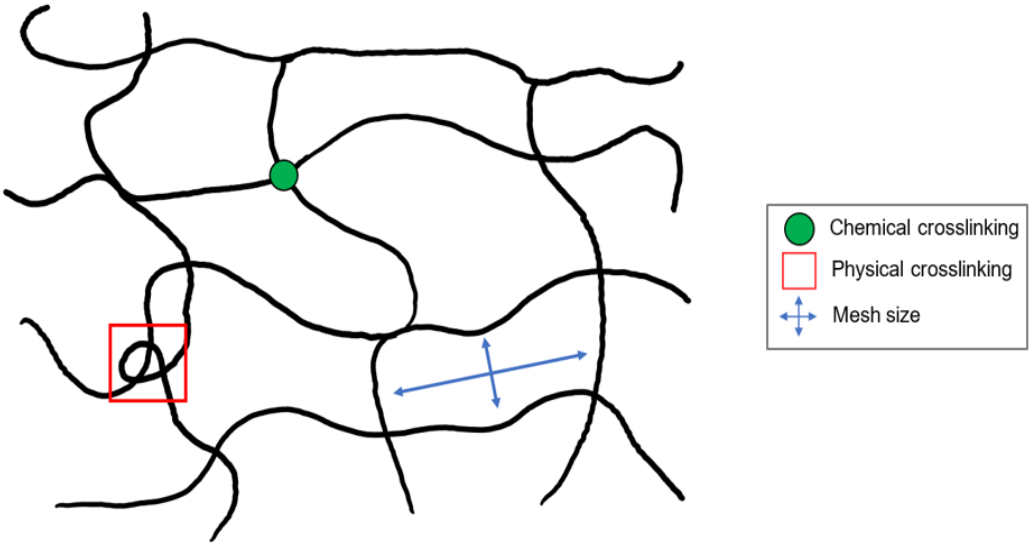


Figure 2.8. Exemplar two-dimensional hydrogel network highlighting the physical and chemical crosslinks along with the mesh size.

Hydrogels have primarily been used for biomedical applications as an analogue for human tissue (Aswathy *et al.*, 2020). As hydrogels can be designed and tailored to meet specific requirements, there has been an increase in the research and utilisation of hydrogels across a range of different fields, such as agriculture, the food industry, pharmaceuticals, and biosensors (Ahmed, 2015; Aswathy *et al*., 2020). Within forensic science, there has been preliminary work on the application of hydrogels as a collection medium for trace particulates, namely 2,4,6-trinitrotoluene (TNT) (Choodum *et al*., 2016) as well as amino acids and DNA from fingermarks (van Helmond *et al*., 2018). However, the potential contributions of hydrogels as a tool for the recovery of forensic materials remains largely untapped. As such, this thesis explored the use of a novel gelatine-based hydrogel formulation for the collection of explosive and drug residues from a variety of surfaces (Chapters 3 and 5).

2.9 Summary and Research Questions

The focus of this research lies within the context of the forensic process and addresses a current debate within the literature regarding the importance and necessity of a growing body of empirical research to inform each stage of that process. Trace evidence is often a valuable part of that process offering both evidence and intelligence that can inform an investigation. An understanding of the transfer and persistence of forensic materials is fundamental in enabling robust crime reconstructions and therefore a priority for research. In order to

obtain intelligence from these materials, they must first be successfully recovered. As such, this thesis was designed to address the transfer, persistence, and collection of forensic materials.

The experimental work presented within this thesis explores the use of a novel gelatine-based hydrogel for the collection of explosive and drug resides on a variety of porous and non-porous surfaces. Such work sought to expand upon the forensic tool kit investigators can rely upon to recover evidence from a given scene. Additionally, a complementary approach to understanding evidence dynamics was undertaken, focusing on the mechanics of transfer of both drugs and explosive traces through methodological reductionism, a method not often explored with respect to evidence dynamics. To achieve this, a new method for assessing the impact of certain variables on the transfer of forensic materials is presented which employs an Instron ElectroPuls E3000 to independently control for the force of contact, the duration of contact, and the amount of rotation applied during contact. Lastly, to assess persistence, a different approach was adopted, focusing on the persistence of forensic materials following activity, rather than assessing persistence over time which is typically reported in the literature.

The aim of these studies was therefore threefold: (1) to address the poor recovery of forensic materials from porous surfaces by exploring the use of a novel hydrogel formulation; (2) to expand upon an empirical evidence base through implementing a reductionist approach to understand the mechanics of how a particular form of trace may have been transferred, and (3) offer insights into how a certain activity can impact upon the persistence of a given trace. As such, this thesis was designed to assess:

1. the recovery efficiency of a novel hydrogel formulation in collecting explosive (Chapter 3) and drug (Chapter 5) particulates on a variety of porous and non-porous surfaces;
2. the independent impact of force, time, and rotation on the transfer of explosive (Chapter 4) and drug (Chapter 5) particulates using a new method and bespoke application of an Instron ElectroPuls E3000;
3. the persistence of drug particulates on paper and polymer banknotes following activity (Chapter 5).

The results from these studies can indicate the best approaches for the collection of forensic materials on different surfaces as well bolster understanding of trace evidence dynamics and the importance of the context that such data provide for effective inferences to be made.

Chapter 3

Collection of Trace Evidence: A Novel Gel Formulation

It is widely accepted that an explosion will not consume 100% of the explosive compound involved. Traces of the explosive will remain in the form of residues and decomposition products, which will be widely scattered at an explosion scene. The key aim for a forensic scientist is therefore to recover traces of the unconsumed explosive. (Yu et al., 2016; p. 721)

Outline

Forensic scientists must be able to recover traces of any original explosive materials not consumed in the detonation, in a careful controlled manner to aid a crime reconstruction. In current sampling techniques, the collection efficiency of post-blast residue is highly variable and often dependent on the swabbing materials and solvent systems used. To address these method limitations, this study presents a gelatine-based sampling medium and assesses its capabilities for the collection of ammonium nitrate. Common surfaces were spotted with a known concentration of ammonium nitrate, the unset gel applied, allowed to set, and then peeled from the surface. The gel was dissolved, and solid phase extraction employed to isolate the target explosive compound and remove the constituents of the gel. The eluate was concentrated and subsequently analysed and quantified. Overall, the gel formulation was able to collect ammonium nitrate from all of the test surfaces, with recoveries ranging from 0.1% to 61.7%. This study presents a gelatine-based formulation that has the potential to become a valuable asset in the forensic tool kit for the collection of explosive traces. A key attribute of the gel is that it offers an alternative recovery tool to conventional swabbing and solvent extraction methods.

3.1 Introduction

The use of explosive materials by individuals and groups to propagate terror poses a significant threat to public safety, political stability, and the global economy (United States Department of Homeland Security, 2014). When explosives have been detonated, the residual explosive material deposited at a crime scene is of significant value in forensic investigations and subsequent crime reconstructions. Recovery of these traces can assist investigators in determining the type of explosive compound(s) used, identifying potential sources of

improvised explosive device components, comparing physical evidence from the scene to a potential suspect, corroborating statements, and potentially linking criminal cases (Vermette, 2012). However, the majority of explosive residues consist of relatively simple anions, cations, and vapours which have little diagnostic value (Yinon & Zitrin, 1996; Bell, 2006; Abdul-Karim *et al.*, 2013). Therefore, the primary goal of the forensic scientist is to recover traces of the original explosive materials that were not consumed in the detonation (Abdul-Karim *et al*., 2014).

3.1.1 Explosives

Chemical explosives can be differentiated from one another based on their explosive behaviour, which is characterised by their molecular structure (Yallop, 1980). They can be divided into three classes based on their relative sensitivities: primary, secondary, and tertiary (**Table 3.1**). Primary explosives are the most sensitive and undergo a rapid transition from burning to detonation and are able to propagate a detonation wave to less sensitive explosives. Unlike primary explosives, secondary explosives are relatively insensitive to shock, friction, flame, or heat and are initiated by a strong explosive shock wave that is produced by the explosion of a primary explosive. Upon detonation, the compounds of secondary explosives will dissociate instantaneously (Akhavan, 2011). Tertiary explosives are insensitive to shock and therefore the most stable. They require a secondary explosive, functioning as a booster, to cause detonation (Hopler, 2012).

Table 3.1. The three classes of chemical explosives based on their relative sensitivities and some examples of the explosive compounds in each class.

Class	Examples
Primary	Lead azide, lead styphnate, mercury fulminate and some organic peroxides
Secondary	Research Department Explosive (RDX), 2,4,6-trinitrotoluene (TNT), and pentaerythritol tetranitrate (PETN)
Tertiary	Ammonium nitrate-fuel oil (ANFO)

Explosives can also be classified based on their provenance: military, commercial, or home-made (Hopler, 2012). Unlike military and commercial explosives, home-made explosives do not have a legitimate use and recently, there has been an increase in the use of organic peroxides in explosive attacks

(Bali, 2013). This development in recent explosive material selections demonstrates the dynamic shift in criminal choices as explosion detection capabilities of security forces becomes enhanced. This trend is exemplified in the changing nature of materials used in terrorist attacks that have occurred over the last quarter-century (Gordon, 2004; Ducibella & Cunningham, 2010; Dyson, 2011; Bali, 2013).

3.1.2 Explosive Residues

Scientific literature pertaining to the origin and distribution of explosive residues is limited. It has been posited that explosive residues are derived from the partial reaction of the thin outer layer of an explosive charge. This results from the reflection of the shock wave from the surface back into the reaction zone of the device (Kelleher, 2002). Whilst the exact mechanism by which these traces persist during detonation events is unknown, it is accepted that they do, and can subsequently be found at post-blast scenes. Explosives undergoing deflagration (rapid burning) as opposed to detonation are termed low explosives and leave large deposits of unconsumed particulates which are easier to identify by investigators (Strobel, 2012; Abdul-Karim *et al*., 2014). Residue recoveries from explosives undergoing detonation are much more challenging (Strobel, 2012); in such events, undetonated explosive residues are still present as a result of the incomplete combustion of the explosive (Abdul-Karim *et al*., 2014). These trace residues are likely to be present in low amounts, potentially at the nanogram level, and therefore extreme care needs to be taken in their recovery and analysis (Murray, 2012).

3.1.3 Explosive Recovery

Residual traces of explosives can persist for long periods of time depending on the material upon which they are adsorbed or deposited. The collection method used to recover post-blast residues depends on a number of factors including the physical form of the residue, the type of deposition surface, and whether a low explosive or high explosive was used (Strobel, 2012). Mechanical vacuuming or physical sweeping of the epicentre of the explosion is considered to be the most efficient manner of collecting the unconsumed particulates of low explosives, whereas swabbing and solvent washes are frequently employed in the recovery of residues from high explosives (Todd *et al.*, 2010; Strobel, 2012)

The collection efficiency of post-blast residue based on current sampling techniques is highly variable, between approximately 10 and 90 per cent, and is often dependent on the swabbing medium and solvent used (DeTata *et al.*, 2013). Certain solvents such as acetone can damage certain substrates resulting in interfering background material being collected which can reduce recovery efficiency (Song-im *et al.*, 2012a). Also affecting the recovery efficiency is the unavoidable delay that exists between the time of collection and subsequent extraction and analysis. During this time period there is a decline in the concentration of volatile explosives as well as an increase in the formation of breakdown products (DeTata *et al.*, 2013). Cotton swabs moistened with an ethanol and water mixture are commonly used for the collection of both inorganic and organic explosive residues (DeTata *et al.*, 2013; McEneff *et al.*, 2018). Due to the low recoveries obtained from porous surfaces using traditional swabbing methods, vacuum sampling is also employed (Yu *et al.*, 2016, 2017).

In recent years, the increased use of high-energy improvised explosive materials, such as organic peroxides and homemade mixtures of inorganic and organic explosive compounds, in terrorist attacks has been observed around the world (Song-im *et al.*, 2012a; McEneff *et al.*, 2018). As such, there is a demand for an optimised sampling procedure capable of recovering a wide array of post-blast residues that may be encountered at these scenes. In addition, a single-step extraction method for both organic and inorganic compounds is required as traditional sequential extraction methods, which make use of multiple solvents with differing polarities, results in the loss of certain compounds (Song-im *et al.*, 2012b), which may have significance for the interpretation of this form of intelligence or evidence in the investigation context.

3.1.4 Hydrogels

Hydrogels have primarily been used in the medical and pharmaceutical sectors; however, recent studies have shown hydrogels, are being designed and tailored for a variety of different applications (Ahmed, 2015). Previous research into the use of hydrogels in the recovery of particulates and forensic sampling has been conducted, including the incorporation of hydrogels in a testing kit for TNT (Choodum *et al.*, 2016) and the use of hydrogels for the collection of amino acids and DNA from fingermarks (van Helmond *et al.*, 2018). Hydrogels are comprised

of a cross-linked polymeric network. Due to the presence of hydrophilic functional groups attached to the polymer backbone, hydrogels have the ability to absorb large quantities of water within the spaces available among the polymeric chains (Okay, 2009; Pal *et al.*, 2009). Water absorbed by the hydrogel allows for the free diffusion of solute molecules (Okay, 2009). The amount of water absorbed by the hydrogel, or total water holding capacity, is dictated by the cross-linking density (Miyata, 2002). As cross-linking density increases, there is a decrease in equilibrium swelling as a result of decrease in the hydrophilic groups (Pal *et al.*, 2009). Physical properties, such as diffusivity of the entrapped molecules and mechanical strength, are dictated by the mesh size of the polymeric network (Singh *et al.*, 2010).

3.1.5 Aim

The purpose of this study was to create and then evaluate the use of a gelatine-based medium for the collection of trace particulate explosive residues. The versatility of a gel medium means it can be used on a wide array of surfaces and has the potential to provide a new sample collection technique that can be employed by forensic investigators at post-blast scenes to overcome the limitations of conventional collection techniques. Therefore, the study sought to:

1. create a gelatine-based collection medium to provide an easy application method, requiring little to no scientific knowledge or technical expertise;
2. assess the degree to which this gelatine-based collection medium could offer a more efficient sampling technique, resulting in better collection efficiencies than the current methods being employed.

3.2 Materials and Methods

The gelatine-based collection medium constituents (gelatine powder, arrowroot powder, and glycerine) and test surfaces used were all sourced from local vendors. The inorganic explosive, ammonium nitrate (99.0% purity), along with the solvents used, were acquired from Sigma-Aldrich (Gillingham, Dorset, UK); all solvents were high-performance liquid chromatography (HPLC) grade unless otherwise stated. Bromelain tablets (500 mg), used to prevent the gelatine from resetting, were purchased from Holland & Barrett (Nuneaton, Warwickshire, UK). Oasis® MCX solid-phase extraction (SPE) cartridges (30 µm copolymer reverse-

phase 60 mg / 20 mL) from Waters (Elstee, Herts, UK) were used for the extraction and isolation of ammonium nitrate from the gel matrix.

3.2.1 Pilot Study

A series of gelatine-based formulations were assessed (**Table 3.2**). Arrowroot powder (starch) was used to provide structural support, through the formation of cross-links (Koev *et al.*, 2020), and glycerine, a humectant, was added to bind water, facilitating hydrogen bonding within the polymeric network (Xia *et al.*, 2019).

Table 3.2. The twenty-six gelatine-based test formulations assessed with the fit-for-purpose formulation highlighted. Amounts in grams.

Test	Gelatine	Arrowroot Powder	Glycerine	Water	Ratio
A	6	1	1	30	6:1:1:30
B	6	1	1	20	6:1:1:20
C	6	1	1	10	6:1:1:10
D	6	2	1	30	6:2:1:30
E	6	2	1	20	6:2:1:20
F	6	2	1	10	6:2:1:10
G	6	3	1	30	6:3:1:30
H	6	3	1	20	6:3:1:20
I	6	3	1	10	6:3:1:10
J	6	1	2	30	6:1:2:30
K	6	1	2	20	6:1:2:20
L	6	1	2	10	6:1:2:10
M	6	1	3	30	6:1:3:30
N	6	1	3	20	6:1:3:20
O	6	1	3	10	6:1:3:10
P	6	2	2	30	3:1:1:15
Q	6	2	2	20	3:1:1:10
R	6	2	2	10	3:1:1:5
S	6	2	3	30	6:2:3:30
T	6	2	3	20	6:2:3:20
U	6	2	3	10	6:2:3:10
V	6	3	2	30	6:3:2:30
W	6	3	2	20	6:3:2:20
X	6	3	2	10	6:3:2:10
Y	6	3	3	30	2:1:1:10
Z	6	3	3	20	6:3:3:20
AA	6	3	3	10	6:3:3:10

In order to be fit-for-purpose, the formulation needed to meet the following criteria: (1) fast setting; (2) easily removable (peelable) from a surface once set; (3) leave minimal residue on the surface once removed; and (4) able to collect an explosive compound from a surface. Microscopy was used to assess the degree to which each formulation was able to recover ammonium nitrate from a test surface (glass microscope slide) as well as the amount of residue left behind by the gel (see **Figure 3.1**). Based on these criteria, test formulation 'R' was deemed to be fit-for-purpose (see **Table 3.2**).

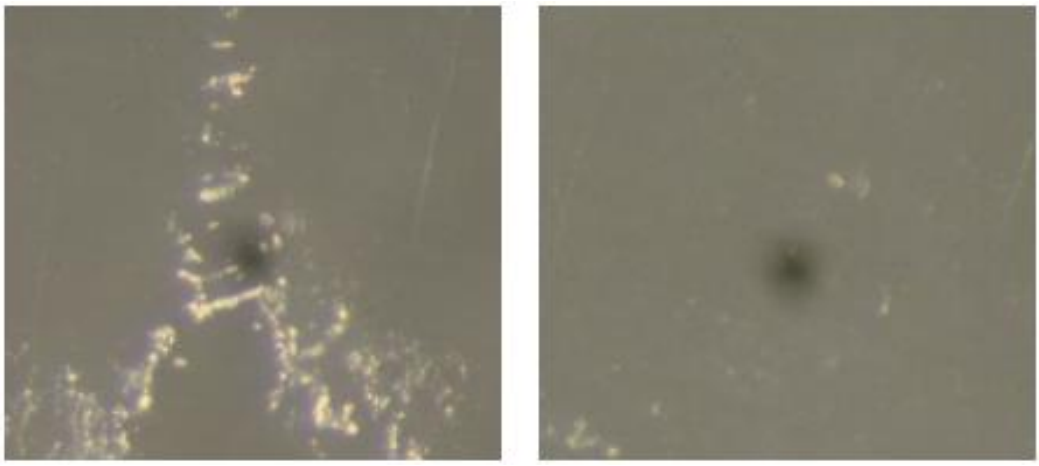
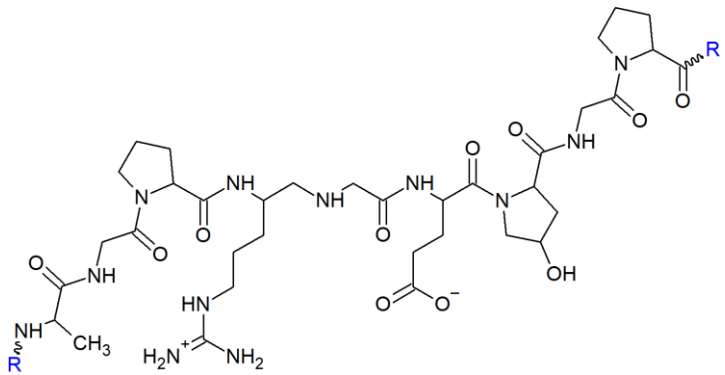
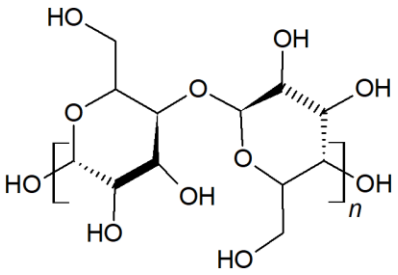
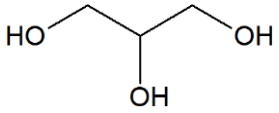
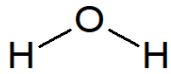


Figure 3.1. Before and after comparison. Ammonium nitrate on a glass microscope slide following deposition is shown on the left. The same surface is shown following application and removal of gel (test formulation 'R') on the right.

3.2.2 Final Formulation

The individual components of the gel formulation are readily obtainable from shops and do not have to be acquired through chemical suppliers, affording a cost-effective sampling medium. The gel was formulated by combining gelatine powder, arrowroot powder, glycerine, and water (**Table 3.3**) in a mass ratio of 3:1:1:5. Using a hotplate with stirrer, gelatine powder was added to water in a beaker and heated to 75° C until dissolved, stirring continually. Once dissolved, the arrowroot powder was added followed by the glycerine. The mixture was then kept at 75° C for 5 minutes, stirring continually.

Table 3.3. Molecular structures of the four components comprising the gelatine-based medium.

Component 1	Component 2	Component 3	Component 4
			
Gelatine ($C_{102}H_{151}O_{39}N_{31}$)	Starch $(C_6H_{10}O_5)_n$	Glycerine ($C_3H_8O_3$)	Water (H_2O)

3.2.3 Recovery Surfaces

A total of six different surfaces, cut into 2 cm x 2 cm squares, were used in this study: copper, acrylonitrile butadiene styrene (ABS; chosen to represent plastics), glass, carpet, a cotton polyester blend fabric, and unfinished wood (**Figure 3.2**) to represent common surfaces where explosive particulate recovery is often required. Each surface was cleaned with methanol (MeOH) prior to analysis to ensure they were free from contamination.

Figure 3.2. Non-porous (top) and porous (bottom) surfaces used in this study.

3.2.4 Ammonium Nitrate

Ammonium nitrate (**Figure 3.3**) has been used as an explosive in numerous terrorist incidents (Oppenheimer, 2008; Ducibella & Cunningham, 2010; Yücel *et al.*, 2018). It was selected as the target analyte for this study due to its relative safety and stability, as well as the ease of use and availability of the compound. A 1,000 mg/L working solution of ammonium nitrate was prepared by weighing 0.1 g of ammonium nitrate in a 100 cm^3 volumetric flask and diluting to volume with MeOH. Using a pipette, 100 µL of ammonium nitrate stock solution was spotted onto the centre of each of the surfaces (five replicates of each test surface) and allowed to evaporate in order to recrystallise the explosive from the solvent.

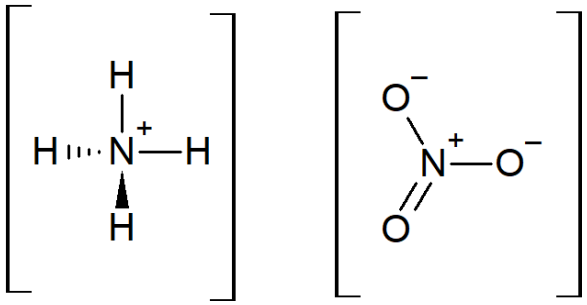


Figure 3.3. Molecular structure of ammonium nitrate (NH_4NO_3).

3.2.5 Application

Using a disposable syringe, 2 cm^3 of gel was applied to each surface (each surface was analysed in quintuplicate; n = 5 for each of the six surfaces) and left to solidify for 10 minutes. Once solidified, the gel was peeled from the surface with forceps and placed into a 10 cm^3 glass vial. The vial was then placed onto a hotplate, 5 cm^3 of water added, and heated at 75° C until dissolved. In order to prevent the gel from solidifying, bromelain (an enzyme found in the stems of pineapples) (**Figure 3.4**) was added to each sample (0.2 g of crushed (powdered) bromelain for every 6 g of gelatine used). During method development, bromelain did not impact the chromatographic profile obtained when added to solutions of ammonium solution.

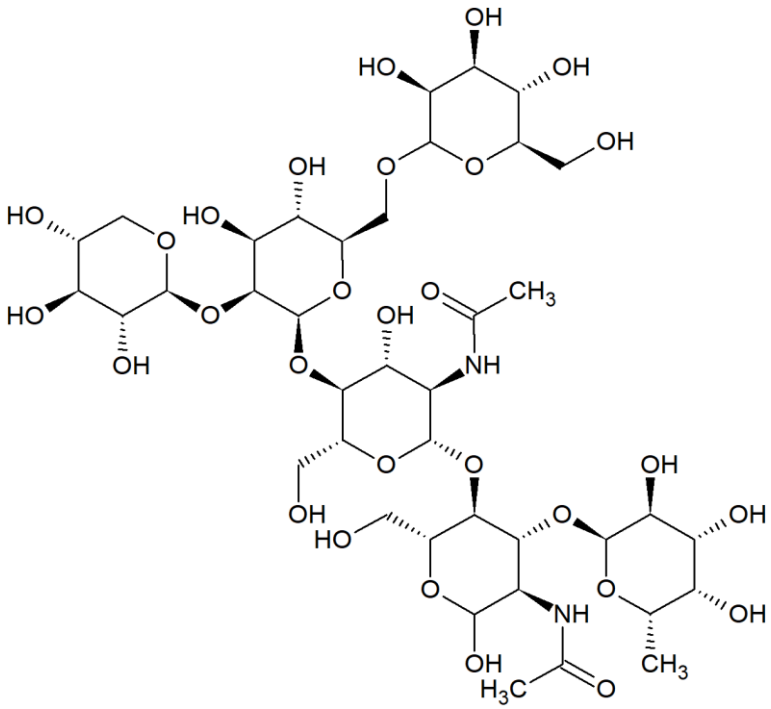


Figure 3.4. Molecular structure of bromelain ($C_{39}H_{66}N_2O_{29}$), a mixture of enzymes found in the stems of the pineapple plant (*Ananas comosus*).

3.2.6 Extraction

Following dissolution of the samples, SPE was employed as a sample clean-up technique to isolate the analytes of interest from the gelatine-based matrix. The SPE cartridges were conditioned with 5 cm^3 of MeOH, and then equilibrated with 10 cm^3 water. The samples (v = 5 cm^3) were then loaded onto the cartridges and allowed to elute under vacuum. The cartridges were then washed with 5 cm^3 of water and dried under vacuum. Finally, 5 cm^3 of acetonitrile (ACN) was added to the cartridges and the eluates collected. The eluates were dried under a stream of nitrogen gas and reconstituted in 1 cm^3 of MeOH prior to analysis. The absolute recovery of the SPE method was assessed by loading 1 cm^3 of a 1,000 mg/L ammonium nitration solution directly onto the cartridge and the eluate quantified. The absolute recovery with respect to ammonium nitrate was 92% ± 5% (mean ± standard deviation, n = 5).

3.2.7 Analysis

This was a targeted study which focused on the detection of the nitrate ion (NO_3^-) of ammonium nitrate. As such, the method employed is not amenable to mixtures and instances in which other unretained compounds may interfere with the signal obtained from the detector. A Prominence UFLC (Ultra Fast Liquid Chromatograph) system from Shimadzu fitted with an Acclaim™ Explosive E2 column was used to analyse the eluates. The method used (**Table 3.4**) was adapted from the US Environmental Protection Agency (EPA) Method 8330B for nitroaromatics, nitramines, and nitrate esters (United States Environmental Protection Agency, 2006).

Table 3.4. Liquid chromatography (LC) operating conditions for the analysis of ammonium nitrate.

Analytical Conditions	Method
Column	Acclaim Explosives E2 (3.0 x 150 mm, 3 µm, 120 Å)
Flow rate	0.3250 mL/min
Eluent	48/52 v/v MeOH/water
Gradient	Isocratic
Temperature	30 °C
Detection	UV 214 nm

Although the recommended wavelength for the detector was 230 nm, during the method development stage, it was found that setting the detector to 214 nm yielded a greater response for ammonium nitrate. As such, 214 nm was selected as the detector wavelength. Using this method, the retention time (RT) of ammonium nitrate was determined to be 1.8 minutes.

3.2.8 Quantification

In order to quantify any ammonium nitrate recovered, a nine-point calibration curve was established (1 mg/L to 1,000 mg/L). The average peak area was plotted against the concentration of each calibration standard (run in triplicate) injected onto the column and the linearity evaluated using the R^2 coefficient of determination. The R^2 value indicated that a good reliability existed in the linear relationships between the analyte concentrations and the peak areas (**Figure 3.5**). Once the calibration curve had been constructed and the linear range determined, two calibration checks were performed to assess the reliability of the calibration curve in quantifying ammonium nitrate. This was carried out using two independently prepared solutions at concentrations of 250 and 850 mg/L. The accuracy, with respect to determining the true concentration of ammonium nitrate in a sample, was determined to be 99.5% and 99.2% respectively.

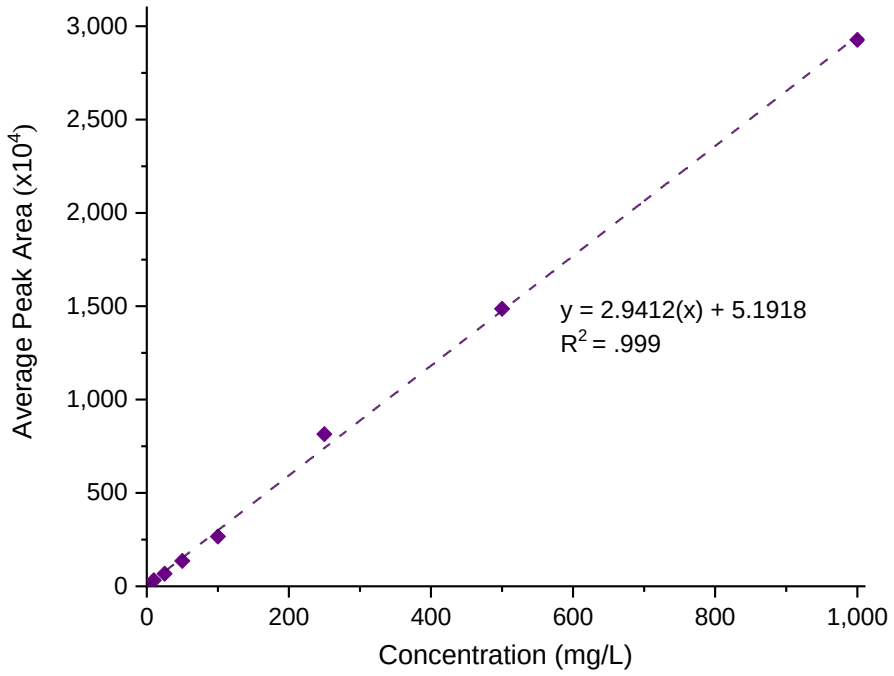


Figure 3.5. LC calibration curve of ammonium nitrate (reference standard).

3.2.9 Negative Controls

A series of negative controls was carried out to ensure that the response obtained from analysis was a direct result of ammonium nitrate being detected and not a background contaminate present on the surfaces used, or a constituent of the gel. To assess this, the procedure outlined above was repeated for each of the six different surface types which were not spotted with ammonium nitrate. Additionally, the gel formulation was analysed separately, having not been applied to a surface.

3.2.10 Statistics

Data was analysed using IBM® SPSS® Statistics (version 26). A two-sample *t*-test assuming unequal variances was employed to assess whether the mean recoveries obtained from porous and non-porous surfaces differed significantly ($p < .05$). A one-way analysis of variance (ANOVA) was conducted to compare the recovery of ammonium nitrate across the three different porous surfaces in order to assess if the means significantly ($p < .05$) differed. This was repeated for non-porous surfaces.

3.3 Results and Discussion

3.3.1 Negative Controls

No response at 1.8 minutes was measured in any of the negative controls. As such, the peak obtained in the recovery samples was a direct result of ammonium nitrate being detected in eluates and not resulting from a background contaminate present on the test surfaces or in the gel formulation.

3.3.2 Porous Surfaces

Recoveries from the porous surfaces ranged from 0.1% to 14.9%, with the highest recoveries obtained from carpet (average recovery of 6.8%) and the lowest recoveries from untreated wood (average recovery of 1.1%) (**Table 3.5**). The recoveries obtained from the three porous surfaces are graphically represented in **Figure 3.6**. When comparing the recoveries of ammonium nitrate obtained from wood, fabric and carpet, the results did not statistically differ, $F(2,12) = 3.68$, $p = .057$.

Table 3.5. Gel recovery results of ammonium nitrate from three porous surfaces. Units of concentration are mg/L.

Surface	Conc.	Avg. Conc. ± SD (%CV)	Recovery (%)	Avg. Recovery (%)
Wood	13.5 6.0 22.7 1.8 10.0	10.7 ± 7.3 (68.2%)	1.4 0.6 2.3 0.1 1.0	1.1
Fabric	11.2 5.0 30.4 20.4 13.7	16.1 ± 8.7 (53.7%)	1.1 0.5 3.0 2.0 1.4	1.6
Carpet	3.8 22.9 46.0 149.3 117.0	67.8 ± 55.9 (82.5%)	0.4 2.3 4.6 14.9 11.7	6.8

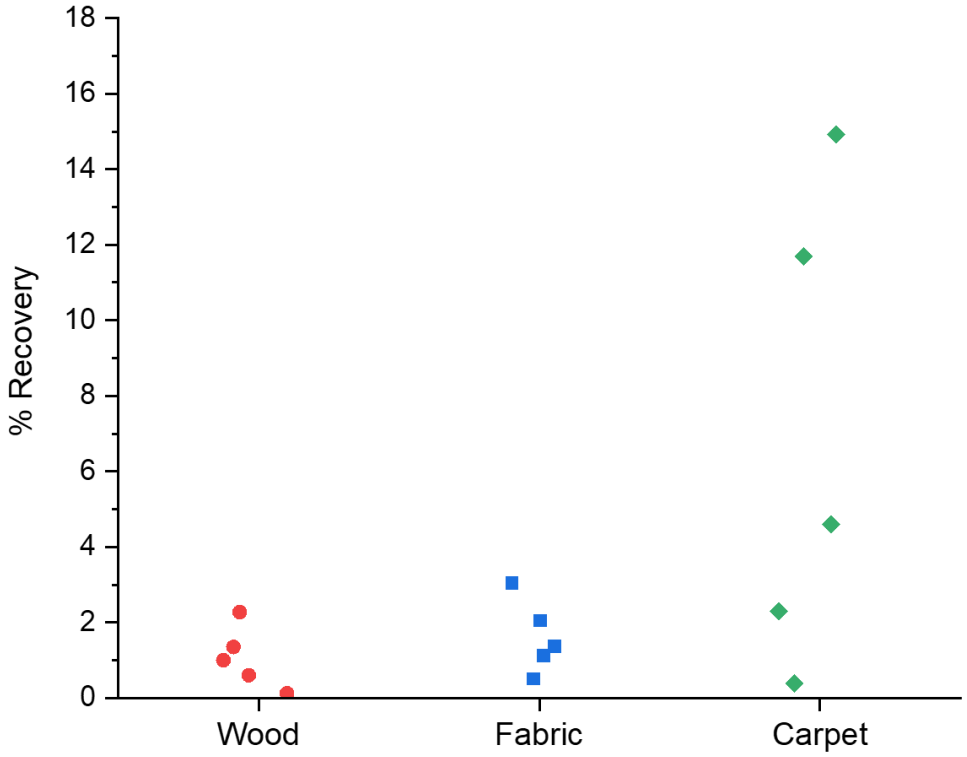


Figure 3.6. Individual value plot of the recoveries (%) obtained from the three porous surfaces tested.

3.3.3 Non-Porous Surfaces

Recoveries from non-porous surfaces ranged from 10.0% to 61.6%, with the highest recoveries obtained from ABS (average recovery of 38.6%) and the lowest recoveries from copper (average recovery of 18.4%) (**Table 3.6**). The recoveries obtained from the three non-porous surfaces are graphically represented in **Figure 3.7**. When comparing the recoveries of ammonium nitrate obtained from copper, glass, and ABS, the results did not statistically differ, $F(2, 12) = 2.75$, $p = .104$.

Table 3.6. Gel recovery results of ammonium nitrate from three non-porous surfaces. Units of concentration are mg/L.

Surface	Conc.	Avg. Conc. ± SD (%CV)	Recovery (%)	Avg. Recovery (%)
Copper	166.9	184.1 ± 56.4 (30.6%)	16.7	18.4
	100.3		10.0	
	217.2		21.7	
	167.3		16.7	
	268.8		26.9	
Glass	101.3	240.9 ± 93.2 (38.7%)	10.1	24.1
	359.7		36.0	
	180.9		18.1	
	319.9		32.0	
	242.5		24.3	
ABS	459.0	385.8 ± 188.0 (48.8%)	45.9	38.6
	163.5		16.4	
	526.5		52.7	
	616.0		61.6	
	163.9		16.4	

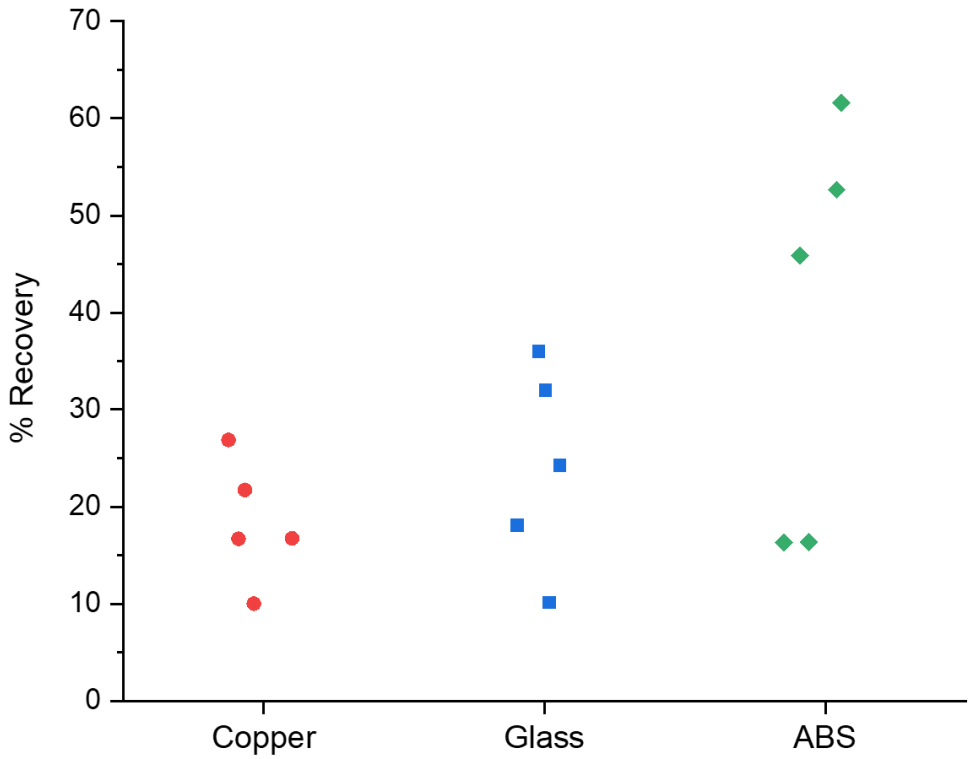


Figure 3.7. Individual value plot of the recoveries (%) obtained from the three non-porous surfaces tested.

3.3.4 All Surfaces

The average recoveries obtained from each surface are visually displayed in **Figure 3.8**. When comparing the recoveries obtained from porous and non-porous surfaces (**Figure 3.9**), a significantly, $t(28) = 5.68$, $p < .001$, higher recovery was obtained from the non-porous test surfaces (average recovery of 27.0%) than the porous test surfaces (average recovery 3.2%). The recoveries of ammonium nitrate from porous surfaces yielded higher coefficients of variation (%CV: wood = 68.2%, fabric = 53.7%, and carpet = 82.5%) than those from non-porous surfaces (%CV: copper = 30.6%, glass = 38.7%, and ABS = 48.8%). As such, a more consistent recovery was obtained when the gelatine-based collection medium was applied to non-porous surfaces.

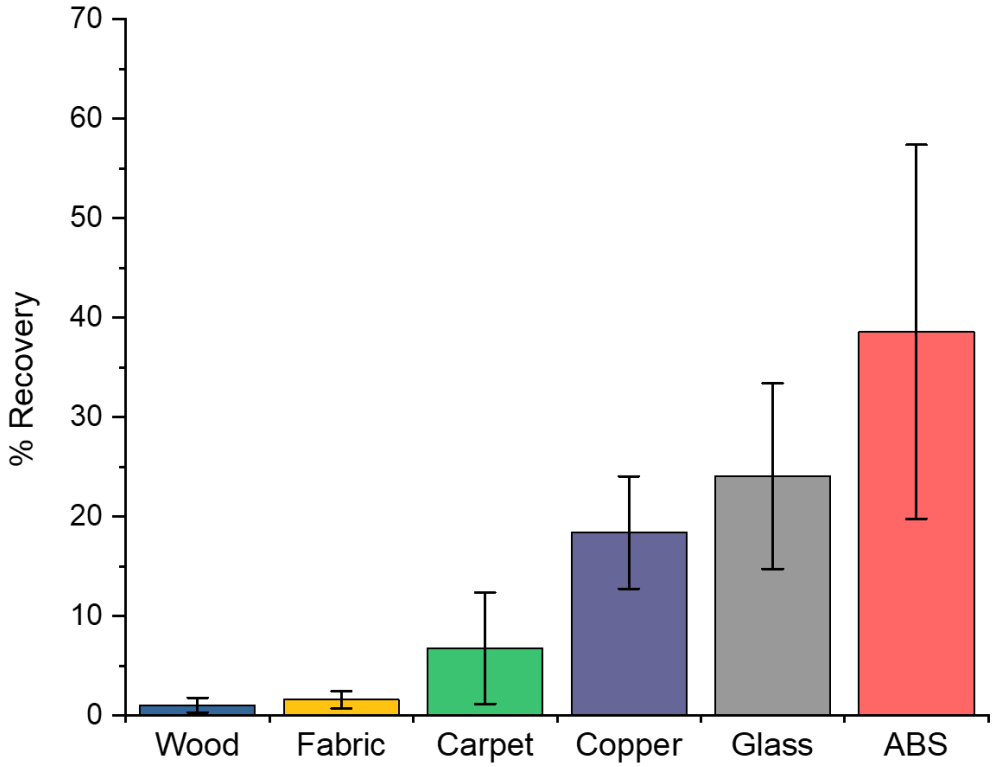


Figure 3.8. Mean recoveries (%) obtained from each surface. Error bars represent one standard deviation.

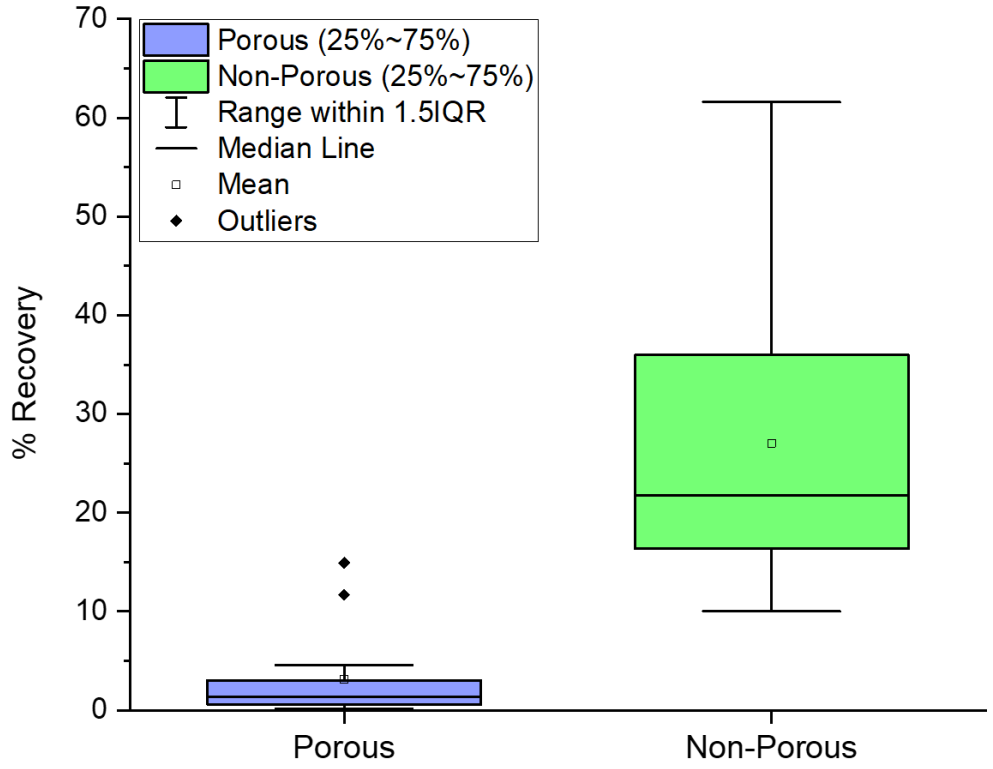


Figure 3.9. Box plot comparing the recoveries (%) obtained from porous (n=15) and non-porous surfaces (n=15).

The gel-based formulation was able to recover ammonium nitrate from all three of the porous surfaces tested, although the recoveries were low. Previous published studies such as Yu *et al.* (2016), have highlighted the difficulties of recovering trace explosive compounds from porous surfaces. With respect to the non-porous test surfaces, the recovery range obtained (10 – 62%) was much higher and falls within the range of conventional swabbing methods (10 – 90%) (DeTata *et al*., 2013).

3.3.5 Further Work

This study presents the formulation and application of a gelatine-based recovery method for explosive residues, specifically ammonium nitrate. In order to ascertain the circumstances in which the application of a gelatine-based hydrogel might be more appropriate than other recovery methods, direct comparison studies are now required, accounting for larger sampling areas and a wide array of explosive compounds (both organic and inorganic). Additionally, the ability of the gel to recover trace quantities of these residues should be assessed in order to determine the amount of explosive that can be feasibly recovered using hydrogels. The application of hydrogels as a collection medium could be extended to other traces, such as drug (Chapter 5) and gunshot residues. Hydrogels may afford other advantages that require assessment, including the long-term encapsulation of particulates during transfer and storage, the ability to be applied across complex surfaces, and the potential coverage across large surface areas.

3.4 Conclusion

This study set out to create a gelatine-based medium and then assess the suitability of the gel for the collection and recovery of explosive residues on a selection of common porous and non-porous surfaces. This study has demonstrated that:

1. it has been possible to create a low cost, effective gelatine-based formulation that is effective at recovering explosive residues from a range of different surfaces;
2. using the gelatine-based formulation, ammonium nitrate was recovered from all of the test surfaces;

3. a significantly higher recovery rate was obtained from the non-porous surfaces tested than the porous surfaces;
4. the recovery rates across the test surfaces ranged from 0.1% to 61.6%, with the highest recoveries obtained from ABS and the lowest from untreated wood.

The use of gel formulations could become a valuable asset in the forensic tool kit for the collection of explosive traces. A key attribute of the gel is that it offers an alternative recovery tool, with comparable recoveries from non-porous surfaces, to conventional swabbing and solvent extraction methods. As such, the results of this study provide a starting point from which further research can now be conducted. The method presented here offers an alternative collection approach that does not rely on the conventional swabbing materials and solvent systems. The results of this study are therefore promising and have the potential to offer a more effective method for inclusion in the tool kit which investigators tasked with the forensic collection and recovery of post-blast residues at bomb sites can rely upon.

Chapter 4

Transfer of Trace Evidence: A Reductionist Approach

There is a need to experimentally investigate plausible scenarios in which secondary transfers may occur. Such work will enhance our ability to accurately assess the likelihood that a secondary transfer has taken place in a forensic situation… [E]xperimentation that mimics forensic reality contributes to our understanding of trace evidence behaviour and therefore enhances our ability to interpret it within an investigative context. (French et al., 2014; p. 57)

Outline

Empirical studies evaluating the conditions under which the transfer of forensic materials occurs can provide contextual information and offer insight into how that material may have been transferred in a given scenario. Here, a reductionist approach was taken to assess the impact of force, time, and rotation on the transfer of an explosive compound. An Instron ElectroPuls E3000 material testing instrument was used to bring porous and non-porous surfaces adulterated with an ammonium nitrate into direct contact with a human skin analogue, controlling for the force of contact, duration of contact, and rotation applied during contact. Quantifiable amounts of ammonium nitrate were recovered from all of the recipient surfaces demonstrating that ammonium nitrate is readily transferred from one surface to another, even when contact occurs for a short duration with a relatively low force. More particulates were transferred from non-porous surfaces onto the human skin analogue, but the amount of ammonium nitrate transferred did not depend upon the force of contact, duration of contact, or the amount of rotation applied. However, when contact occurred and involved rotation, a greater transfer of ammonium nitrate was observed, compared to those contacts occurring without rotation being applied. This approach complements more commonly used holistic experiments that test multiple interacting variables in a realistic setting by isolating these variables, allowing them to be examined individually. This can be utilised to better understand the individual impact that specific variables have on the transfer of trace evidence in relevant crime reconstruction contexts.

4.1 Introduction

The need for data to underpin the evaluation and interpretation of trace materials has been highlighted in a series of high-profile reports in the US (President's Council of Advisors on Science and Technology, 2016), Canada (Centre for Forensic Science and Medicine, 2013), and UK (Forensic Science Regulator, 2015, 2017, 2018; House of Lords Science and Technology Select Committee, 2019). Establishing the significance of trace materials relies on having an evidence base that can support source, activity, and offence level propositions (Morgan *et al*., 2020). When identifying competing propositions, a number of factors are important, such as the circumstances of the case, availability of empirical support, observations that have been made, and the expertise of the scientist (Cook *et al*., 1998b). This study sought to generate data that can be used to provide support for activity level propositions in an attempt to answer "how" a trace was deposited by incorporating transfer properties.

Particle traces can address a wide range of questions facing the forensic investigator and contribute to crime reconstructions (Morgan, 2017a). Given the principle of contact and transfer (**Fig. 4.1**), empirical studies assessing the conditions under which transfers of specific traces occur can provide contextual information and offer insight into how that trace may have been transferred in a given scenario (Trejos *et al.*, 2020).

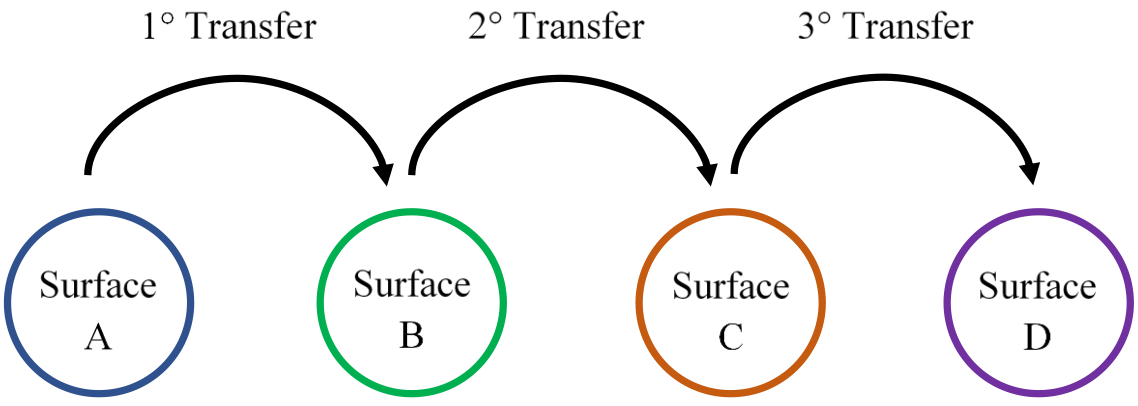


Figure 4.1. Overview of primary (1°), secondary (2°), and tertiary (3°) transfer. This is predicated on sufficient trace being available for transfer.

Use of explosive materials by criminals and terrorist groups poses a significant threat to society through impacting public safety, political stability, and the global economy (United States Department of Homeland Security, 2014). The direct handling of explosive materials by individuals has been shown to result in a significant transfer of that material onto their hands (Perret *et al*., 2008). Once acquired onto the hands, the subsequent transfer of these explosive residues onto various surfaces is therefore likely to occur when that individual comes into contact with them. As such, it is important to understand the dynamics of how explosive materials may have been transferred onto various surfaces through contact and handling. Transfers resulting from illicit activities is of relevance to crime reconstructions and can provide valuable intelligence that can inform an investigation (Morgan, 2017a).

With respect to other forensic materials, there is a relatively small literature base pertaining to the transfer and persistence of explosive traces. Previous studies have assessed the transfer and persistence of explosive materials onto the hands of individuals through direct handling (Perret *et al*., 2008), in consecutive fingerprints (Lees *et al*., 2018), and in hair (Oxley *et al*., 2005) under controlled conditions. There have also been studies that have sought to offer insights into the transfer and persistence of traces through experimental designs that seek to mimic forensically relevant scenarios (Brayley-Morris *et al*., 2015; Oldfield *et al*., 2018; Morgan *et al*., 2019; Scott *et al*., 2019). However, this approach is unable to separate individual variables within a specific scenario to establish the mechanics of what is being observed. A reductionist approach that seeks to assess each variable one at a time in a controlled and repeatable manner offers a complementary approach that can identify how each individual variable impacts on others in a complex system. These data can then be incorporated with data derived from ‘real world’ studies to develop models that can be used to underpin inference and evaluation of specific traces.

The ElectroPuls is a state-of-the-art dynamic instrument for material testing produced by Instron. It applies a force to a sample and measures the resulting change in length, allowing material properties, such as stress and strain, to be evaluated (Illinois Tool Works, 2015). These devices, of which those produced by Instron are the most common, are frequently used in the automotive, biomedical,

and construction industries to assess the mechanical properties of newly developed materials (Illinois Tool Works, n.d.). The ElectroPuls E3000 provides a range of dynamic and static loads up to 3,000 N, as well as providing torsional forces (Illinois Tool Works, 2015). All are highly controllable with great precision and reproducibility (Mårtensson *et al*., 2011; Lindberg *et al*., 2013; Völlner *et al*., 2019). In this reductionist experimental study, an Instron ElectroPuls E3000 was used to bring either a porous or non-porous surface adulterated with an explosive residue into direct contact with a human skin analogue (recipient surface), controlling for the force of contact, duration of contact, and rotation applied during contact. The amount of explosive residue transferred onto the recipient surface was then measured to isolate the individual impact that force, time, and rotation have on the percentage of the residue transferred. This can aid in developing an understanding of the proportion of explosive particulates transferred, through direct handling of explosive compounds and subsequent contact surfaces as well as innocent transfer arising from contact with contaminated surfaces, under different testing parameters.

4.2 Materials and Methods

4.2.1 Transfer and Recipient Surfaces

Unfinished medium-density fibreboard (MDF) (a porous transfer surface) and stainless steel (non-porous transfer surface) were sourced from local vendors. Each surface type was cut into 1 x 1 cm^2 squares (MDF: n = 36; stainless steel: n = 36), placed into individual plastic holders, and affixed with Blu Tack®. This was to prevent cross contamination as well as the trace being exposed to the environment, thereby minimising any loss of the trace. Chamois (*R. rupicapra*) leather (Amazon, UK) was selected as a human skin analogue for the recipient surface as it has been shown to simulate both the mechanical and frictional contact behaviour of human skin (Dąbrowska *et al*., 2016; Fenton *et al*., 2020). In order to reproduce the oil content of human skin, the leather was preconditioned by wearing under clothing for a period of eight hours to transfer dermal oils onto the recipient surface. The leather was then cut into 1 cm^2 circles (n = 72), each stored in a 5 cm^3 glass vial with a plastic lid.

4.2.2 Application of Ammonium Nitrate to Transfer Surface

Ammonium nitrate was selected as the explosive compound for this study due to its relative safety and stability. A working solution of ammonium nitrate with a concentration of 1,000 mg/L was prepared by weighing out 500 mg of ammonium nitrate (99.0% purity; Sigma Aldrich, Gillingham, Dorset, UK) and diluting to volume with MeOH (HPLC grade; Sigma Aldrich, Gillingham, Dorset, UK) in a 0.5 L volumetric flask. The approximate centre point of each of the transfer surfaces (MDF and stainless steel) was spotted with 100 µL of the working solution and left uncovered for thirty minutes to allow the solvent (MeOH) to evaporate and the solute (ammonium nitrate) to recrystallise onto the surface.

4.2.3 Instron® ElectroPuls™ E3000

An Instron ElectroPuls E3000 (Instron, High Wycombe, UK) was used to independently control the contact force (in newtons), duration of contact (in seconds), and rotation (in degrees) applied during contact, in order to assess their impact on the amount of ammonium nitrate transferred onto a recipient surface, a human skin analogue, following the controlled contact with a porous and non-porous surface adulterated with ammonium nitrate. The variables tested were contact force, duration of contact, and rotation applied during contact (**Table 4.1)**.

Table 4.1. The control factors and variables tested in each experimental run.

Experiment	Control Factors			Variables	
	Force (N)	**Rotation (°)**	**Recipient Surface**	**Transfer Surface**	**Time (s)**
1	150	0	Leather *(Human skin analogue)*	MDF *(Porous)*	2
2					60
3					150
4					300
5				Stainless Steel *(Non-porous)*	2
6					60
7					150
8					300
Experiment	**Control Factors**			**Variables**	
	Time (s)	**Rotation (°)**	**Recipient Surface**	**Transfer Surface**	**Force (N)**
9	30	0	Leather *(Human skin analogue)*	MDF *(Porous)*	10
10					60
11					120
12					240
13				Stainless Steel *(Non-porous)*	10
14					60
15					120
16					240
Experiment	**Control Factors**			**Variables**	
	Time (s)	**Force (N)**	**Recipient Surface**	**Transfer Surface**	**Rotation (°)**
17	30	150	Leather *(Human skin analogue)*	MDF *(Porous)*	90
18					180
19					270
20					360
21				Stainless Steel *(Non-porous)*	90
22					180
23					270
24					360

Operation of the ElectroPuls E3000 was controlled through Instron's WaveMatrix™ software (*version 1.8.383.0*). For dynamic and static loading, the ElectroPuls E3000 was fitted with a ±250 N Dynacell™ (dynamic load cell) with a load weighing accuracy of ±0.5% of measured load or ±0.005% of the load cell capacity (whichever is greater). Using the auto-tune function in the software, the stiffness value for both sample surfaces (MDF and stainless steel) was established to be 1.446 N/m. Once the instrument had been tuned, a three-step method was created to deliver precise

and reproducible contact between the surfaces (**Fig. 4.2**). The first step involved establishing a ramp waveform which controlled the speed at which the two surfaces were brought into contact with one another (0.05 mm/s). The second step related to the hold waveform which dictated the time and force which the two surfaces were held under compression. The final step was a ramp waveform which dictated the speed at which the two surfaces were pulled apart from one another (0.05 mm/s).

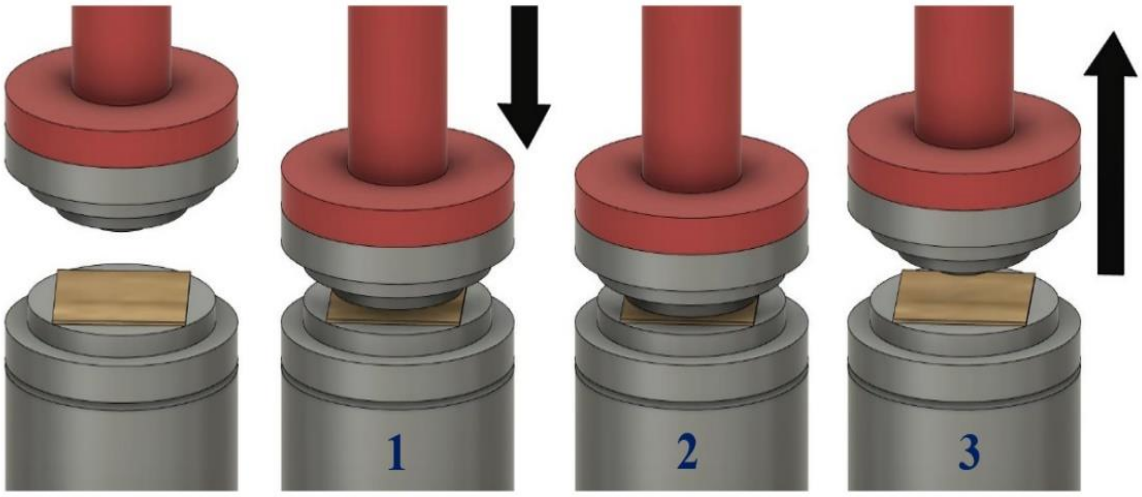


Figure 4.2. Three-step method used when assessing the amount of ammonium nitrate transferred onto the recipient surface as a function of force and time. (1) The surfaces were brought into contact with one another at a speed of 0.05 mm/s until the defined contact force had been obtained. (2) The two surfaces were held under the compression for the defined duration of time. (3) Contact between the two surfaces was removed at a speed of 0.05 mm/s.

For the torsional testing, the ElectroPuls E3000 was fitted with a ±5 kN Dynacell with ±25 N·m dynamic torque capacity. For these tests, the three-step method remained in place but with an additional rotation applied on the constant force hold (step 2) (**Fig. 4.3**). This rotation was applied in degrees per second. The angular velocities for each rotation (90°, 180°, 270°, and 360°) were as follows: 3°/s; 6°/s; 9°/s; 12°/s, with the hold waveform modified to apply these angular velocities over the duration of the hold time (30 s).

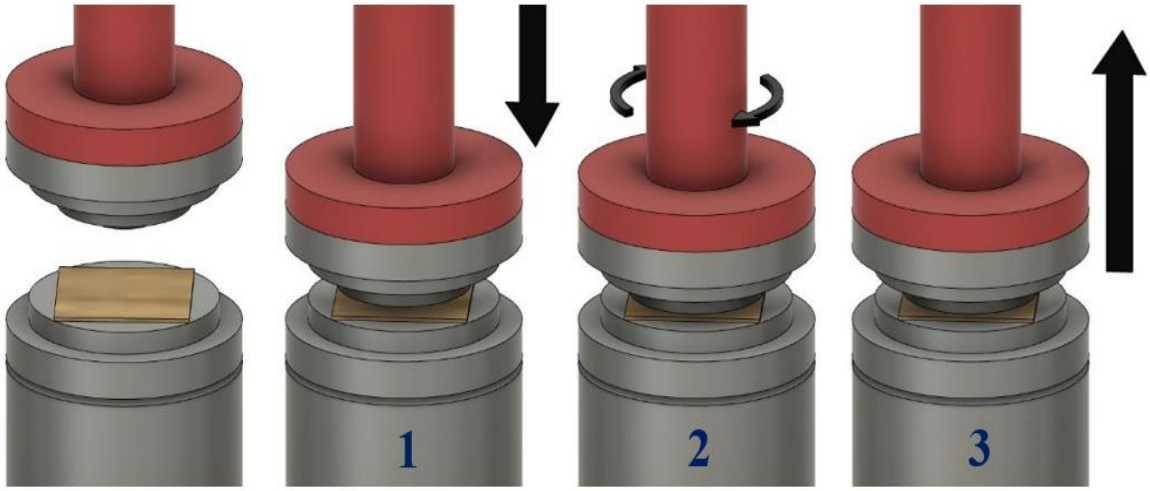


Figure 4.3. Method used when assessing the amount of ammonium nitrate transferred onto the recipient surface as a function of the amount of rotation applied during contact. (1) The surfaces were brought into contact with one another at a speed of 0.05 mm/s until compression at 150 N was achieved. (2) The two surfaces were held under the compression at 150 N for 30s during which a rotation was applied with an angular velocity of either 3°/s, 6°/s, 9°/s, or 12°/s depending on the extent of rotation assigned. (3) Contact between the two surfaces was removed at a speed of 0.05 mm/s.

The transfer and recipient surface were affixed to compression discs (platens) using double-sided tape. The compression platen with the transfer surface was physically attached to the dynamic load cell and the compression platen with the recipient surface was magnetically attached to the static load cell (**Fig. 4.4**). Each contact force, duration of contact, and amount of rotation during contact were assessed in triplicate for both surface types (porous and non-porous). For each run, the load registered by the dynamic load cell was recorded.

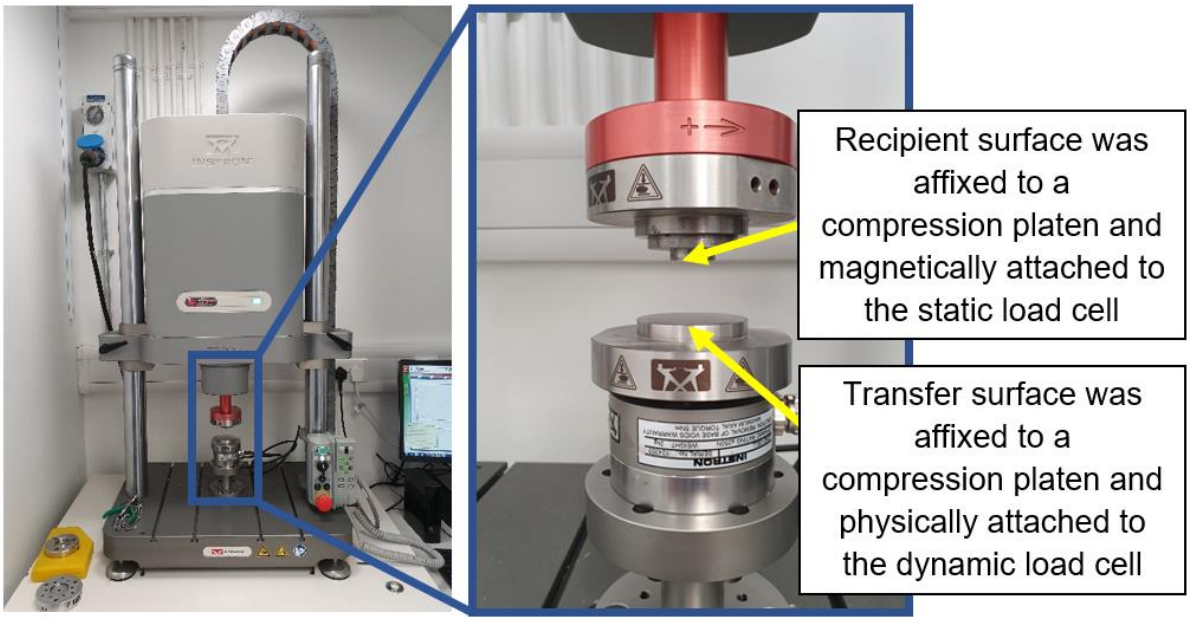


Figure 4.4. Instron's ElectroPuls E3000 system (left) and annotated close-up picture of the load cells highlighting the placement of the transfer and recipient surfaces (right).

4.2.4 Extraction of Ammonium Nitrate from Recipient Surface

To each of the recipient surfaces, 1 cm^3 of MeOH was added. Each sample was then placed on a Cole-Parmer™ Stuart™ See-Saw Rocker at 70 revolutions per minute (rpm) for 15 minutes. Following this time, the leather samples were wrung dry using a pair of forceps (cleaned with MeOH between samples) and the solution transferred into a 2 cm^3 screw cap autosampler vial. Each sample was analysed and quantified using LC as described in **Section 4.2.5**. The extraction efficiency of the method employed was assessed by spotting five leather samples with 100 µL of the stock solution (1,000 mg/L) of ammonium nitrate and quantifying the amount of ammonium nitrate recovered following extraction.

4.2.5 Quantification of Ammonium Nitrate in the Extracts

All analyses were performed on a Shimadzu Prominence UFLC System with ultraviolet (UV) detection. With respect to ammonium nitrate, the nitrate ion (NO_3^-) absorbs UV radiation and is therefore amenable to UV detection (Calcerrada *et al*., 2016). The analytical method used was adapted from the EPA Method 8330B and the detector wavelength optimised for the analysis of ammonium nitrate (see **Table 4.2**) (United States Environmental Protection Agency, 2006). The analysis of a

standard solution of ammonium nitrate yielded a single Gaussian peak on flat baseline with a corresponding RT of 1.8 minutes. As this was a targeted study to quantify the amount of ammonium nitrate transferred onto a recipient surface, the method employed is not amenable to mixtures, or instances in which other unretained compounds in a given sample may interfere with the signal obtained from the detector.

Table 4.2. Chromatographic system and method used to quantify the percentage of ammonium nitrate transferred onto each recipient surface.

Liquid Chromatograph	Shimadzu Prominence UFLC
Detector Column	UV-Vis (SPD-20A)
Column Flowrate	Acclaim Explosives E2 (3.0 x 150 mm, 3 µm, 120 Å)
Flowrate	0.3250 mL min^{-1}
Mobile Phase	48:52 (v/v) of MeOH/water
Gradient	Isocratic
Injection Volume	2.0 µL
Temperature	30°C
Detection	214 nm

In order to quantify the amount of ammonium nitrate present in each extract, a ten-point calibration curve was established (concentration range: 1 mg/L to 1,000 mg/L). Each of the ten calibration standards were injected onto the column in triplicate and the areas of the resultant peaks recorded. The peak areas were then plotted against the concentration of each calibration standard and the linearity evaluated (**Fig. 4.5**). The resultant R^2 coefficient of determination (0.999) indicted a linear relationship between concentration and peak area over the range tested. Two calibration checks were performed to assess the reliability of the calibration curve in quantifying ammonium nitrate. This was carried out using two independently prepared solutions at concentrations of 350 mg/L and 800 mg/L. The accuracy, with respect to determining the true concentration of ammonium nitrate in a sample, was determined to be 99.1% and 99.6% respectively. Using the established calibration curve (y = 2.941(x) + 5.193), the concentration of ammonium nitrate in each extract was calculated by correlating its resultant peak area to a concentration.

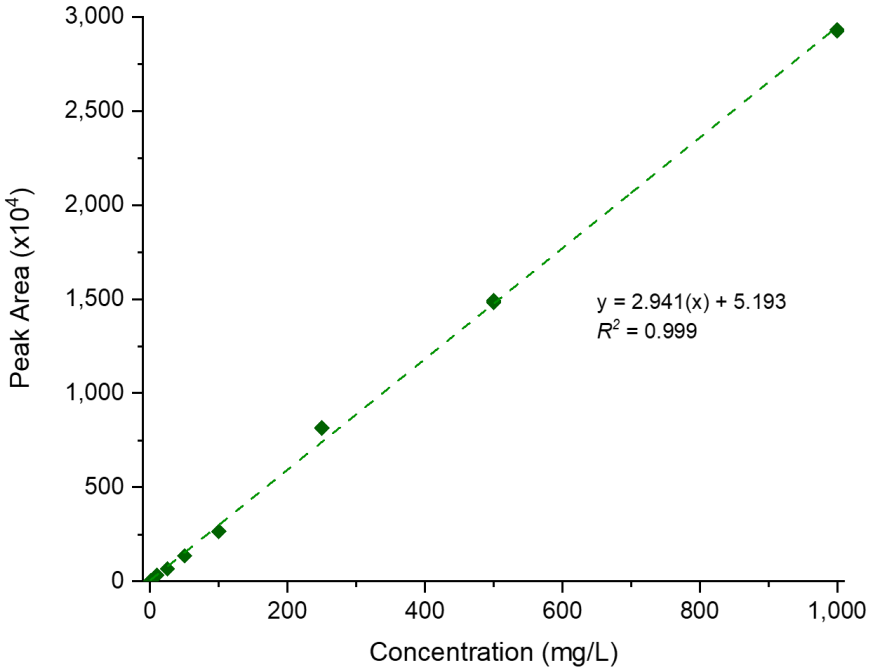


Figure 4.5. LC calibration curve of ammonium nitrate (reference standard).

4.2.6 Negative Controls

A series of negative controls were carried out to ensure that the response obtained from the analysis was a direct result of ammonium nitrate being detected and not a background contaminate present on the transfer and recipient surfaces. MeOH washes of each surface type were analysed along with a MeOH blank and water blank. No measurable response at 1.8 minutes was observed in any of the negative controls.

4.2.7 Statistical Analyses

Data was analysed using IBM SPSS Statistics (version 26). A one-way ANOVA was conducted to determine if the mean amount of ammonium nitrate transferred significantly ($p < .05$) differed with increasing force of contact, duration of contact, or extent of rotation applied during contact. Regression models were then fitted to sample data to assess if a relationship existed between the three contact parameters (contact force, duration of contact, and extent of rotation applied over contact time) and the amount of ammonium nitrate transferred. Based on these models, a two-sample *t*-test (assuming unequal variances) was conducted to compare the mean amount of ammonium nitrate transferred when contact involved rotation, to those

contacts that did not involve rotation. Finally, in order to assess the impact of surface type on the transfer of ammonium nitrate, a two-sample *t*-test (assuming unequal variance) was performed. This was to ascertain if the mean amount of ammonium nitrate transferred onto the recipient surface significantly ($p < .05$) differed when the contact occurred with either a porous or non-porous surface. Cohen's *d*, the standardised mean difference, was used to measure the effect size. The means plus or minus one standard deviation are presented throughout. An alpha level of .05 (5%) was used for all statistical tests.

4.3 Results

4.3.1 Accuracy of Instron® ElectroPuls™ Test Methods

As this is the first reported use of Instron's ElectroPuls for application to forensic science, it is important to demonstrate the repeatability and reproducibility of the process. Therefore, the following aspects were assessed:

- load accuracy – the load achieved in relation to the method;
- load consistency – fluctuations in load across the hold period;
- rotational accuracy- the rotation angle achieved in relation to the method;
- hold time accuracy- the length of time under prescribed load in the method.

In all tests, the load delivered was within 1.6% of the load requested. In the experimental setup, the actuator needed to be lowered down on to the sample, meaning it had to travel in free space before contact with the two surfaces occurred (**Figure 4.6**). As a consequence, the initial load at the point of contact was slightly higher than the load requested by the method. This was followed by an automatic adjustment by the instrument to compensate for this and is likely to be the reason for the small variation in loads observed. Once the hold level was reached, the load during the entire hold period was stable to within < ±0.5%: 10.03 ± 0.03 N; 60.60 ± 0.04 N; 121.13 ± 0.07 N; and 242.03 ± 0.13 N (**Figure 4.7**). In the same way, the set rotation rates of 3°/s, 6°/s, 9°/s, and 12°/s were constant ($R^2 < .99$) (**Figure 4.8**). The duration of the hold periods was confirmed by the time length of the hold step. **Table 4.3** demonstrates that the hold time for each test achieved the desired time within an average of 0.09 ± 0.01 seconds over the hold length.

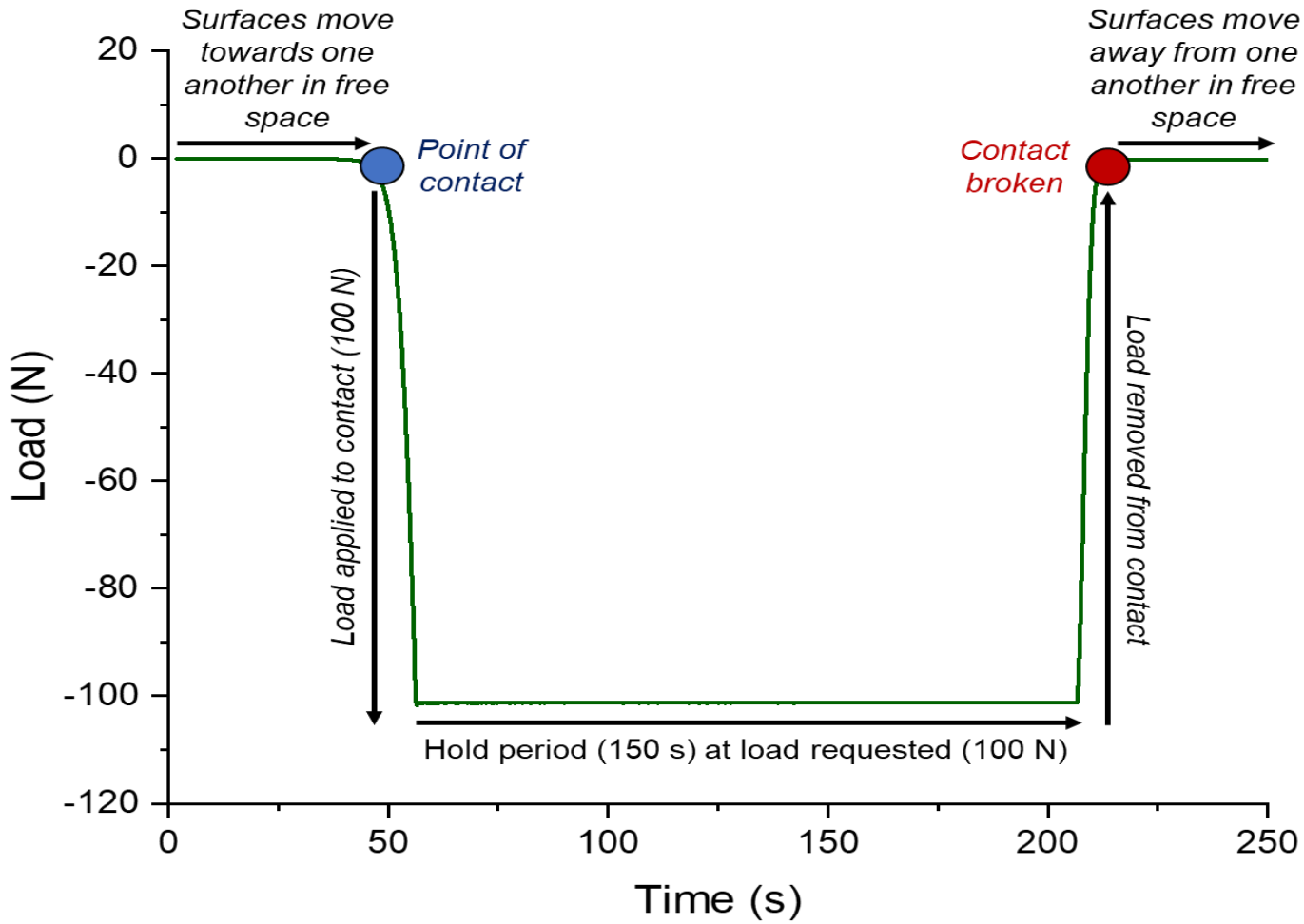


Figure 4.6. Annotated exemplar load *versus* time plot from data acquired by the ElectroPuls E3000 showing the execution of the method in which two surfaces were brought into contact at a specified force (100 N) and held in contact for a prescribed period of time (150 s).

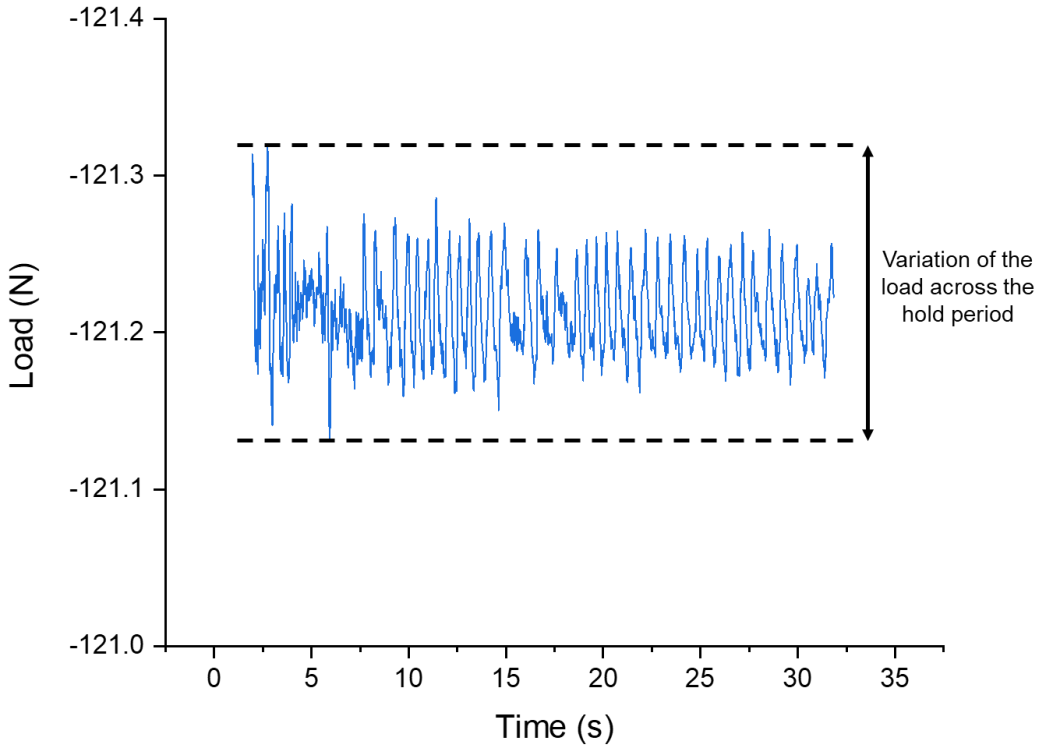


Figure 4.7. Annotated load *versus* time plot for contact occurring at 120 N. The plot has been truncated to show the load recorded over the hold period.

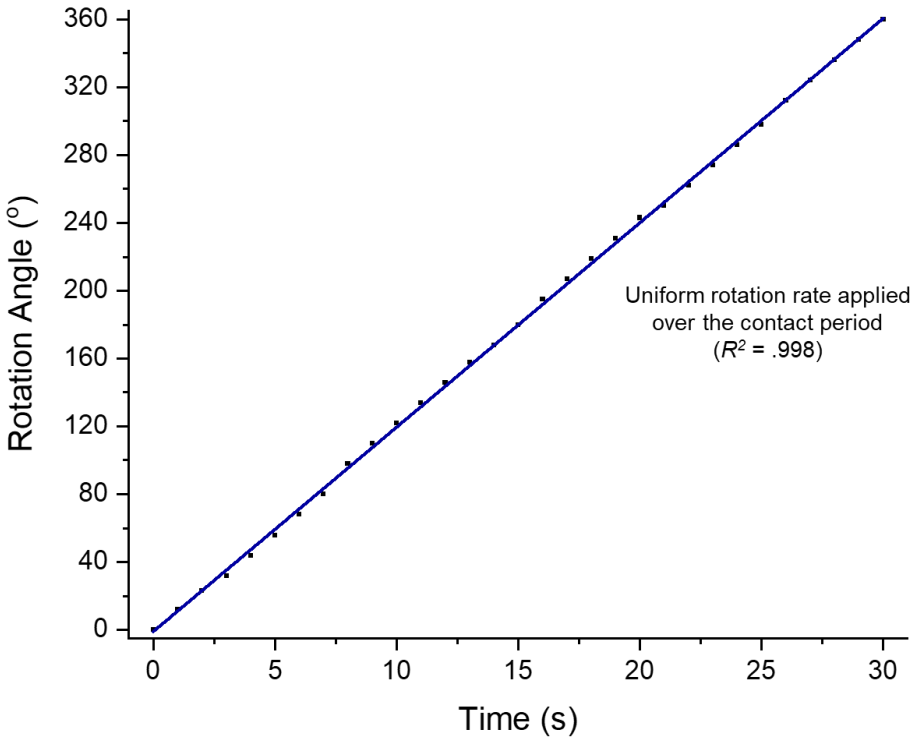


Figure 4.8. Exemplar rotation angle *versus* time plot. A uniform rotation rate (12°/s) was achieved over the hold period (30 s) to deliver 360° of rotation.

Table 4.3. The actual hold times as compared to each of the four programmed times. %CV denotes coefficient of variation expressed as a percentage.

	Actual hold time (s)						
Prog. hold time (s)	**Run 1**	**Run 2**	**Run 3**	**Run 4**	**Run 5**	**Run 6**	**Avg. ± SD (%CV)**
2.00	2.11	2.09	2.11	2.09	2.10	2.09	2.10 ± 0.01 (0.43%)
60.00	60.10	60.09	60.08	60.10	60.09	60.09	60.09 ± 0.01 (0.01%)
150.00	150.08	150.07	150.09	150.08	150.08	150.11	150.09 ± 0.01 (0.01%)
300.00	300.12	300.09	300.09	300.10	300.06	300.08	300.09 ± 0.02 (0.01%)

4.3.2 Extraction Efficiency

The extraction efficiency of the method employed to recover the ammonium nitrate transferred onto the recipient surface was determined to be 26.1 ± 0.6% (**Table 4.4**). As such, the measured amount of ammonium nitrate in each sample was adjusted, accounting for the extraction efficiency, to provide a more accurate representation of the actual amount of ammonium nitrate that was transferred. Although a relatively low recovery was obtained, this was not unexpected as other studies have also highlighted the difficulties of recovering explosive compounds from porous surfaces (Yu *et al*., 2016, 2017). Despite this, the extraction method provided consistent results (%CV = 2.3%) and as such, enabled meaningful comparisons with respect to the transfer of ammonium nitrate (expressed as a percentage) under different testing parameters.

Table 4.4. The recovery efficiency of the extraction technique employed prior to analysis in order to assess the average amount of ammonium nitrate recovered during this process as well as the repeatability of the extraction method employed. Units of concentration are mg/L.

Leather Sample	**Conc. Spotted**	**Conc. Recovered**	**Recovery (%)**	**Avg. Recovery ± SD (%CV)**
1	1000.0	260.7	26.1	26.1 ± 0.6 (2.3%)
2	1000.0	254.3	25.4	
3	1000.0	265.8	26.6	
4	1000.0	268.7	26.9	
5	1000.0	257.1	25.7	

4.3.3 The Impact of Force on the Transfer of Ammonium Nitrate

As the surface area of contact remained constant, the pressure was considered to be the best representation of force. The surface area for each sample was 1 cm^2 and therefore the pressures exerted ranged from 100 kilopascals (kPa) (when 10 N was applied) to 2,400 kPa (when 240 N was applied). As the average pressure of contact during a handshake for adult males is between 550 kPa and 690 kPa (De Boos, 2019), the pressures assessed are representative of real-world scenarios.

Ammonium nitrate was readily transferred from both porous and non-porous surfaces onto the recipient surface, with non-porous surfaces consistently transferring more ammonium nitrate than porous surfaces (**Table 4.5**). The average percentage of ammonium nitrate transferred onto the recipient surface following contact with a porous surface was 11.5 ± 1.9%. For contact with non-porous surfaces this was 38.8 ± 2.3%. The amount of transfer was significantly different between the porous and non-porous surfaces ($t(22) = 30.24$, $p < .001$, $d = 13.47$), but the amount of ammonium nitrate transferred did not depend on the force of contact (non-porous surfaces: $R^2 = .17$, $F(1, 10) = 2.12$, $p = .176$; porous surfaces: $R^2 = 0.08$, $F(1, 10) = 0.88$, $p = .371$) (**Fig. 4.9**).

Table 4.5. Quantification of the percentage of ammonium nitrate transferred (%T) onto the recipient surface (human skin analogue) following contact with an adulterated surface, either MDF (porous surface) or stainless steel (non-porous surface), at four different contact forces.

	Porous (MDF)		Non-Porous (Stainless Steel)	
Force (N)	**%T**	**Avg. %T ± SD (%CV)**	**%T**	**Avg. %T ± SD (%CV)**
10	11.2 10.5 13.0	11.6 ± 1.1 (9.1%)	36.2 36.0 38.8	37.0 ± 1.3 (3.4%)
60	13.9 14.4 7.7	12.0 ± 3.0 (25.4%)	38.6 37.2 36.8	37.5 ± 0.8 (2.1%)
120	13.9 11.3 11.5	12.2 ± 1.2 (9.7%)	43.1 41.4 39.1	41.2 ± 1.6 (4.0%)
240	9.7 10.8 10.3	10.3 ± 0.4 (4.4%)	37.1 42.6 38.2	39.3 ± 2.4 (6.0%)

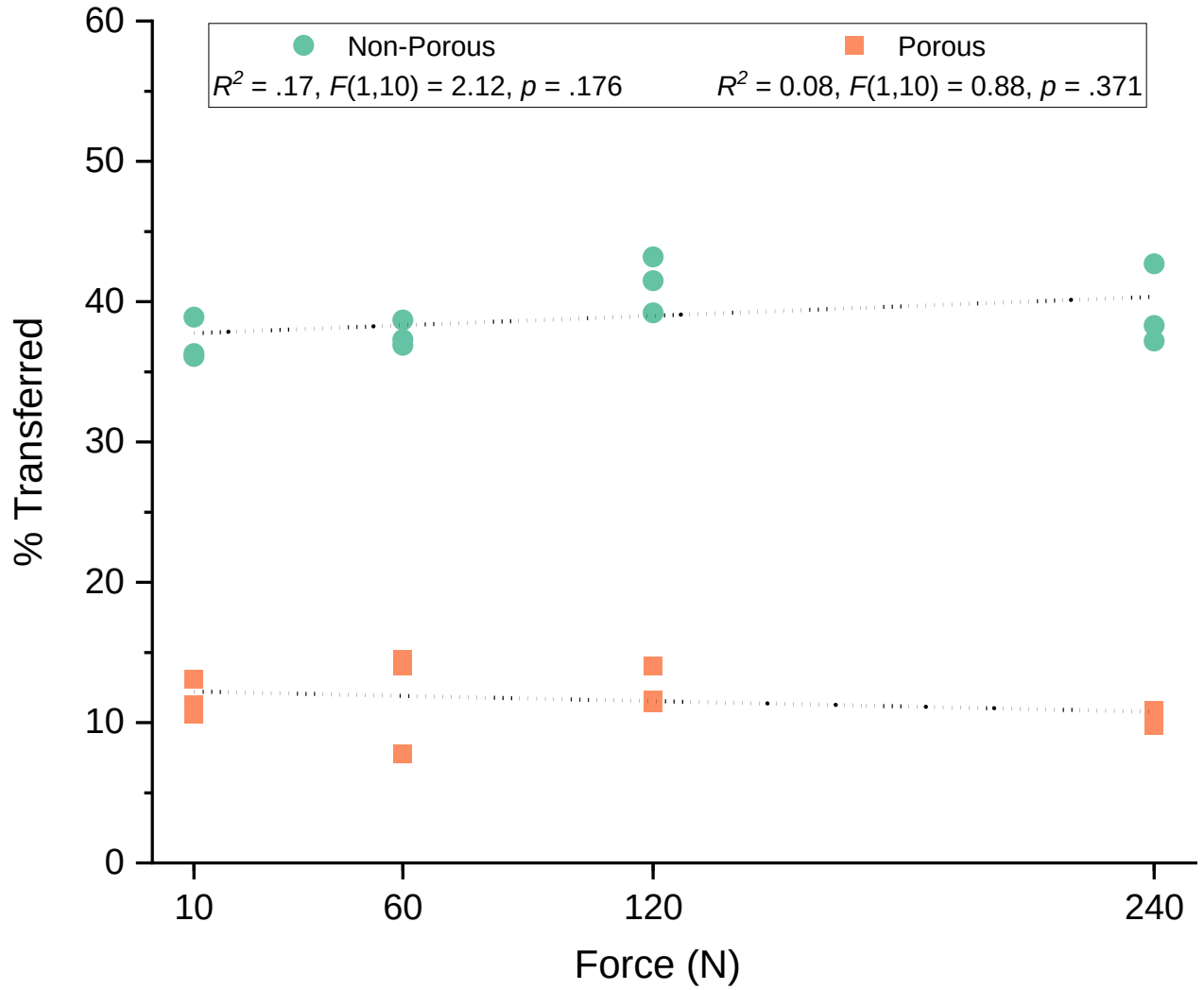


Figure 4.9. The percentage of ammonium nitrate transferred onto the recipient surface following contact with an adulterated porous or non-porous surface at four different contact forces. No trend was observed.

4.3.4 The Impact of Time on the Transfer of Ammonium Nitrate

The contact times selected in this study provide a range, from a brief contact (2 s), such as that involved by brushing against a surface or a handshake, to an extended contact (300 s), in order to encompass a wide array of contact scenarios that might be encountered throughout routine day-to-day activities.

As was observed with force, non-porous surfaces consistently transferred more ammonium nitrate than porous surfaces ($t(22) = 15.26$, $p < .001$, $d = 7.33$) (**Table 4.6**). The average recovery from contact with a porous surface was 11.4 ± 3.0% and 34.0 ± 3.9% from contact with an adulterated non-porous surface. The amount of ammonium nitrate transferred did not depend on contact time (non-porous surfaces: $R^2 = 0.02$, $F(1, 10) = 0.22$, $p = .653$; porous surfaces: $R^2 = 0.08$, $F(1, 10) = 0.86$, $p = .374$) (**Fig. 4.10**).

Table 4.6. Quantification of the percentage of ammonium nitrate transferred (%T) onto the recipient surface (human skin analogue) following contact with an adulterated surface, either MDF (porous surface) or stainless steel (non-porous surface), at four different contact times.

	Porous (MDF)		Non-Porous (Stainless Steel)	
Time (s)	**%T**	**Avg. %T ± SD (%CV)**	**%T**	**Avg. %T ± SD (%CV)**
2	15.0 8.0 15.3	12.8 ± 3.4 (26.4%)	30.6 29.9 33.2	31.2 ± 1.4 (4.5%)
60	12.0 15.3 7.5	11.7 ± 3.2 (27.6%)	34.4 34.4 38.3	35.7 ± 1.8 (5.1%)
150	10.6 12.0 10.4	11.1 ± 0.7 (6.5%)	33.9 38.6 42.8	38.4 ± 3.6 (9.5%)
300	8.7 14.8 7.5	10.4 ± 3.2 (30.9%)	31.2 29.8 30.8	30.6 ± 0.6 (1.9%)

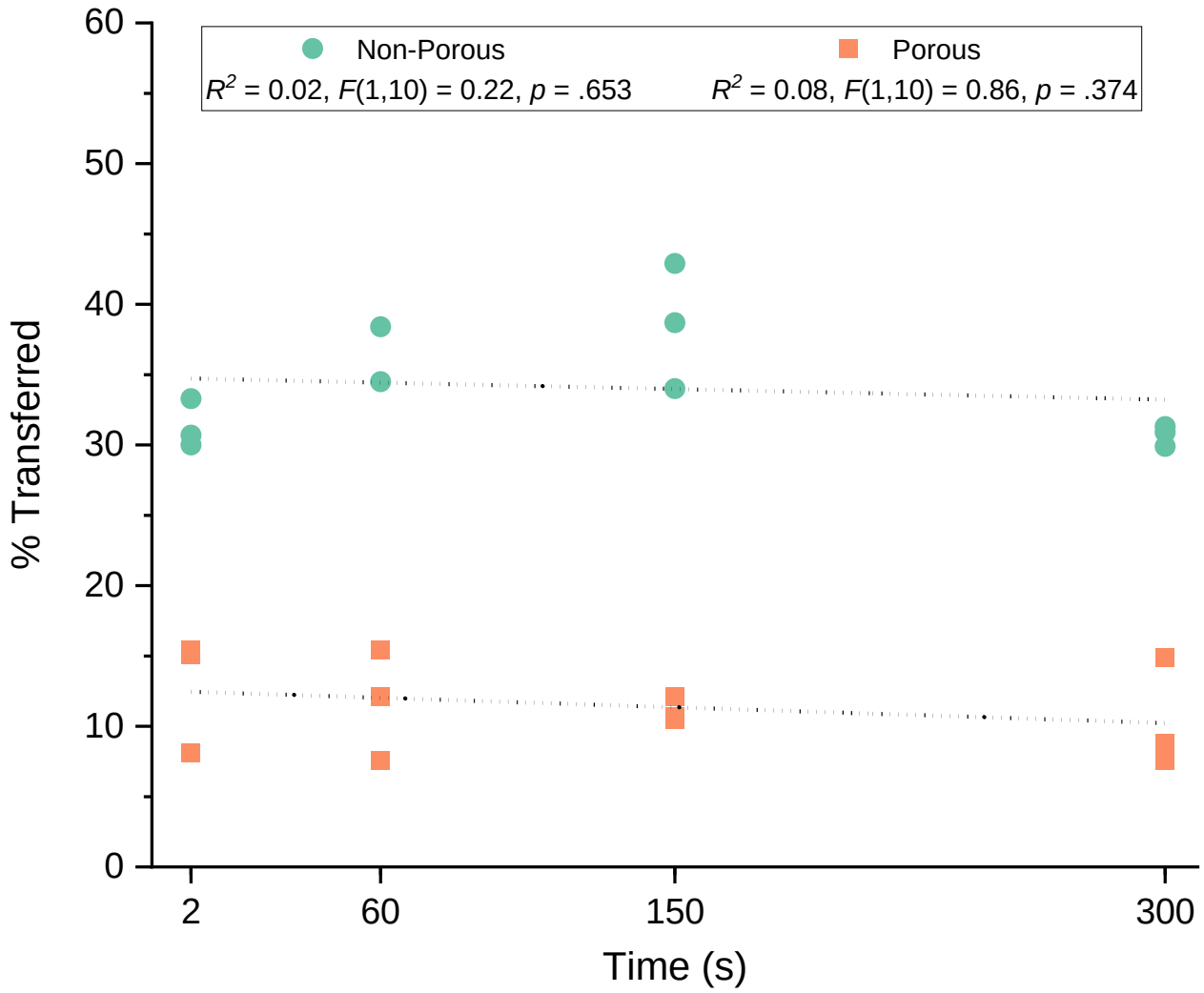


Figure 4.10. The percentage of ammonium nitrate transferred onto the recipient surface following contact with an adulterated porous or non-porous surface at four different contact times. No trend was observed.

4.3.5 The Impact of Rotation on the Transfer of Ammonium Nitrate

As well as force and time, contact is also likely to involve rotation; a variable that has not previously been independently assessed with respect to the transfer of forensic materials. Again, non-porous surfaces consistently transferred more ammonium nitrate than porous surfaces ($t(22) = 27.80$, $p < .001$, $d = 12.33$) (**Table 4.7**). The average percentage of ammonium nitrate transferred onto the recipient surface following contact with a porous surface with rotation applied was 22.2 ± 2.3%. For contact with non-porous surfaces this was 51.6 ± 2.7%. Although the amount of ammonium nitrate transferred did not depend upon the extent of rotation applied (non-porous surfaces: $R^2 = 0.13$, $F(1, 10) = 1.54$, $p = .244$; porous surfaces: $R^2 = 0.19$, $F(1, 10) = 2.37$, $p = .155$) (**Fig. 4.11**), contact which involved any amount of rotation resulted in a significantly larger transfer of ammonium nitrate (non-porous: $t(34) = 11.63$, $p < .001$, $d = 4.11$; porous surfaces: $t(34) = 12.09$, $p < .001$, $d = 4.27$) when compared to contacts occurring without rotation (**Fig. 4.12**).

Table 4.7. Quantification of the percentage of ammonium nitrate transferred (%T) onto the recipient surface (human skin analogue) following contact with an adulterated surface, either MDF (porous surface) or stainless steel (non-porous surface), with four different rotations applied during contact.

	Porous (MDF)		Non-Porous (Stainless Steel)	
Rotation (°)	**%T**	**Avg. %T ± SD (%CV)**	**%T**	**Avg. %T ± SD (%CV)**
90	19.6	20.9 ± 2.5 (11.8%)	48.4	50.5 ± 2.2 (4.4%)
	18.7		53.5	
	24.3		49.3	
180	19.4	21.7 ± 2.3 (10.4%)	50.0	51.0 ± 1.7 (3.3%)
	21.0		49.4	
	24.8		53.2	
270	20.5	22.4 ± 1.6 (7.3%)	56.6	52.5 ± 3.5 (6.6%)
	24.5		48.1	
	22.3		52.6	
360	21.4	23.6 ± 1.8 (7.6%)	50.9	52.9 ± 2.1 (4.0%)
	25.8		55.7	
	23.7		51.7	

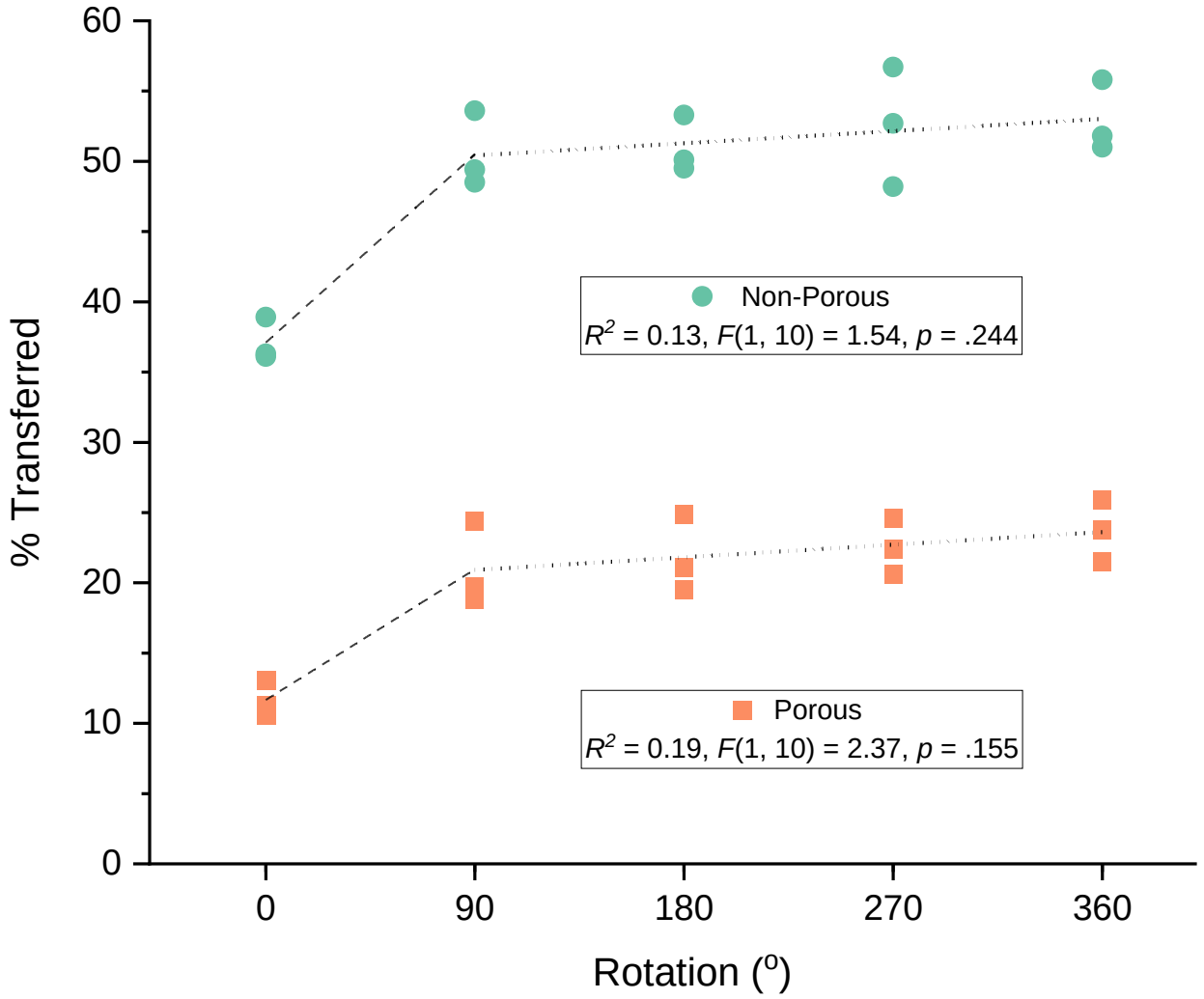


Figure 4.11. The percentage of ammonium nitrate transferred onto the recipient surface following contact with an adulterated porous or non-porous surface without rotation and with four rotations applied during contact. The application of rotation during contact was significant; however, no trend was observed with additional rotation applied.

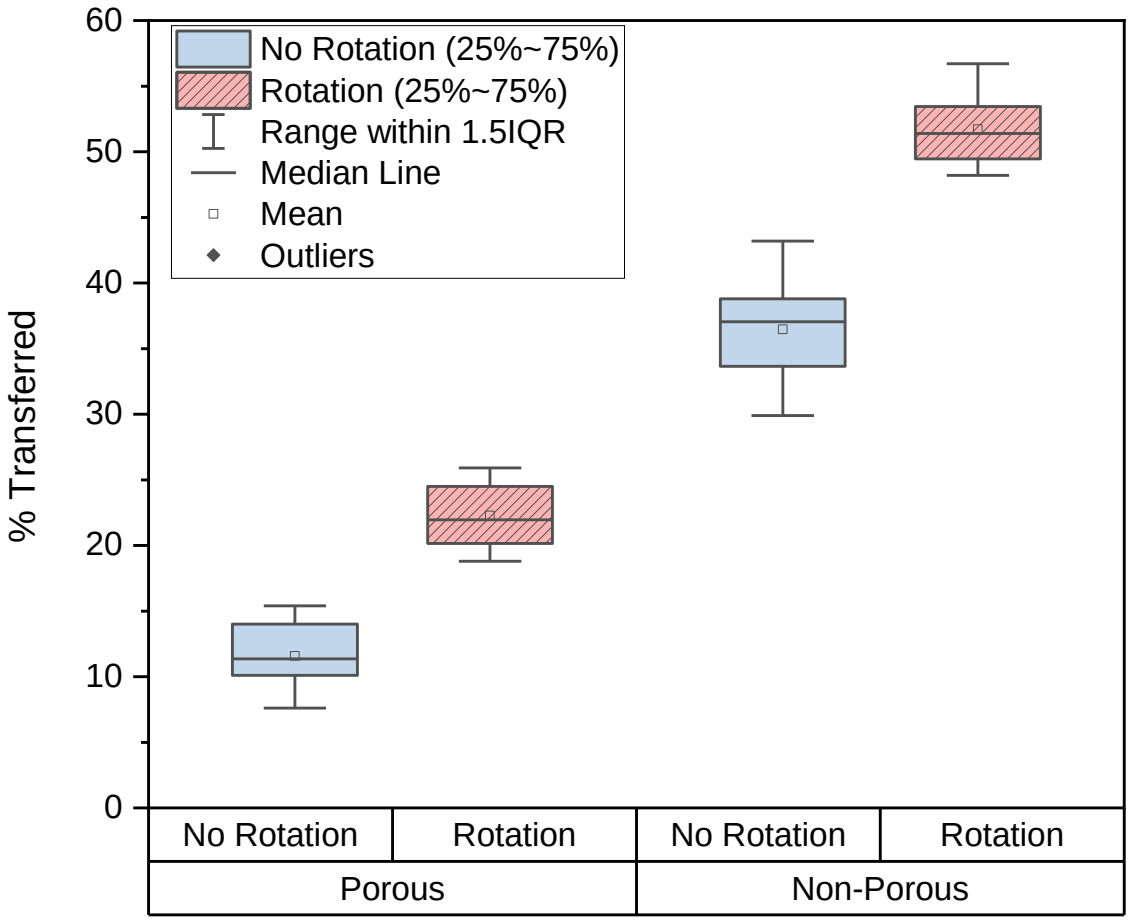


Figure 4.12. Box plot comparing the amount of ammonium nitrate transferred onto a recipient surface from an adulterated porous or non-porous surface across contacts occurring without rotation and those with rotation.

4.3.6 Summary of Results

There was a substantial difference in the percentage of ammonium nitrate particulates transferred from porous and non-porous surfaces (**Fig. 4.13**). On average, 41.5 ± 8.0% of the ammonium nitrate particulates on adulterated non-porous surfaces transferred onto a recipient surface through contact. The average transfer from adulterated porous surfaces was 15.0 ± 5.6%; approximately 25% fewer particulates than transferred from non-porous surfaces. The amount of ammonium nitrate transferred onto a recipient surface from an adulterated porous or non-porous surface across all of the different contact times, forces, and rotations is summarised in **Fig. 4.14**.

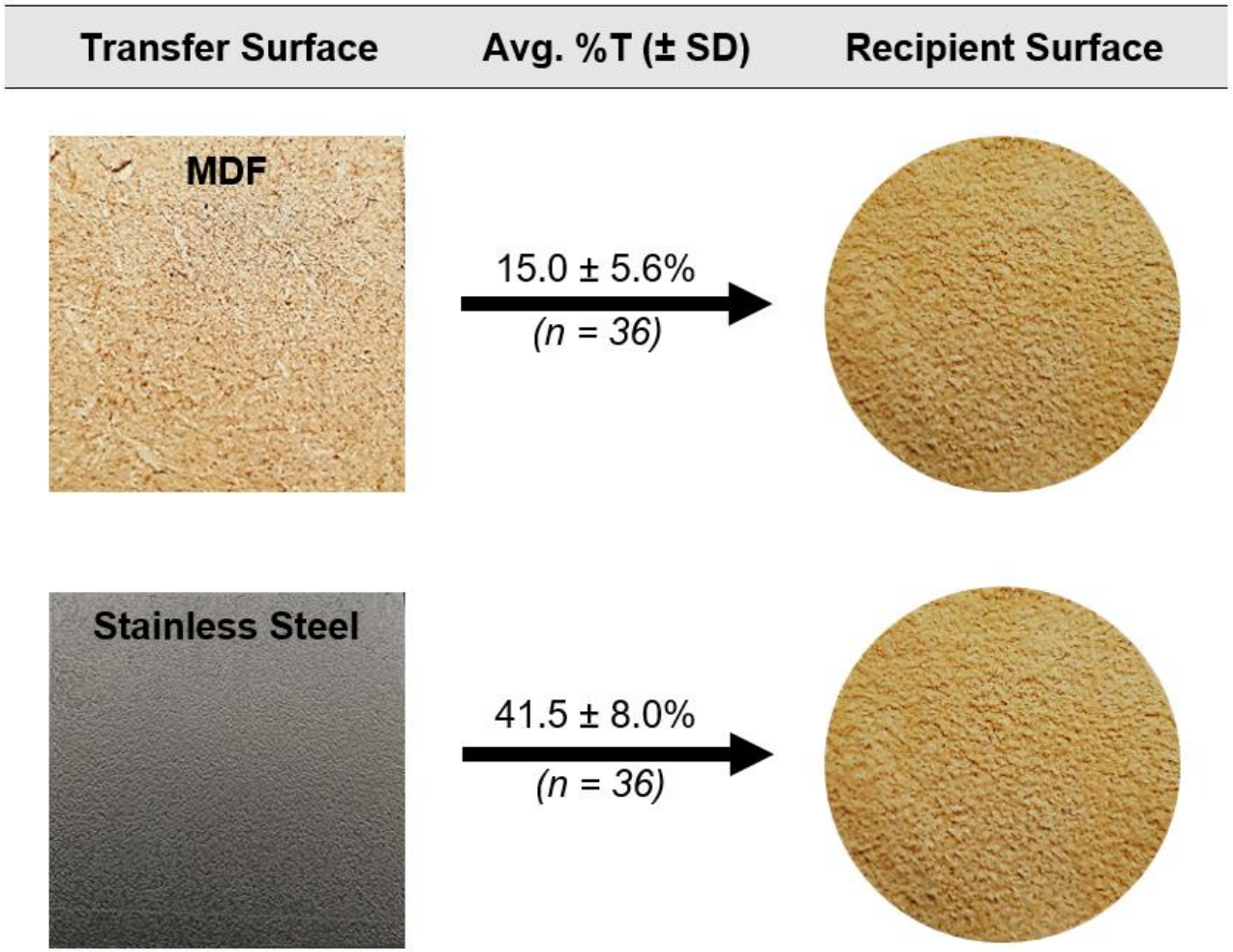


Figure 4.13. Comparison of the impact of surface type on the transfer of ammonium nitrate. The average percentage transfer obtained across all experiments for that surface type are presented.

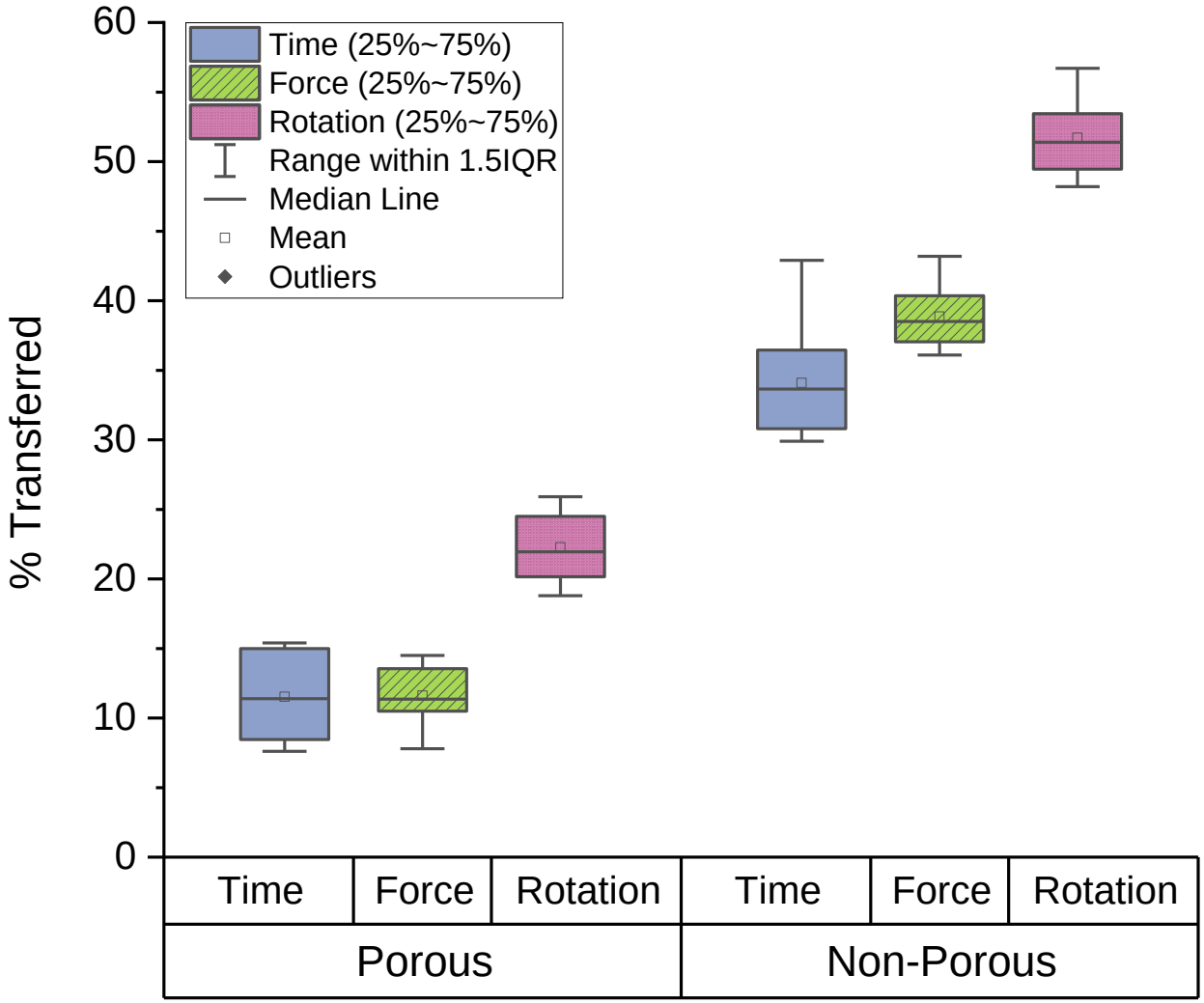


Figure 4.14. Box plot comparing the amount of ammonium nitrate transferred onto a recipient surface from an adulterated porous or non-porous surface across different contact times, forces, and rotations.

4.4 Discussion

We have demonstrated a reliable and repeatable method to assess the transfer of particulates from one surface to another, in a manner that ensures accurate control over the contact force, duration of contact, and amount of rotation applied over the duration of contact. This is the first reported study on the use of the ElectroPuls E3000 for forensic applications and opens up the opportunity for using similar devices to isolate and explore fundamental aspects of transfer in forensic research.

In all scenarios, and under all of the testing parameters assessed, quantifiable amounts of ammonium nitrate were recovered on the recipient surface. This highlights that ammonium nitrate is readily transferred from one surface to another, even with low pressure (100 kPa when 10 N was applied across a 1 cm^2 surface) and short duration (2 s). A greater transfer of ammonium nitrate was observed when the transfer originated from an adulterated non-porous surface. This was expected and is in agreement with other studies that have assessed the impact of surface type on the transfer and recovery of forensic materials (Goray *et al*., 2010a, 2010b; Yu *et al*., 2016, 2017; Sisco *et al.*, 2018). It is likely that this is due to the surface topography (as shown in the SEM micrographs of the surfaces (**Fig. 4.15**)) allowing particulates of ammonium nitrate to be retained within the pores of the MDF surface. Additionally, as the recovery surface (leather) was also porous, it is likely that this is what accounted for the low recovery observed in the extraction phase with direct spiked samples.

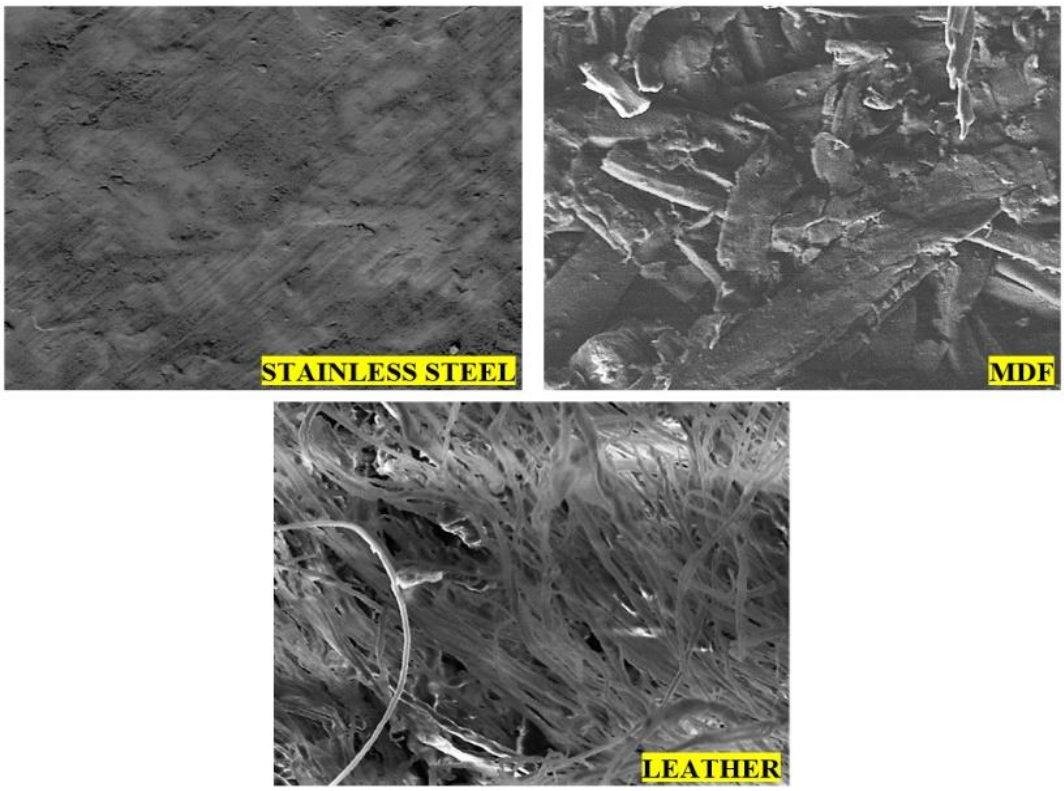


Figure 4.15. SEM micrographs of the two transfer surfaces (top) and the recipient surface (bottom) at 500x magnification (HV = 18.0 kV; WD = 14 mm). Micrographs acquired using a Pemtron PS-230 scanning electron microscope fitted with an Oxford Instruments X-act energy-dispersive X-ray spectrometer system. Surfaces were sputter coated (time = 2 min; vacuum = 0.2 Torr; current = 0.2 mA) with gold prior to analysis.

For both porous and non-porous surfaces, the force and duration of contact did not significantly impact the amount of ammonium nitrate transferred onto the recipient surface. This is in contrast to the work conducted by Gherghel *et al.*, where an increase in transfer was observed with increasing contact times (Gherghel *et al*., 2016) and a decrease over time when contact involved friction (Gherghel *et al*., 2019). However, these studies involved the transfer of volatile organic compounds rather than solid particulates which is likely to account for the difference in findings. The impact of pressure has also been assessed with respect to the transfer of DNA, whereby it was found that a larger portion of DNA is transferred with increasing pressure both directly from skin onto a surface (Tobias *et al*., 2017) as well as between two surfaces (Goray *et al*., 2010a). Again, these findings contrast with the results presented here and further highlights the need to understand the transfer dynamics of different forensic trace materials.

When contact involved rotation, a statistically larger transfer of ammonium nitrate was observed, compared to those contacts not involving a rotation. However, this was not dependent upon the extent of rotation applied. This suggests that any rotation at all will increase transfer, but once transfer has occurred, any additional rotation has no measurable effect. This is an important finding given that contacts, in most instances, will inherently involve an amount of rotation.

In order to build a robust empirical evidence base, a larger surface set should be assessed under similar testing conditions using a variety of surface types that one might encounter in a given day, and in certain scenarios. Additionally, the impact of these variables on the transfer of other evidence types (for example gunshot residue, pollen, glass, fibres, and drug residues (Chapter 5)) needs to be assessed in order to better understand transfer dynamics across a wide range of evidence types in order to aid and assist with forensic reconstructions. Other factors likely to impact upon the transfer of forensic materials, such as the particle size of the material being transferred and the electrostatic forces involved, are also important to consider in understanding evidence dynamics and warrant future investigation. Additionally, once transferred, the persistence of that material on a surface then becomes an important consideration.

Although the results of this study are akin to secondary transfer scenarios, this method can be expanded to include tertiary transfer scenarios. As such, this opens up an opportunity for having a powerful tool to assess the primary, secondary, and tertiary transfer dynamics of a number of different evidence types across different combinations of surface types.

This work provides a starting point for future studies in which controlled experimental studies addressing specific aspects of trace evidence dynamics can be conducted to better understand the individual impact that specific variables have on the transfer of trace evidence. Establishing the contribution of individual variables opens up possibilities for creating simulation models that can be adapted for specific crime reconstruction contexts.

4.5 Conclusion

This study sought to assess the impact of three variables, contact force, duration of contact, and the amount of rotation applied during contact, on the transfer of ammonium nitrate. Using the Instron's ElectroPuls E3000, a reliable and repeatable method was established to assess these variables independently by having automated control of the parameters in which the contact between the transfer and recipient surfaces occurred. The amount of ammonium nitrate transferred onto the recipient surface from both a porous and non-porous surface as a function of each of the three variables was then quantified. The results from this study have demonstrated that:

- ammonium nitrate is readily transferred from both porous and non-porous surfaces over a range of contact forces, durations of contact, and rotations applied during contact;
- a greater transfer of ammonium nitrate was observed when the transfer originated from an adulterated non-porous surface;
- when contact occurred and involved rotation, a larger transfer of ammonium nitrate was observed, compared to contact not involving a rotation.

The results from this study provide empirical data on the independent impact of force, time, and rotation on the transfer of ammonium nitrate. This study highlights that when contact occurs, even with the smallest forces and shortest contact times, it is possible that detectable amounts of the explosive compound transferred can be recovered. These findings indicate that there is value for broader crime reconstruction endeavours in taking a reductionist approach when seeking to understand the mechanics of trace transfers. The potential such insights offer include the creation of simulation models where specific parameters can be adjusted for a given case in which the transfer of explosive residues, as well as other evidence types, may have occurred. The creation of such datasets will be valuable for modelling the movements of traces in order to enable more transparent and reproducible interpretations of pertinent trace materials in crime reconstructions going forward.

Chapter 5

Cocaine on Banknotes: Evidence Dynamics Case Study

A significant proportion of banknotes in worldwide circulation are contaminated with cocaine…The analysis of drug traces on banknotes…can provide important information about the types of substances that are used in a geographical region. Banknotes can also be contained during drug deals, but the most important cause of contamination is their direct use, after being rolled up, to snort illicit substances. (Troiano et al., 2017; p. 1098)

Outline

It is well established that a large proportion of paper banknotes in circulation contain traces of cocaine. Being able to discriminate between the innocent transfer of illicit drug particles acquired through everyday interactions with surfaces such as banknotes, as opposed to transfer resulting from criminal activities can provide valuable intelligence that can inform an investigation. With many countries adopting polymer banknotes as legal tender, it is important to consider the transfer of cocaine from these surfaces as well as the retention of these particulates on polymer banknotes for evaluative interpretation in crime reconstruction. This comparison study assessed three contact variables (force, time, and rotation) on the transfer of cocaine particulates from paper and polymer banknotes onto a human skin proxy. The persistence of cocaine particulates was assessed through a realistic scenario which mimicked a cash transaction. Quantifiable amounts of cocaine were transferred onto the human skin proxy across all the contacts assessed, with a greater transfer observed with contacts involving polymer banknotes and those contacts which involved rotation. Following extensive handling, cocaine persisted on both banknote types, with paper banknotes retaining larger amounts of cocaine than polymer banknotes. These findings show that cocaine can persist on both paper and polymer banknotes for extended periods of time following handling and is therefore available for transfer. This transfer then readily occurs, even when contact is brief and involves relatively small forces. A key distinction between the banknote types was that cocaine particulates are more likely to transfer from polymer banknotes due to the lower retention rate of particulates on this surface. Such insights can aid in

evaluating the relevance of illicit drug particles identified on items or persons of interest in crime reconstruction approaches.

5.1 Introduction

In 2019, an estimated 20 million people worldwide used cocaine (0.4% of the total population between 15 and 64 years old) (United Nations Office on Drugs and Crime, 2021). Over the same period, cocaine (benzoylmethylecgonine) (**Fig. 5.1**) was the second most commonly used illicit drug in Europe, with 3.5 million users (1.2% of the total population aged 15 to 64) (European Monitoring Centre for Drugs and Drug Addiction, 2021). Cocaine, a Class A drug under the UK Misuse of Drugs Act (1971), is a common environmental contaminate, with cocaine traces found on approximately 13% of fingerprints from self-reported drug-free volunteers (Ismail *et al*., 2018) and on many paper banknotes in circulation (Hudson, 1989; Oyler *et al*., 1996; Negrusz *et al*., 1998; Sleeman *et al*., 2000; Jenkins, 2001; van der Heide & Russell, 2016; Poupko *et al*., 2018; Pinorini *et al*., 2020). A person handling adulterated banknotes is therefore likely to receive traces of drugs onto their hands. Historically, banknotes have been made from cotton and linen papers (cellulose fibres), a porous substrate (Daniels, 1996) but since Australia replaced paper banknotes with polymer banknotes in 1996 (Wilson, 1998), other countries have either adopted, or have started to adopt, polymer banknotes as legal tender (Jones *et al*., 2003; Singh, 2008; Spencer, 2011; McClintock & Whymark, 2016). These polymer banknotes are commonly produced using biaxially oriented polypropylene (BOPP), a non-porous substrate (Scotcher & Bradshaw, 2018).

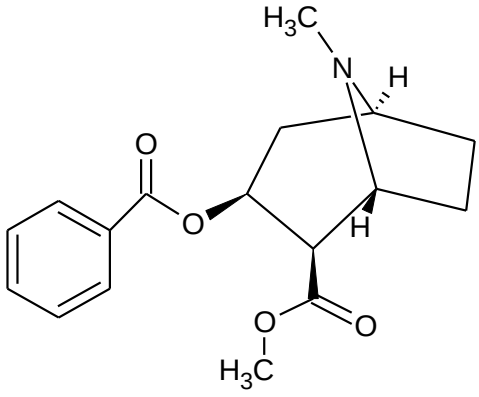


Figure 5.1. Chemical structure of cocaine ($C_{17}H_{21}NO_4$), a naturally occurring alkaloid found in the leaves of the coca plant (*Erythroxylum coca* and *Erythroxylum novogranatense*).

Despite the decline of cash economy (Cohen *et al*., 2020), cash is still often used in organised crime transactions (Riccardi & Levi, 2018), and the change from paper to polymer banknotes is likely to impact upon the transfer and persistence of cocaine traces on and to these surfaces. Being able to discriminate between the innocent transfer of drug traces acquired from contaminated surfaces, such as banknotes, as opposed to transfer resulting from illicit activities is of relevance to crime reconstructions as particle traces provide valuable intelligence that can inform an investigation (Morgan, 2017a). It is important to be able to identify traces, and also to understand the dynamics of those traces to establish how and when they have been transferred to an object or a person (Chisum & Turvey, 2000; French *et al*., 2012). Empirical studies assessing the dynamics of how these traces may have been transferred, under what conditions they may persist, and over what timescales can these traces persist, can provide contextual information and enable robust crime reconstructions (Morgan *et al*., 2020; Trejos *et al*., 2020).

This chapter presents an experimental study which was designed to compare the transfer and persistence of cocaine particulates on paper and polymer banknotes that were intentionally adulterated with controlled amounts of cocaine. A reductionist approach was taken to understand the individual impact that time, force, and rotation have on the percentage of cocaine transferred from paper and polymer banknotes onto a human skin proxy. The duration of contact, force of contact, and rotation applied during contact were controlled using an Instron ElectroPuls E3000, a method previously used to characterise transfer of explosive particulates (Amaral *et al.*, in press). To assess persistence, paper and polymer banknotes were handled up to fifty times, and the amount of cocaine present on each banknote quantified following each prescribed handling period. Results from studies such as this can aid in developing an understanding of the proportion of cocaine that can be transferred by direct handling of adulterated banknotes under different testing parameters as well as how long these particulates will persist on banknotes following repeated handling. These insights can then be used to create an evidence-based approach for the interpretation of trace particles such as cocaine in crime reconstructions.

5.2 Materials and Methods

5.2.1 Banknotes

Foreign currency banknotes were drawn over the counter from a local eurochange® (Crickhowell, UK). For this study, paper US one-dollar banknotes and polymer Vietnamese twenty thousand đồng banknotes were used (see **Fig. 5.2**). Of the paper currency issued in the US, the one-dollar banknote, made from a blend of 25% linen and 75% cotton, is the most common banknote in circulation (United States Federal Reserve System, 2021). Vietnam was an early large-scale adopter of polymer banknotes (Goher, 2012; CCL Secure, 2021) and the twenty thousand đồng banknote, made from BOPP, was selected as having an approximate equivalent value (~ $0.9 USD). To remove any previously transferred particulates each note was twice washed with isopropyl alcohol (Lachance *et al*., 2016), and subsequently allowed to dry, before the experiment was undertaken; cocaine was not detectable in the negative controls. It was outside the remit of this study to assess the prior odds of cocaine contamination (background rates) on banknotes.

Figure 5.2. Paper US one-dollar (1 USD) (left) and polymer Vietnamese twenty thousand đồng (20,000 VND) (right) banknotes used in this study.

5.2.2 Transfer Study

With such a high number of banknotes in circulation adulterated with cocaine traces, coupled with the frequency with which banknotes are handled in society, drug traces are likely to be transferred onto the hands of individuals who make contact with banknotes. As such, this study was designed to quantify the amount of cocaine transferred onto a human skin proxy from paper and polymer banknotes following a controlled contact. Chamois leather (Amazon, UK) was used as a proxy for human skin and was preconditioned by wearing under clothing for a period of eight hours to transfer dermal oils. This material was chosen as it has been shown to mimic both

the mechanical and frictional contact behaviour of human skin (Dąbrowska *et al*., 2016; Fenton *et al*., 2020). All leather samples were cut into 1 cm^2 circles (n = 90).

5.2.2.1 Adulteration of Banknotes with Cocaine Hydrochloride

A 1,000 mg/L working solution of cocaine hydrochloride (HCl) (≥ 97.5% purity; Sigma Aldrich, Gillingham, Dorset, UK) was prepared by dissolving 0.1 grams of cocaine HCl powder in 100 cm^3 of MeOH (HPLC grade; Sigma Aldrich, Gillingham, Dorset, UK). Each banknote was subdivided into ten approximately equal sections (**Fig. 5.3**). To the centre of each section, 250 µL of the working solution was spotted. Banknotes were then left to air dry for one hour to allow the solvent (MeOH) to evaporate and the solute (cocaine HCl) to recrystallize onto the surface.

Figure 5.3. Exemplar template of how each banknote was subdivided into ten sections.

5.2.2.2 Controlled Contact

An Instron ElectroPuls E3000 material testing instrument (Instron, High Wycombe, UK) was used to control the duration of contact, force of contact, and extent of rotation applied during contact (**Table 5.1**). In each run, the adulterated banknote was affixed to the dynamic load cell and the leather recipient surface was affixed to the static load cell of the instrument. Operation was controlled through Instron WaveMatrix software using a three-step method. The first step brought the banknote and leather recipient surface into contact at a speed of 0.05 mm/s. Once contact occurred, the second step controlled the time and force at which the surfaces were held under compression. Rotation was applied by delivering a fixed angular velocity for 4, 8, or 12 s to provide rotation of 120, 240, or 360 degrees. The third step broke the contact between the two surfaces, moving the surfaces away from one another

at a speed of 0.05 mm/s. Each experimental run was assessed five times and the mean and standard deviation calculated.

Table 5.1. The control factors and variables tested in each experimental run. A ±250 N Dynacell (dynamic load cell) was fitted for experiments one through twelve (those assessing the impact of force and time) and a ±5 kN Dynacell, with a torsional range of ±25 Nm, fitted for experiments thirteen through eighteen (those assessing the impact of rotation).

Experiment	Control Factors			Variables	
	Force (N)	Rotation (°)	Recipient Surface	Transfer Surface	Time (s)
1	100	0	Chamois leather	Paper banknote	2
2					60
3					300
4				Polymer banknote	2
5					60
6					300
Experiment	**Control Factors**			**Variables**	
	Time (s)	**Rotation (°)**	**Recipient Surface**	**Transfer Surface**	**Force (N)**
7	30	0	Chamois leather	Paper banknote	10
8					120
9					240
10				Polymer banknote	10
11					120
12					240
Experiment	**Control Factors**			**Variables**	
	Time (s)	**Force (N)**	**Recipient Surface**	**Transfer Surface**	**Rotation (°)**
13	30	100	Chamois leather	Paper banknote	120
14					240
15					360
16				Polymer banknote	120
17					240
18					360

5.2.2.3 Extraction of Cocaine Hydrochloride

To extract the cocaine from each of the leather samples, 1 cm^3 of MeOH was added and the samples were then agitated at 70 rpm for 30 minutes using a Cole-Parmer Stuart See-Saw Rocker. Following this, the leather samples were wrung dry (using forceps that were cleaned between samples) and the extracts transferred into 2 cm^3 autosampler vials. The amount of cocaine present in extract was quantified using gas chromatography with a flame ionization detector (FID) as outlined in **Section**

5.2.4. The extraction efficiency of the method was assessed by spotting five leather samples with 100 µL of the stock solution of cocaine HCl and quantifying the amount recovered following extraction.

5.2.3 Persistence Study

A further study was designed to compare the persistence of cocaine on adulterated paper and polymer banknotes following handling. A realistic scenario was designed to mimic an everyday cash transaction whereby cash is removed from, and returned to, a wallet.

5.2.3.1 Adulteration of Banknotes with Cocaine Hydrochloride

Each banknote was divided into five sections (**Fig. 5.4**) and 250 µL of the working solution of cocaine HCl (1,000 mg/L) spotted onto the centre point of each section. The banknotes were left to air dry for one hour, to allow for the solvent (MeOH) to evaporate and the drug (cocaine HCl) to recrystallise on the surface.

Figure 5.4. Exemplar template of how each banknote was subdivided into five sections.

5.2.3.2 Handling

To assess the persistence of cocaine on banknotes following handling, an adulterated banknote was placed inside a billfold wallet, which was then folded, and subsequently placed inside a trouser pocket. The wallet was then removed from the pocket, unfolded, and the banknote taken out. This process was repeated ten times after which the first section of the banknote was sampled. Subsequent sections of banknote were sampled following the note being handled twenty, thirty, forty, and fifty times. The whole experiment was then replicated to generate data across five runs for each banknote type (paper and polymer). Each banknote type was assessed

independently, and a new banknote used for each replicate (a total of five of each type of banknote). A new billfold wallet was used for each experimental run (total of 10 wallets).

5.2.3.3 Recovery of Cocaine Hydrochloride

Following the method outlined by Amaral *et al.* (2020), a sampling gel was prepared by combining gelatine powder, arrowroot powder, glycerine, and water in a mass ratio of 3:1:1:5. An aqueous gel composition was chosen as the hydrochloride salt of cocaine is water soluble. Using a disposable plastic syringe, a thin layer of gel was applied over the surface area of the banknote section being sampled (~ 19 cm^2). The gel was left to dry for 15 minutes, before being peeled from the surface using forceps, and placed into a beaker. The gel was dissolved in 25 cm^3 of water at 75°C with 0.6 g of powdered bromelain added to prevent re-solidification. Cocaine HCl was isolated from the gelatine-based matrix using SPE (**Fig. 5.5**) following which the eluates were dried under a stream of nitrogen gas and reconstituted in 1 cm^3 of MeOH prior to analysis. The recovery efficiency of the method was assessed by spotting five sections of a paper and polymer banknote with 250 µL of the stock solution of cocaine HCl, applying the gel to each section and then isolating cocaine HCl from the gel matrix using SPE. Cotton wool swabs wetted with MeOH were used to sample the billfold wallets for cocaine.

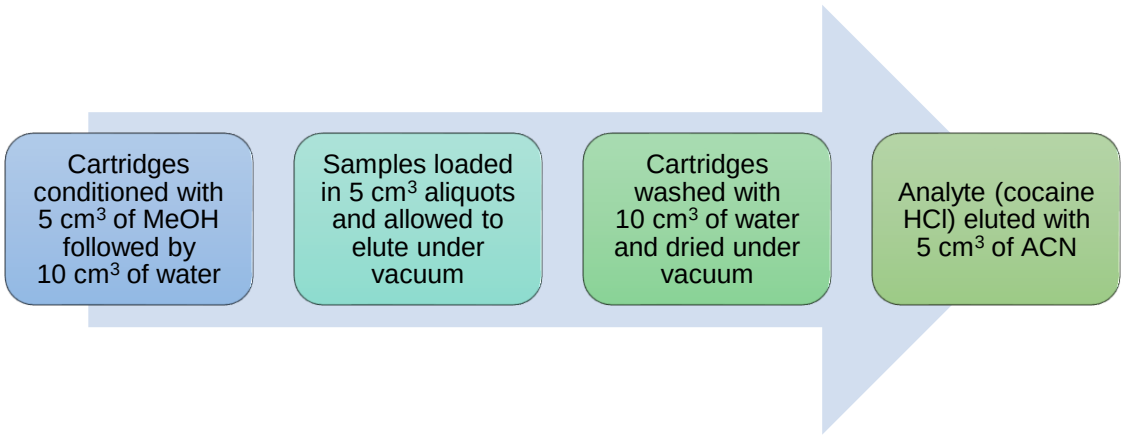


Figure 5.5. SPE method used to isolate cocaine HCl from the gelatine-based matrix.

5.2.4 Analysis and Quantification

All analyses were carried out using a Shimadzu Nexis GC-2030 gas chromatograph (GC) with an AOC-20i Plus auto injector and FID. The method used was adapted from the United Nations Office on Drugs and Crime (2012) (**Table 5.2**). Analysis of a standard solution of cocaine HCl yielded two resolved Gaussian peaks: a solvent peak at 1.7 mins and an analyte peak at 5.2 mins.

Table 5.2. GC operating conditions used for identification and quantification of cocaine HCl.

Detector	FID
Column	SH-Rxi™-5ms (30 m, 0.25 mm, 0.25 µm)
Carrier gas	Helium 1.14 mL/min
Injector temperature	280°C
Detector temperature	280°C
Oven temperature	250°C
Injection volume	2 µL
Split ratio	25:1
Run time	7 min

To quantify the amount of cocaine HCl present in each extract, a ten-point calibration curve was established (concentration range: 1 mg/L to 1,000 mg/L). Each of the calibration standards were injected onto the column in triplicate, and the average peak area calculated. This was then plotted against the concentration of each calibrator and the linearity evaluated (**Fig. 5.6**). The resultant coefficient of determination (R^2 = .99) indicated a linear relationship over the concentration range tested. Using the established line equation, y = 374.71(x) – 7142.87, the concentration of cocaine HCl in each extract was calculated by correlating its resultant peak area (y) to a concentration (x).

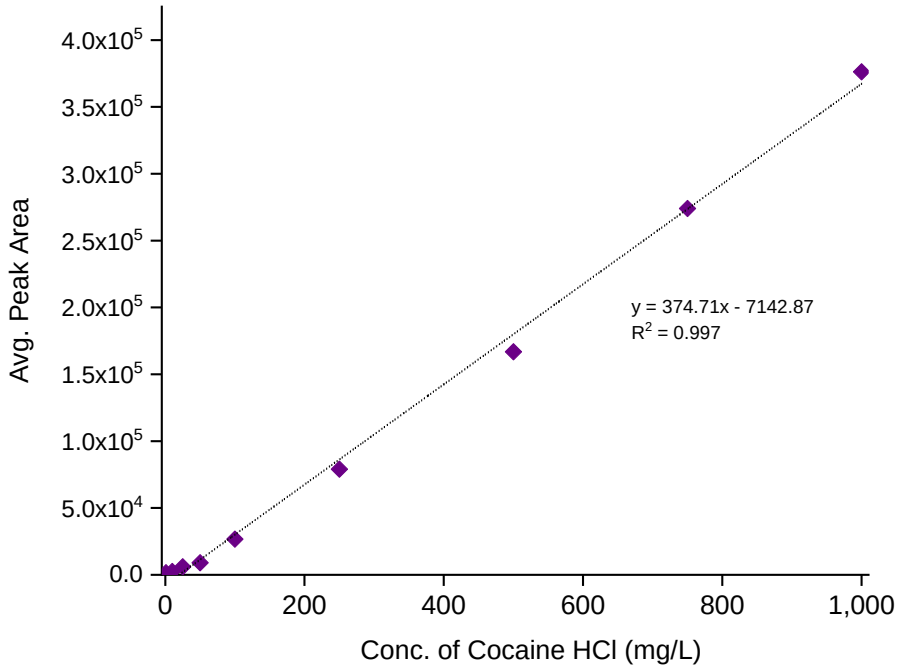


Figure 5.6. GC-FID calibration curve of cocaine HCl (reference standard).

5.2.5 Statistical Analyses

Data was analysed using IBM SPSS Statistics (version 26). The Shapiro-Wilk test was used to determine whether the data followed the Normal distribution or not. In all instances, the tests did not show evidence of non-normality ($p > 0.05$). Based on this, and the visual examination of the Q-Q plots, parametric tests were used. A one-way ANOVA was conducted to determine if the mean amount of cocaine HCl transferred depended on the duration of contact, force of contact, or extent of rotation applied. The strength of the relationship between the contact parameters (time, force, and rotation) and the amount of cocaine HCl transferred was then assessed using Pearson product-moment correlation coefficients. Based on the regression models, a two-sample *t*-test (assuming unequal variances) was carried out to compare the mean amount of cocaine HCl transferred when contact involved rotation, to those contacts that did not involve rotation. The impact of banknote type (paper and polymer) on the transfer of cocaine transferred was also assessed using a two-sample *t*-test (assuming unequal variances).

The relationship between the number of times a banknote was handled and the amount of cocaine present on the banknote was evaluated using the Pearson

product-moment correlation coefficient. A two-sample *t*-test (assuming unequal variance) was then carried out to compare the mean amount of cocaine recovered from a billfold wallet containing an adulterated polymer banknote to the recovery of cocaine from a wallet containing an adulterated paper banknote.

Cohen's *d*, the standardised mean difference, was used to measure the effect size (values >0.8 indicated a large effect size, where the results are likely to show a meaningful difference between the two groups (Cohen, 1988)). The means +/- 1 standard deviation, along with the coefficients of variation, are presented throughout. An alpha level of .05 (5%) was used for all statistical tests.

5.3 Results

5.3.1 Transfer Study

The recovery efficiency of the extraction technique employed to liberate cocaine HCl from the recipient surface (chamois leather) was determined to be 20.4 ± 0.9% (**Table 5.3**). Even though the percentage recovery was relatively low, the recovery was consistent (%CV = 4.2%). To provide a more accurate representation of the actual amount of cocaine HCl transferred, the measured amount of cocaine HCl in each sample was adjusted to account for the average percentage lost (79.6%) in the recovery process.

Table 5.3. Recovery results of cocaine HCl from the recipient surface. Units of concentration are mg/L.

Leather Sample	Conc. Spotted	Conc. Recovered	Recovery (%)	Avg. Recovery ± SD (%CV)
1	1,000.0	194.8	19.5	20.4 ± 0.9 % (4.2%)
2	1,000.0	204.7	20.5	
3	1,000.0	217.4	21.7	
4	1,000.0	206.5	20.7	
5	1,000.0	194.1	19.4	

Across the three parameters tested (the duration of contact, force of contact, and extent of rotation applied during contact), polymer banknotes consistently transferred more cocaine HCl to the skin proxy than paper banknotes. This finding was significant ($t(88) = 15.91$, $p < .001$, $d = 3.35$), and substantial, with approximately twice the amount of cocaine HCl transferred to skin from a polymer banknote than a

paper note. The amount of cocaine HCl transferred from paper banknotes did not depend on the duration or force of contact (time: R^2 = .19, F(1, 13) = 3.04, p = .105; force: R^2 = .08, F(1, 13) = 1.08, p = .318) With contacts occurring with polymer banknotes, larger contact forces resulted in a greater transfer of cocaine HCl, R^2 = .75, F(1, 13) = 39.25, p < .001. No trend was observed for the duration of contact, R^2 = .02, F(1, 13) = 0.25, p = .623 (see **Tables 5.4–5.5** and **Fig. 5.7**).

Table 5.4. The impact of time on the transfer of cocaine HCl from paper and polymer banknotes onto a human skin proxy.

	Paper Banknote		Polymer Banknote	
Time (s)	**%T**	**Avg. %T ± SD (%CV)**	**%T**	**Avg. %T ± SD (%CV)**
2	14.5 18.3 23.0 13.1 11.8	16.2 ± 4.6 (28.2%)	46.2 40.2 43.2 46.6 44.7	44.2 ± 2.6 (28.2%)
60	18.8 23.8 16.9 17.7 21.5	19.7 ± 2.9 (14.5%)	47.4 40.0 46.2 44.3 48.0	45.2 ± 3.2 (7.1%)
300	20.7 21.8 16.8 24.2 20.6	20.8 ± 2.7 (12.9%)	41.7 47.7 45.7 42.5 49.1	45.3 ± 3.2 (7.0%)

Table 5.5. The impact of force on the transfer of cocaine HCl from paper and polymer banknotes onto a human skin proxy.

	Paper Banknote		Polymer Banknote	
Force (N)	**%T**	**Avg. %T ± SD (%CV)**	**%T**	**Avg. %T ± SD (%CV)**
10	15.0 21.5 23.6 10.8 19.9	18.1 ± 5.2 (28.6%)	41.4 38.5 42.7 39.4 42.0	40.8 ± 1.8 (4.3%)
120	24.2 17.1 20.8 13.3 21.9	19.4 ± 4.3 (21.9%)	43.3 47.8 49.1 51.2 46.4	47.6 ± 3.0 (6.3%)
240	25.7 19.8 17.0 22.6 19.4	20.9 ± 3.4 (16.1%)	50.2 51.8 48.1 57.6 52.6	52.1 ± 3.5 (6.8%)

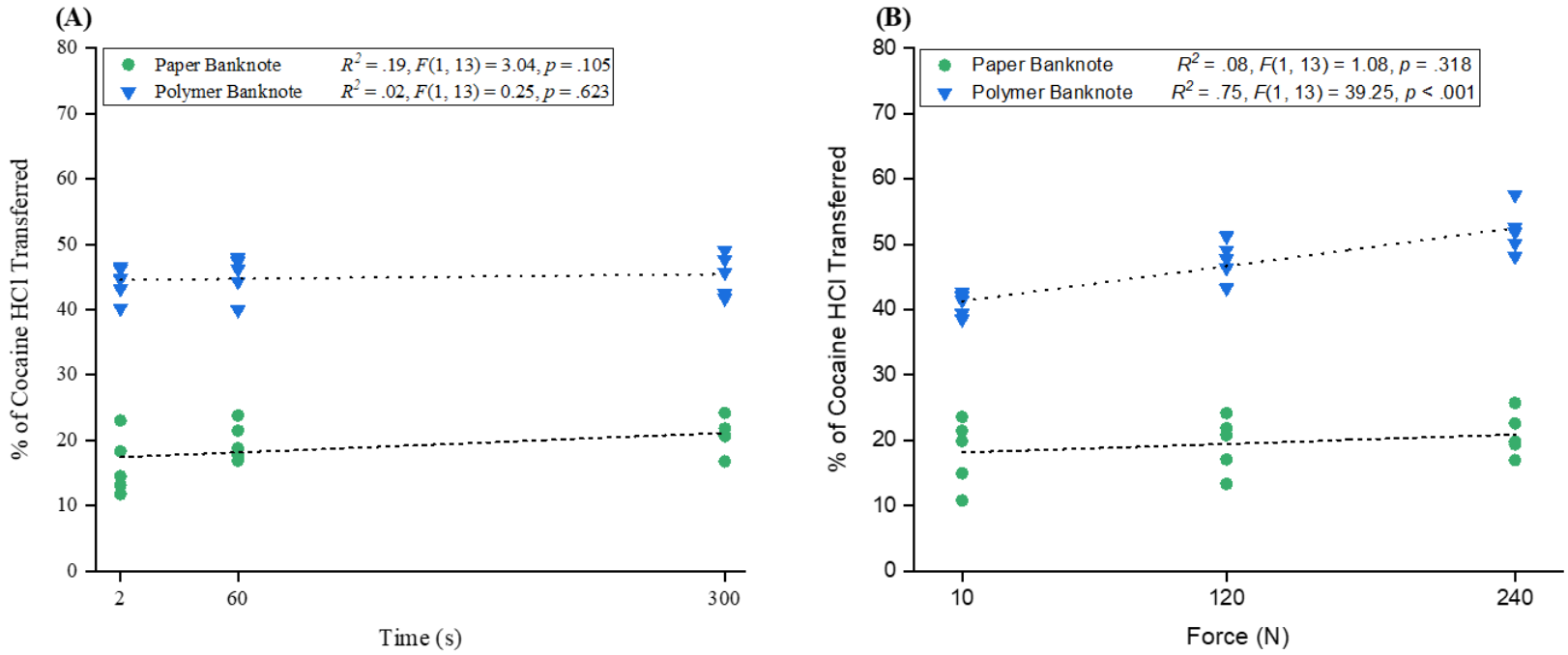


Figure 5.7. The percentage of cocaine HCl transferred from adulterated paper and polymer banknotes onto the recipient surface following contacts occurring at three different times (A) and forces (B).

For both polymer and paper banknotes, contact which involved rotation resulted in a larger transfer of cocaine HCl onto the recipient surface than those contacts not involving rotation (paper: $t(43) = 9.25$, $p < .001$, $d = 2.93$; polymer: $t(43) = 7.80$, $p < .001$, $d = 2.47$). With paper banknotes, increasing the amount of rotation applied over the contact period resulted in an increased transfer of cocaine HCl onto the recipient surface, $R^2 = .34$, $F(1, 13) = 6.71$, $p = .022$. No trend was observed for polymer banknotes, $R^2 = .02$, $F(1, 13) = 0.22$, $p = .649$. The results are summarised in **Table 5.6** and **Fig. 5.8**.

Table 5.6. The impact of rotation on the transfer of cocaine HCl from paper and polymer banknotes onto a human skin proxy.

	Paper Banknote		Polymer Banknote	
Rotation (°)	**%T**	**Avg. %T ± SD (%CV)**	**%T**	**Avg. %T ± SD (%CV)**
120	24.7 30.7 34.0 23.4 29.4	28.5 ± 4.3 (15.3%)	56.1 53.0 58.7 54.9 57.4	56.0 ± 2.2 (28.6%)
240	26.9 37.3 32.9 42.5 39.0	35.7 ± 6.0 (16.9%)	51.5 59.1 55.5 56.6 52.9	55.1 ± 3.0 (5.5%)
360	43.4 35.6 27.0 40.7 42.4	37.8 ± 6.8 (17.9%)	53.0 53.8 59.5 52.8 57.1	55.2 ± 3.0 (5.4%)

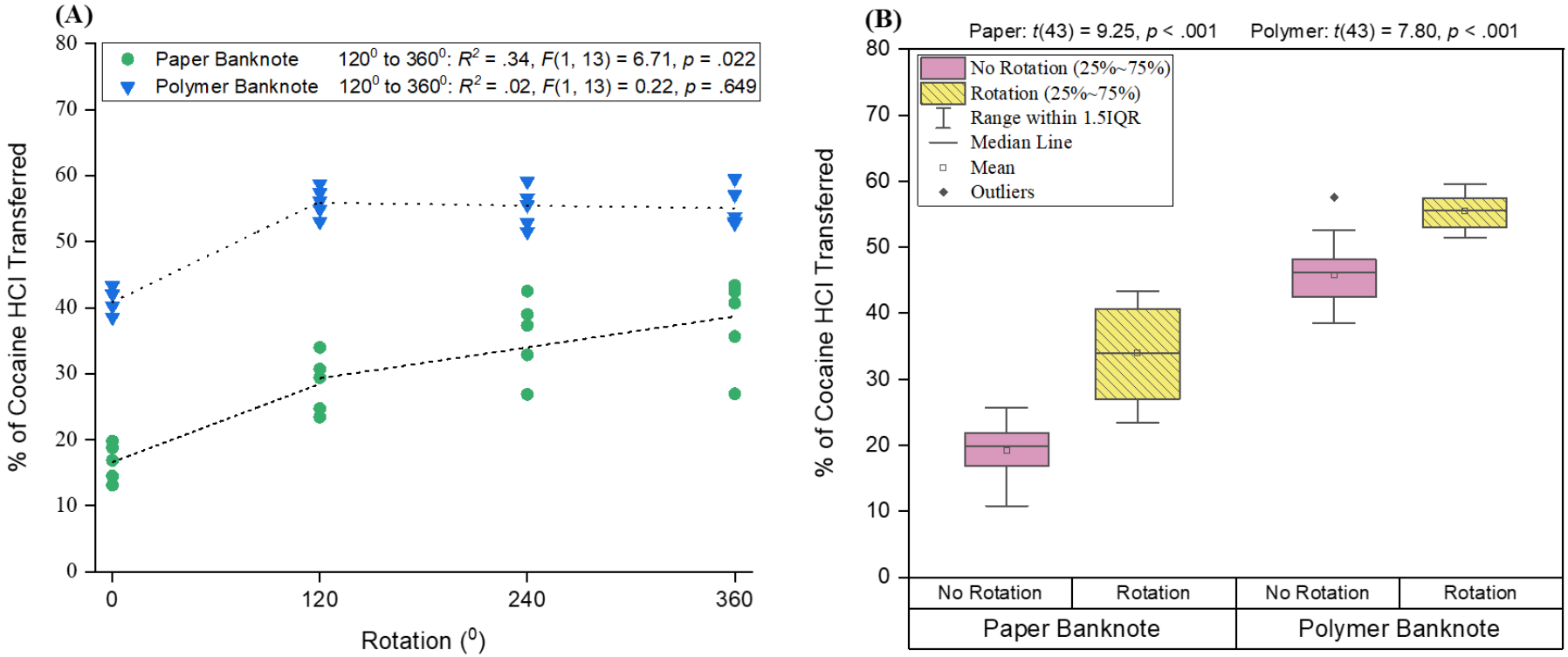


Figure 5.8. (A) The percentage of cocaine HCl transferred from adulterated paper and polymer banknotes onto the recipient surface following contacts occurring without rotation and with three rotations applied during contact. (B) Box plot comparison of the amount of cocaine HCl transferred from adulterated paper and polymer banknotes across contacts occurring without rotation and those with rotation.

5.3.2 Persistence Study

Using the gelatine-based collection medium, the recovery of cocaine HCl from banknotes was determined to be 9.5 ± 1.3% for paper banknotes and 38.9 ± 1.8% for polymer banknotes (**Table 5.7**). The recoveries of cocaine from paper banknotes yielded higher coefficients of variation (%CV = 13.6%) than those from polymer banknotes (%CV = 4.6%). As such, the percentage variability in the recovery of cocaine HCl was less from polymer banknotes. Following handling, the measured amount of cocaine present on each section of the paper and polymer banknotes was adjusted for, accounting for the average percentage recovery of the technique on both banknote types.

Table 5.7. Recovery results of cocaine HCl from the recipient surface. Units of concentration are mg/L.

Surface	Conc. Spotted	Conc. Recovered	Recovery (%)	Avg. Recovery ± SD (%CV)
Paper	1,000.0	77.2	7.7	9.5 ± 1.3 % (13.6%)
	1,000.0	104.7	10.5	
	1,000.0	83.1	8.3	
	1,000.0	111.4	11.1	
	1,000.0	99.8	10.0	
Polymer	1,000.0	368.2	36.8	38.9 ± 1.8 % (4.6%)
	1,000.0	393.7	39.4	
	1,000.0	418.3	41.8	
	1,000.0	372.9	37.3	
	1,000.0	391.4	39.1	

On average, 399.8 ± 30.3 mg/L of cocaine HCl remained on the surface of paper banknotes following handling. For polymer banknotes, the average was 158.0 ± 32.4 mg/L (**Table 5.8**). Greater quantities of cocaine were therefore persisting on paper banknotes after handling. This finding was significant, $t(48) = 27.25$, $p < .001$, $d = 7.71$. With increased handling, the concentration of cocaine HCl present on polymer banknotes decreased, $R^2 = .85$, $F(1, 23) = 131.12$, $p < .001$. No trend was observed for paper banknotes, $R^2 = .02$, $F(1, 23) = 0.36$, $p = .556$ (**Fig. 5.9**).

Table 5.8. The concentration of cocaine HCl on paper and polymer banknotes (as determined by the amount quantified in the eluates from the recovery process) following handling. Units of concentration are mg/L.

	Paper Banknote		Polymer Banknote	
Number of Times Handled	**Conc. of Cocaine HCl on Banknote**	**Avg. Conc. ± SD (%CV)**	**Conc. of Cocaine HCl on Banknote**	**Avg. Conc. ± SD (%CV)**
10	420.0 429.5 353.7 391.1 452.6	409.4 ± 38.1 (9.3%)	186.6 195.4 206.4 228.2 212.9	205.9 ± 16.0 (7.8%)
20	444.2 376.8 410.5 399.7 381.1	402.5 ± 27.1 (6.7%)	180.2 176.6 163.5 172.8 186.3	175.9 ± 8.5 (4.8%)
30	353.7 446.3 385.3 403.4 387.9	395.3 ± 33.7 (8.5%)	168.4 162.7 150.9 141.7 133.2	151.4 ± 14.5 (9.6%)
40	340.0 435.8 407.4 377.2 382.7	388.6 ± 35.7 (9.2%)	134.8 130.9 147.8 128.1 123.9	133.1 ± 9.1 (6.9%)
50	413.7 387.4 433.7 371.2 410.6	403.3 ± 24.3 (6.0%)	131.1 118.6 135.0 125.8 108.3	123.8 ± 10.6 (8.6%)

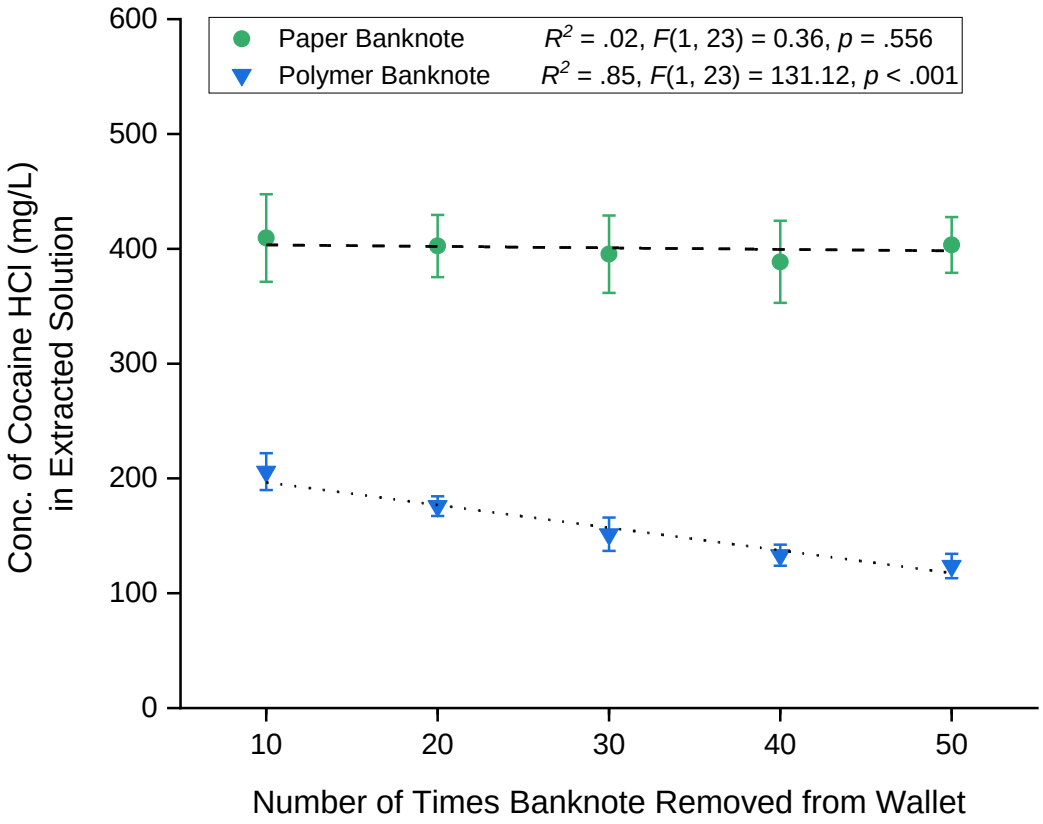


Figure 5.9. The average amount of cocaine HCl quantified in the extracts. Each banknote was sampled following handling a defined number of times. Error bars represent ± one standard deviation from the mean.

Following housing adulterated banknotes, each wallet was swabbed and the amount of cocaine HCl present quantified (the recovery efficiency of the swabbing method was 39.2%). The results obtained showed that swabs from wallets which housed adulterated paper banknotes had lower concentrations of cocaine HCl than those that held adulterated polymer banknotes (**Fig. 5.10**). This supports the observation that there was a larger transfer of cocaine HCl from polymer banknotes to the billfold wallet than from paper banknotes, $t(8) = 9.09$, $p < .001$, $d = 5.75$.

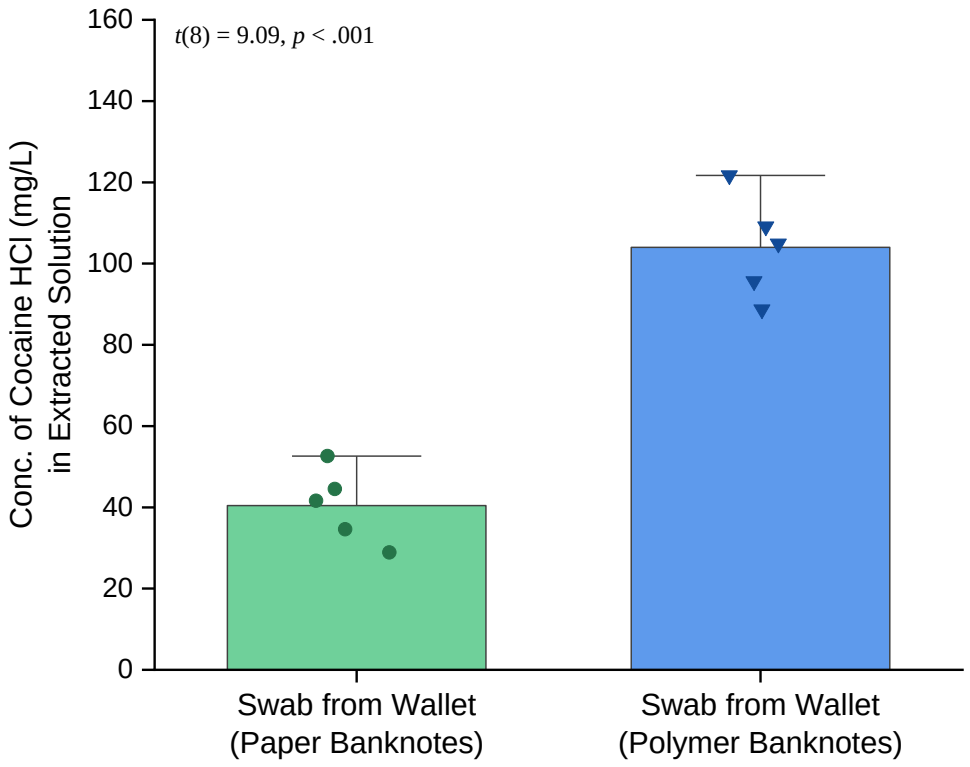


Figure 5.10. Comparison of the amount of cocaine HCl quantified in the extracts after sampling the billfold wallets that housed either adulterated paper or polymer banknotes. Data points represent the amount of cocaine HCl quantified on each swab after sampling the billfold wallets which housed each banknote type.

5.4 Discussion

Chamois leather as a material which could mimic the mechanical and frictional contact behaviour of skin (Dąbrowska *et al*., 2016; Fenton *et al*., 2020) provided a suitable proxy to assess the mechanics of transfer. However, the properties of human skin are complex and continuously changing due to different factors (Dąbrowska *et al*., 2016). As such, this needs to be taken into account when considering the results of this study and how they can be applied to situations of transfer of particles to human skin. The extraction method to recover cocaine from the recipient surface (preconditioned chamois leather) provided consistent results (%CV = 4.2%) and as such, offered insights for meaningful comparisons with respect to the transfer of cocaine (expressed as a percentage) from paper and polymer banknotes under different testing parameters. It was found that cocaine was readily transferred from adulterated paper and polymer banknotes onto a proxy for human skin across all testing parameters. A greater number of cocaine particulates were

transferred from polymer banknotes than from paper banknotes. This is likely due to the differences in the surface topography (as shown in the SEM micrographs (**Fig. 5.11**)). The results suggest that cocaine particulates are more likely to be retained within the porous matrix of paper banknotes while having a lower retention on non-porous polymer banknotes. This was an expected result and is in agreement with other studies that have assessed the impact of surface type on the transfer of trace forensic materials such as DNA (Goray *et al*., 2010a; Verdon *et al*., 2013).

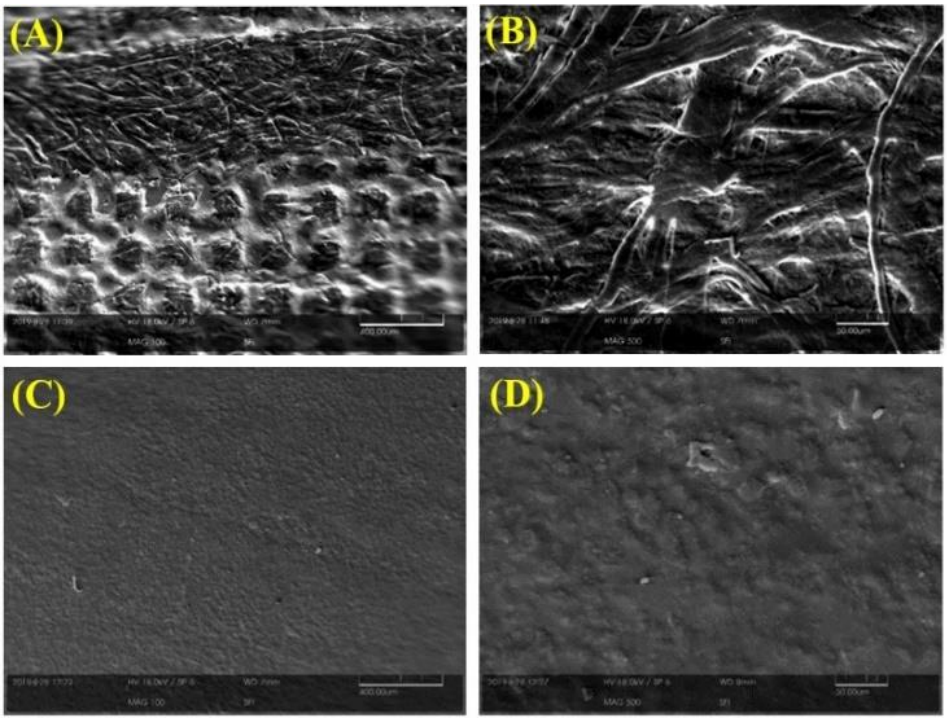


Figure 5.11. SEM micrographs. US one dollar paper banknote (A) 100x magnification and (B) 1,000x magnification. Vietnamese twenty thousand đồng polymer banknote (C) 100x magnification and (D) 1,000x magnification. Micrographs acquired using a Pemtron PS-230 scanning electron microscope fitted with an Oxford Instruments X-act energy-dispersive X-ray spectrometer system. Surfaces were sputter coated (time = 2 min; vacuum = 0.2 Torr; current = 0.2 mA) with gold prior to analysis.

The results indicate that the transfer of cocaine was independent of the duration of contact. A similar finding was noted by Amaral *et al*. (in press) with respect to the transfer of explosive residues. Conversely, contact time has been shown to have an impact on the transfer of fragrances (Gherghel *et al.*, 2019). As such, there is evidence to suggest that the transfer of volatile organic compounds increases with prolonged contact whilst the transfer of solid particulates is independent of contact

time. The impact of the type of evidence being transferred therefore needs to be considered as a factor and warrants further exploration.

Contacts which occurred at higher forces resulted in a larger transfer of cocaine from polymer banknotes whereas force did not impact the amount of transfer from paper banknotes. It is probable that higher contact forces are able to dislodge cocaine particulates from low porosity surfaces more readily than from higher porosity surfaces where there is a higher likelihood of the particulates being trapped within the pores of the surface. The force applied over a given surface area (pressure) has been reported to have an influence on the transfer of trace DNA (Goray *et al*., 2010a; Tobias *et al*., 2017) but not on the transfer of explosive residues (Amaral *et al.*, in press). Again, this highlights the complexity of evidence transfer and the need for further research to address evidence dynamics of relevant traces.

All contacts which involved rotation resulted in greater quantities of cocaine being transferred when compared to contact not involving rotation. Increasing the amount of rotation applied over the contact period resulted in a greater transfer from paper banknotes. This suggests that a larger proportion of cocaine particles are being liberated from the paper matrix of banknotes as the amount of rotation applied during contact increases. For polymer banknotes, only the application of rotation resulted in an increased transfer; the amount of rotation applied was not found to impact upon the amount transferred from polymer banknotes. Although there is limited published research on the impact of rotation on the transfer of forensic materials, Amaral *et al*. (in press) reported similar findings whereby contacts which involved rotation transferred more ammonium nitrate onto the recipient surface than contacts occurring without rotation.

A solution deposition was used to seed the banknotes with cocaine HCl in order to achieve accurate control over the concentration. Additionally, this form of deposition allowed for the percentage of cocaine transferred to be calculated from the concentration of cocaine HCl quantified in the extracts from the recipient surface. An important consideration is that the crystallisation of cocaine from the solvent will be likely to exhibit differences from the dry powdered form. The findings are therefore more relevant to damp environment conditions and in situations where adulterated

banknotes have taken on moisture and then subsequently dried. Building upon the contact parameters assessed in this reductionist approach, the interaction of cocaine particulates with the surface, such as the electrostatic forces involved, should be considered.

Using a gelatine-based sampling medium, the average recovery of cocaine from polymer banknotes (38.9 ± 1.8%) was comparable to the recovery results reported by Amaral *et al.* (2020), for explosive residues on ABS surfaces (38.6%). For paper banknotes, the average recovery (9.5 ± 1.3%) obtained was higher than previously reported recoveries from porous surfaces (wood: 1.1%, fabric: 1.6%, and carpet: 6.8%) (Amaral *et al.*, 2020). As this recovery method had previously only been used to sample explosive residues (Amaral *et al*., 2020) we expand upon the application of this technique as a viable method to also sample drug residues.

The trend in persistence was important to assess rather than the raw amount of cocaine HCl present on the banknote following handling. As such, the differences in the efficiencies of the recovery method for paper and polymer banknotes did not impact upon the ability to assess these trends. With successive handling, a continual decrease in the amount of cocaine present on polymer banknotes was observed. The observed trend was linear and departed from the two-stage model of decay curve first reported by Pounds and Smalldon (1975b), for fibre evidence and subsequently identified for other evidence types including glass (Hicks *et al.*, 1996), pollen (Bull *et al*., 2006), hair (Dachs *et al*., 2003), foam (Wiggins *et al.*, 2002), fragrances (Gherghel *et al.*, 2020), and diatoms (Scott *et al.*, 2021). However, these studies all assessed the persistence over time whereas this research assessed persistence following activity (the handling of banknotes). For paper banknotes, the amount of cocaine recovered remained relatively stable following handling. Therefore, cocaine is likely to persist on paper banknotes for longer periods of time following handling than for polymer banknotes. Following handling, the billfold wallet used to house each banknote was sampled for cocaine residues. A greater amount of cocaine was recovered from wallets that contained adulterated polymer banknotes than from those containing adulterated paper banknotes. This supports the finding that cocaine is more readily lost from polymer banknotes.

5.5 Conclusion

This study compared the transfer and persistence of cocaine on paper and polymer banknotes. The independent impact of three contact variables (time, force, and rotation) on the transfer of cocaine onto a human skin proxy was assessed. Automated control of the parameters in which contact between a given banknote and the recipient surface occurred was achieved using an Instron ElectroPuls E3000. To assess persistence, a realistic scenario was constructed to mimic an everyday cash transaction whereby cash was removed from, and returned to, a wallet. Each banknote was sampled after defined handling periods. The amount of cocaine transferred onto the recipient surface, as well as the amount of cocaine present on each banknote following handling, was quantified using GC-FID. The results from this study demonstrated that:

1. cocaine is readily transferred from both paper and polymer banknotes through contact;
2. a greater transfer of cocaine occurred from polymer banknotes;
3. contacts which involved rotation resulted in a larger transfer of cocaine than contacts not involving rotation;
4. cocaine persisted on both banknote types following extensive handling;
5. following handling, cocaine is more easily lost from polymer banknotes.

The results from this study provide empirical data that offer insights into the factors which underpin the transfer of cocaine from banknotes as well as persistence behaviour of cocaine on paper and polymer banknotes following handling. This study highlights that cocaine is readily transferred from banknotes through contact, even with the smallest forces and shortest contact times. Additionally, cocaine persists on all banknotes for extended periods of time following handling and is therefore available for transfer onto persons. Polymer banknotes were found to have a lower retention rate of cocaine particulates than paper banknotes. As such, cocaine particulates are more available for transfer from polymer banknotes and contact with adulterated polymer banknotes is therefore likely to result in larger quantities of cocaine being transferred onto individuals. Such findings are important for investigators when assessing the probability of innocent transfer of drug traces

acquired from contaminated surfaces, such as banknotes, as opposed to those transfers resulting from illicit activities. These are valuable data that address the likelihood, and likely quantities, of transfers in the course of handling banknotes. These insights have potential to assist with evaluating the relevance of illicit drug particles identified on items or persons of interest in crime reconstruction approaches.

Chapter 6

Summary

[T]race evidence can offer more than a possible/potential "source attribution." Trace materials can be used as exclusionary or associative evidence, and they can provide valuable information at the onset of an investigation to corroborate case activities and eyewitness accounts. The transfer and persistence of trace materials can provide useful information to reconstruct how a criminal event took place, expose relevant links, and reveal factors that can lead to the recovery of additional evidence.
(Trejos et al., 2020; p. 2)

Outline

A key issue to address in forensic science is the misinterpretation of forensic evidence. To ensure accurate interpretations are made, scientific bases to underpin the evidence dynamics of trace particulates are required. This need formed the foundation for the experimental work conducted in this thesis. Upon review of the findings, two key themes emerged with respect to the requirements for methodological approaches in forensic science research:

*(1) solutions need to be **context focused**;*

*(2) solutions need to be **implementable** in practice.*

This chapter synthesises the results from the three experimental studies and outlines a methodological framework for framing and supporting a research culture moving forward. Avenues for further research are also considered.

6.1 Framing a Research Culture Moving Forward

"[T]he traditional forensic sciences in general, and the pattern identification disciplines…do not currently possess – and absolutely must develop – a well-established scientific foundation" (Mnookin, *et al*., 2011*;* pp. 725–726). As such forensic science needs a research culture (Mnookin *et al*., 2011), one that establishes scientific bases to underpin the evidence dynamics of trace materials through structured empirical studies (Morgan *et al.*, 2020). However, over the last decade forensic science research has largely been fragmented and dichotomous.

“Research in forensic science is sorely needed, but it should address primarily forensic science questions – not questions relating to the application of chemistry, biology, statistics, or psychology” (Margot, 2011; p. 801). As such, to be successful, issues must be addressed with context in mind. This informed the research presented in this thesis, with each of the three experimental studies being context focused and adopting a novel methodology to provide implementable solutions to address the problems identified (**Table 6.1**).

Starting with controlled smaller scale studies (Chapters 3 and 4) and adapting these to larger realistic scenarios (Chapter 5), strengthens the pathway for addressing problems in forensic science. It ensures that the individual impact of variables on the transfer and persistence of forensic materials can be understood in a controlled experimental setting which can then be expanded upon through understanding the impact of multiple variables in realistic scenarios which are casework informed. This contrasts with how research in forensic science is generally conducted whereby a dichotomy has emerged in which experiments either mimic case reality or adopt a more controlled laboratory-based approach. Neither approach alone can provide all the answers. Thus, if we are to be successful in framing a research culture moving forward, both a reductionist and casework informed approach are required. This will ensure that the empirical evidence base we rely upon for interpretations is comprehensive and robust. Together, both approaches provide investigators with a holistic understanding of trace evidence dynamics.

Furthermore, we need to challenge the *status quo*; becoming complacent with standard practices and procedures does nothing to advance the field of forensic science and the issues raised by the NAS report in 2009 will continue to plague us. As such, “[c]ontinuing research in the forensic sciences is required to ensure processes do not stagnate but keep abreast of new technologies in related areas of research” (Linacre, 2013; p. 387). This thesis has shown that there are advantages in looking to other fields and disciplines in order to create new methods to answer forensic questions. By doing so, we can expand the tool kit which investigators can rely upon and further contribute to an evidence base upon which sound scientific conclusions can be made.

Table 6.1. Summary table of each of the experimental chapters outlining: (1) the problem addressed, (2) the context focused approach taken, (3) how the research is situated within the forensic science process and hierarchy of propositions, (4) the methodological approach adopted, and (5) the provision of implementable solutions.

	Problem	**Context Focused**	**Situating Research**	**Methodological Approach**	**Implementable Solutions**
Chapter 3	Particulate collection from porous surfaces is challenging, with low recoveries reported from these surfaces.	Designed to address limitations of conventional methods and offer an alternative method accessible to investigators.	'*Collection*' stage of the forensic science process; can support '*Source*' & '*Activity*' level propositions.	Created and evaluated a novel hydrogel formulation for the recovery of particulates from commonplace surfaces.	Offers investigators a cost-effective sampling medium comprised from readily available materials.
Chapter 4	The FSR has stressed the necessity to understand trace evidence dynamics to assist in investigators in answering how and when a given particulate transferred along with the conditions and timeframes they persist.	To support the interpretation of how a particulate arrived on a given surface, a consideration of the mechanics involved is important.	'*Division of matter & transfer*' stage of the forensic science process; can assist with '*Activity*' level propositions.	Adopted a reductionist approach and validated a new method to assess three contact variables under controlled and repeatable conditions.	Provides a new method to study the transfer of forensic materials; contributes to an evidence base upon which investigators can rely upon to interpret evidence.
Chapter 5		With more polymer banknotes entering circulation, the transfer of cocaine residues from these surfaces becomes important for evaluative interpretation.	Pertains to the first three stages of the forensic science process: *transfer*, *persistence*, and *collection*; addresses '*Activity*' level propositions.	Applied the methods created in the first two studies, expanding their application for other evidence types; assessed the impact of activity on persistence.	Supports investigators in assessing the probability of the innocent transfer of materials as opposed to transfers arising from illicit activities

6.2 Avenues for Further Research

To have meaningful impact, research must be context focused, implementable, and beneficial to the forensic community. Based on the results from the work conducted in this thesis, three areas of further research and exploration can be suggested (**Figure 6.1**).

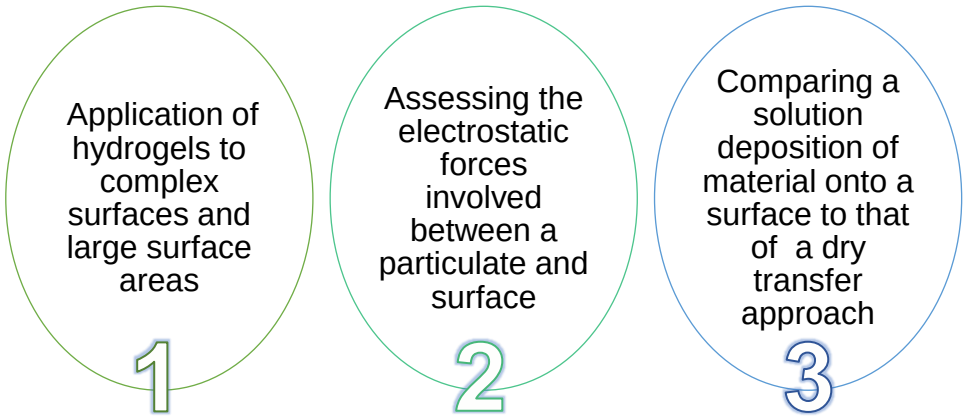


Figure 6.1. Three potential avenues for further research based on the findings of the experimental work conducted in this thesis.

Research into alterative collection techniques for forensic materials should directly address the limitations of conventional methods and offer solutions which are accessible to investigators. Additionally, as the timely collection of evidence at a crime scene is paramount, focus should be on a method which easy to apply, fast, and amenable to a wide range of surfaces and evidence types. Building upon the findings presented in Chapter 3, the application of hydrogels to complex surfaces, such as those which involve joints, bends, and twists, as well as vertical and inverted surfaces should be assessed. Additionally, the ability of hydrogels to sample large surface areas warrants exploration. Such studies are important in establishing the most appropriate method for recovering particulates from different surfaces and substrates encountered at a crime scene. Although this research presented a gelatine-based hydrogel, other formulations should also be explored and evaluated.

Understanding the transfer and persistence properties of forensic materials is important in assisting the court with answering ‘how’ and ‘when’ a given trace may have been deposited. The findings from Chapters 4 and 5 highlighted that the type of surfaces involved in transfer is an important consideration. As such, further research into the individual impact of surface type is warranted. Specifically, electrostatic forces are likely to play a role in the attraction and retention of trace particulates on surfaces. This is likely to influence the ability of that trace to transfer and persist and is therefore an important consideration when interpreting forensic evidence. As such, it would be prudent to assess the relative electrostatic forces involved to gain a better understanding of how particulates interact with the surfaces they come into contact with.

Finally, in the experiments conducted in this thesis, a solution deposition was chosen to ensure accurate control over the concentration and placement of the material being seeded onto the surface. One downside to this approach is that the material is first dissolved and subsequently recrystallised upon deposition which is likely to impact the surface topography and characteristics of the particulates. As such, future research should explore a dry transfer approach to deposit materials onto a surface. This exemplifies the need for both a reductionist and casework informed approach in fully understanding trace evidence dynamics.

Chapter 7

Conclusions

Physical evidence cannot be wrong; it cannot perjure itself; it cannot be wholly absent. Only in its interpretation can there be error. Only human failure to find, study and understand it can diminish its value (Kirk, 1974; p. 2)

Outline

This thesis focused on the recovery, transfer, and persistence of forensic materials, namely explosive and drug residues, across a range of porous and non-porous surfaces. Three experimental studies were conducted (Chapters 3–5), the findings of which support and contribute to a research culture in forensic science, one that is grounded in reality and provides solutions which are implementable in practice. This final chapter of the thesis summarises the key research findings and conclusions with respect to the aims presented in Chapter 1.

7.1 Research Aims and Conclusions

First Aim: *To address the poor recovery of forensic materials from porous surfaces by exploring the use of a novel hydrogel formulation*

A gelatine-based hydrogel formulation was created and evaluated as a sample collection technique for the recovery of explosive (Chapter 3) and drug (Chapter 5) residues from a variety of porous and non-porous surfaces. The final hydrogel formulation consisted of gelatine powder, arrowroot powder, glycerine, and water in a mass ratio of 3:1:1:5. For both residue types, higher recoveries were obtained from non-porous surfaces. On average, the recovery of AN across three non-porous surfaces (copper, ABS, and glass) was 27.0% whereas an average recovery of 3.2% was obtained across the three porous surfaces (carpet, fabric, and wood). Higher recoveries were obtained when collecting cocaine HCl residues from banknotes, with an average recovery of 38.9% from polymer (non-porous) banknotes and 9.5% from paper (porous) banknotes. A key attribute of the hydrogel was that it was able to recover both drug and explosive residues across all the surfaces assessed and

importantly, the four porous surfaces (carpet, paper, wood, and fabric), affording investigators a method which does rely on the conventional swabbing materials and solvent systems and can obtain comparable recoveries to those of traditional recovery methods.

Second Aim: *To expand upon an empirical evidence base through implementing a reductionist approach to understand the mechanics of how a particular form of trace may have been transferred*

A reductionist approach was adopted to understand the individual impact of contact force, duration of contact, and amount of rotation applied during contact, on the transfer of explosive (Chapter 4) and drug (Chapter 5) residues. Also assessed was the impact of surface type on the amount of material transferred. Using an Instron ElectroPuls E3000, a reliable and repeatable method was established, affording the automated control of the parameters in which the contact between two surfaces occurred. It was found that even with the smallest contact forces and shortest contact times, detectable amounts of drug and explosive residues were transferred onto a recipient surface. With respect to the impact of surface type, for both drug and explosive residues, a greater transfer of material was observed from contacts with adulterated non-porous surfaces (an average particulate transfer of 41.6% for AN and 49.1% for cocaine HCl) than from porous surfaces (an average particulate transfer of 15.1% for AN and 24.1% for cocaine HCl). A key finding of this research was that contacts which involved rotation, resulted in a greater transfer of material compared to contacts not involving rotation.

Third Aim: *To offer insights into how a certain activity can impact upon the persistence of a given trace*

A realistic scenario was constructed to mimic an everyday cash transaction whereby paper and polymer banknotes adulterated with a known concentration of cocaine HCl (1,000 mg/L) were removed from, and returned to, a billfold wallet (Chapter 5). Banknotes were sampled after defined handling periods (10, 20, 30, 40 and 50 times) and the amount of cocaine HCl on the banknote quantified. It was found that

cocaine HCl persisted for extended periods of time following handling on both paper and polymer banknotes (on average, 403.3 mg/L of cocaine HCl was recovered from paper banknotes and 123.8 mg/L from polymer banknotes after handling the notes fifty times). Polymer banknotes were found to have a lower retention rate of cocaine particulates than paper banknotes (an average loss of 82.1 mg/L of cocaine HCl was observed from initial to final sampling on polymer banknotes as opposed to a 6.1 mg/L loss from paper banknotes). As such, cocaine particulates are more available for transfer from polymer banknotes and contact with adulterated polymer banknotes is therefore likely to result in larger quantities of cocaine being transferred onto individuals.

7.2 Outcomes and Broader Implications

This research has shown the potential application of hydrogels to the collection and recovery of forensic materials. The work conducted presented an alternative recovery tool that affords researchers and investigators a cost-effective sampling medium that is comprised of readily available materials capable of recovering different forensic materials across a range of surfaces. As such, it provides a method which is easily accessible and implementable to both researchers and practitioners. It is also posited that hydrogels can offer other advantages over conventional methods, such as the long-term encapsulation of particulates during transfer and storage. The results of this research are therefore promising and, with further research, has the potential to offer a more effective method that investigators can rely upon when tasked with the forensic collection and recovery of trace particulates.

Also explored in this thesis is the adoption of a reductionist methodology to understand evidence dynamics. The findings show that this complimentary approach can bolster understanding and aid in crime reconstruction by providing an independent assessment of the variables involved when two surfaces come into contact. Additionally, a bottom-up approach is amenable to the creation of simulation models where specific parameters can be adjusted for a given case in which the transfer of materials has occurred. This can allow for more transparent and reproducible interpretations of trace materials in crime reconstructions moving forward. Furthermore, this work has highlighted the benefit of exploring new methods

and applications for instrumentation from other disciplines, such as Instron's ElectroPuls E3000, in forensic science research.

Additionally, this research has shown that adapting controlled smaller scale studies to larger realistic scenarios is a viable path forward – an approach which parallels evidence-based medicine. In this work the recovery, transfer, and persistence of cocaine on paper and polymer banknotes was assessed by implementing the hydrogel recovery method to recover particulate traces along with the ElectroPuls E3000 to assess the variables likely to impact transfer. The findings contribute to an evidence base which can be used to assess the probability of innocent transfer of drug traces acquired from contaminated surfaces, such as banknotes, as opposed to those transfers resulting from illicit activities. These are valuable data that address the likelihood, and likely quantities, of transfers in the course of handling everyday objects. These insights have potential to assist with evaluating the relevance of forensic materials identified on items or persons of interest in crime reconstruction approaches.

7.3 Final Thoughts

This thesis has demonstrated that there is value in both reductionistic and casework informed studies when compiling an evidence base that can address specific situations and broader challenges in forensic science. Such an empirical evidence base requires research which is context focused and mindful of its intended application setting. This speaks to the dominant conception that forensic science is considered to be a collection of sciences that can be applied to forensic questions rather than a fundamentally defined and distinctive discipline in its own right. We will only move forward when we take both attributes and integrate them into the research culture.

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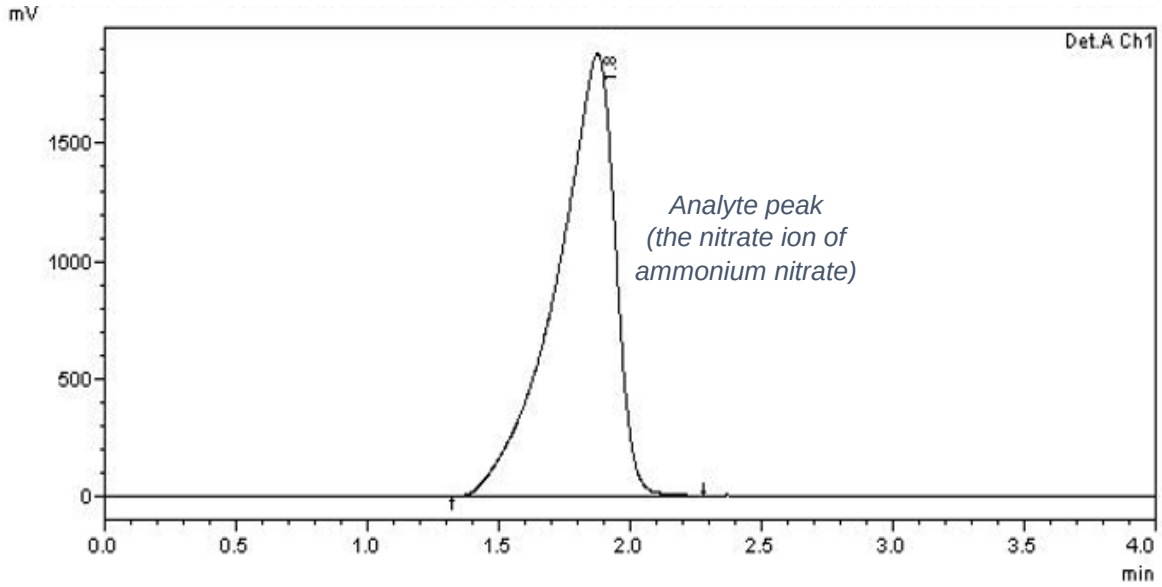
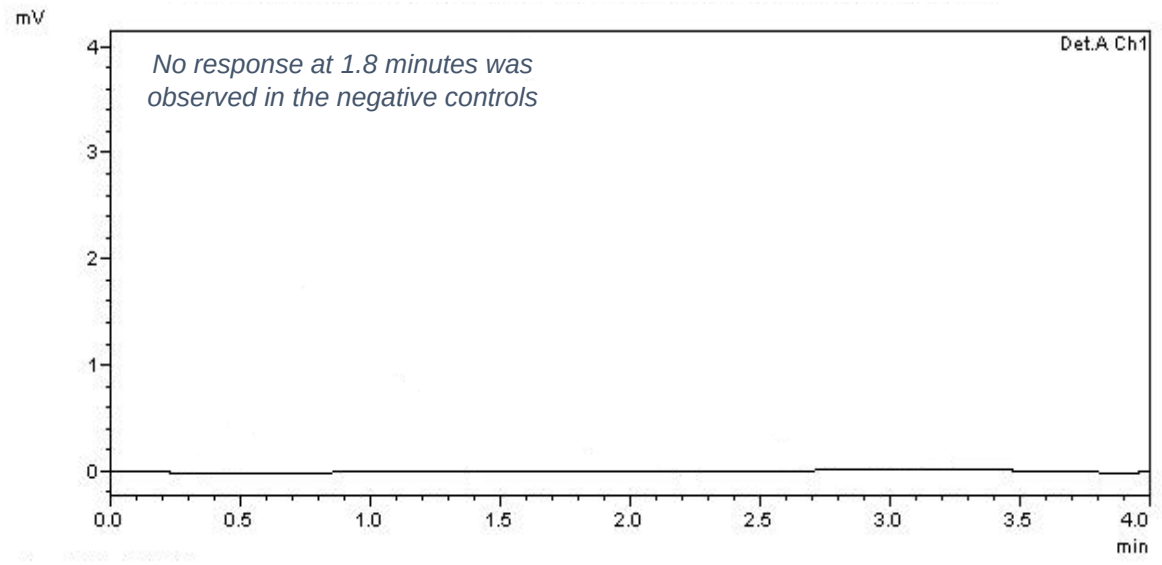
APPENDIX A

Data from LC-UV Analyses

Exemplar chromatogram showing the response of the instrumentation to a standard solution of ammonium nitrate (1,000 mg/L):



Exemplar chromatogram showing the response of the instrumentation to a surface wash extract of ABS (negative control):

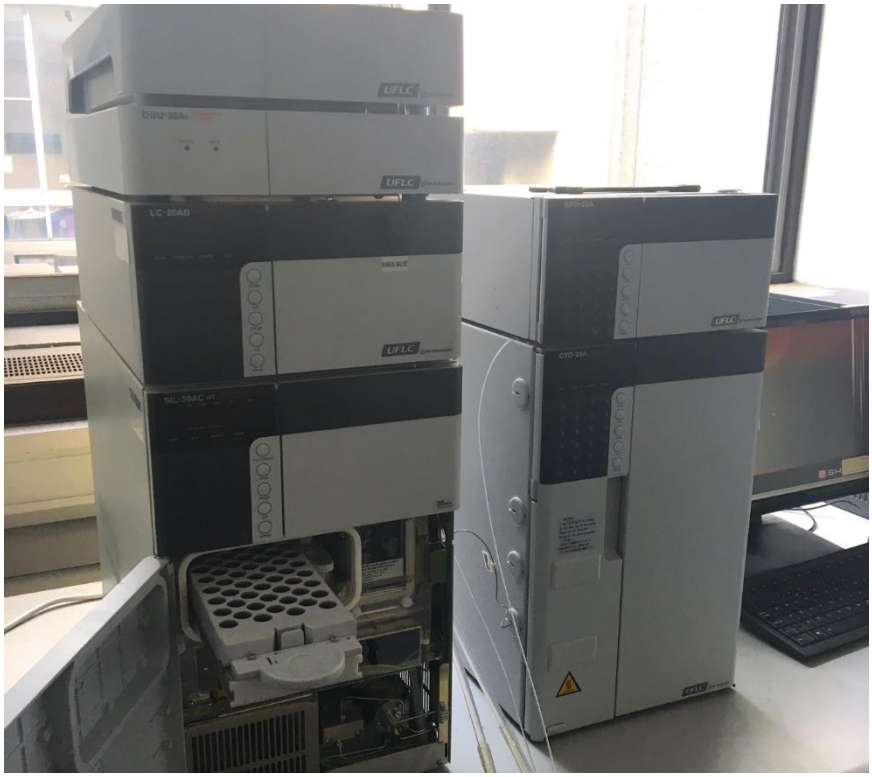


(Chromatograms acquired using a Prominence UFLC system from Shimadzu fitted with an Acclaim Explosive E2 column with UV detection)

Operating conditions:

Liquid Chromatograph	Shimadzu Prominence UFLC
Detector Column	UV-Vis (SPD-20A)
Column Flowrate	Acclaim Explosive E2 (3.0 x 150 mm, 3 µm, 120 Å)
Flowrate	0.3250 mL min^{-1}
Mobile Phase	48:52 (v/v) of MeOH/water
Gradient	Isocratic
Injection Volume	2.0 µL
Temperature	30°C
Detection	214 nm

Shimadzu Prominence UFLC system with an SPD-20A UV-Vis detector:



(1) Calibration curve:

Calibrator	RT (min)	Peak Area	Peak Area ($x10^4$)
1 mg/L	1.789	28673	2.87
	1.779	42241	4.22
	1.782	46071	4.61
5 mg/L	1.786	178757	17.88
	1.792	196237	19.62
	1.788	221946	22.19
10 mg/L	1.794	361790	36.18
	1.786	287474	28.75
	1.791	332551	33.26
25 mg/L	1.789	710786	71.08
	1.797	643601	64.36
	1.784	684063	68.41
50 mg/L	1.795	1412502	141.25
	1.797	1378247	137.82
	1.792	1298636	129.86
100 mg/L	1.789	2686644	268.66
	1.791	2617380	261.74
	1.798	2688284	268.83
250 mg/L	1.786	8193946	819.39
	1.793	8127226	812.72
	1.799	8135839	813.58
500 mg/L	1.803	14907667	1490.77
	1.798	14796135	1479.61
	1.801	14886434	1488.64
1,000 mg/L	1.804	29283626	2928.36
	1.797	29108884	2910.89
	1.803	29410914	2941.09

(2) Independently prepared calibrator checks:

Calibrator Check	RT (min)	Peak Area	Peak Area ($x10^4$)
250 mg/L	1.787	7437271	743.73
	1.799	7338447	733.84
	1.792	7328741	732.87
850 mg/L	1.801	25045941	2504.59
	1.799	24806234	2480.62
	1.798	24704174	2470.42

(3) Negative controls (unadulterated surfaces):

Negative Control	Response at RT = 1.8 min	Peak Area	Peak Area ($x10^4$)
Wood	ND	-	-
Fabric	ND	-	-
Carpet	ND	-	-
Copper	ND	-	-
Glass	ND	-	-
ABS	ND	-	-

ND = 'not detected' (no measurable response recorded between 1.3 and 2.3 min)

(4) Negative controls (gelatine-based hydrogel formulation):

Negative Control	Response at RT = 1.8 min	Peak Area	Peak Area ($x10^4$)
Gelatine	ND	-	-
Starch	ND	-	-
Glycerine	ND	-	-
Water	ND	-	-
Bromelain	ND	-	-

ND = 'not detected' (no measurable response recorded between 1.3 and 2.3 min)

(5) Gelatine-based hydrogel recoveries from porous surfaces:

Surface	RT (min)	Peak Area	Peak Area ($x10^4$)
Wood	1.776	448980	44.90
	1.781	228392	22.84
	1.779	719577	71.96
	1.780	104861	10.49
	1.783	346038	34.60
Fabric	1.778	381334	38.13
	1.782	198978	19.90
	1.784	946042	94.60
	1.779	651922	65.19
	1.780	454861	45.49
Carpet	1.777	163683	16.37
	1.779	725452	72.55
	1.780	1404876	140.49
	1.778	4443129	444.31
	1.781	3493122	349.31

(6) Gelatine-based hydrogel recoveries from non-porous surfaces:

Surface	RT (min)	Peak Area	Peak Area ($x10^4$)
Copper	1.784	4960781	496.08
	1.781	3001944	300.19
	1.779	6440206	644.02
	1.782	4972545	497.25
	1.787	7957863	795.79
Glass	1.783	3031359	303.14
	1.780	10631414	1063.14
	1.778	5372548	537.25
	1.785	9460816	946.08
	1.776	7184328	718.43
ABS	1.791	13552026	1355.20
	1.788	4860780	486.08
	1.786	15537336	1553.73
	1.794	18169710	1816.97
	1.782	4872544	487.25

(1) Calibration curve:

Calibrator	RT (min)	Peak Area	Peak Area ($x10^4$)
1 mg/L	1.769	28676	2.87
	1.771	42191	4.22
	1.768	35892	3.59
5 mg/L	1.770	168833	16.88
	1.773	236238	23.62
	1.769	221888	22.19
10 mg/L	1.774	361753	36.18
	1.771	287459	28.75
	1.767	308566	30.86
25 mg/L	1.775	710778	71.08
	1.770	643643	64.36
	1.772	683811	68.38
50 mg/L	1.777	1412463	141.25
	1.773	1378194	137.82
	1.775	1298629	129.86
75 mg/L	1.776	2185175	218.52
	1.781	2114658	211.47
	1.779	2266627	226.66
100 mg/L	1.777	2696634	269.66
	1.783	2567422	256.74
	1.780	2718279	271.83
250 mg/L	1.782	8213945	821.39
	1.778	8187212	818.72
	1.785	8135754	813.58
500 mg/L	1.791	14907744	1490.77
	1.789	14596091	1459.61
	1.793	15366404	1536.64
1,000 mg/L	1.798	29293567	2929.36
	1.803	29075874	2907.59
	1.801	29280908	2928.09

(2) Independently prepared calibrator checks:

Calibrator Check	RT (min)	Peak Area	Peak Area ($x10^4$)
350 mg/L	1.777	10319549	1031.95
	1.780	10168382	1016.84
	1.782	10270435	1027.04
800 mg/L	1.789	23715510	2371.55
	1.791	23430821	2343.08
	1.794	23311123	2331.11

(3) Negative controls (solvent washes of unadulterated surfaces):

Negative Control	Response at RT = 1.8 min	Peak Area	Peak Area ($x10^4$)
MDF	ND	-	-
Stainless Steel	ND	-	-
Chamois Leather	ND	-	-

ND = 'not detected' (no measurable response recorded between 1.3 and 2.3 min)

(4) Negative controls (solvents):

Negative Control	Response at RT = 1.8 min	Peak Area	Peak Area ($x10^4$)
MeOH	ND	-	-
Water	ND	-	-

ND = 'not detected' (no measurable response recorded between 1.3 and 2.3 min)

(5) Chamois leather recoveries:

Recovery Test Sample #	RT (min)	Peak Area	Peak Area ($x10^4$)
1	1.782	7719117	771.91
2	1.789	7530893	753.09
3	1.793	7869108	786.91
4	1.779	7954397	795.44
5	1.785	7613241	761.32

(6) Recoveries from recipient surface following contact with adulterated porous surfaces (time):

Time	RT (min)	Peak Area	Peak Area ($x10^4$)
2 s	1.771	2588733	258.87
	1.776	1404891	140.49
	1.782	2639469	263.95
60 s	1.777	2081372	208.14
	1.781	2639469	263.95
	1.780	1320331	132.03
150 s	1.775	1844604	184.46
	1.778	2081372	208.14
	1.783	1810780	181.08
300 s	1.781	1523276	152.33
	1.779	2554909	255.49
	1.784	1320331	132.03

(7) Recoveries from recipient surface following contact with adulterated non-porous surfaces (time):

Time	RT (min)	Peak Area	Peak Area ($x10^4$)
2 s	1.789	5227008	522.70
	1.783	5108624	510.86
	1.786	5666720	566.67
60 s	1.781	5869664	586.97
	1.779	5869664	586.97
	1.784	6529233	652.92
150 s	1.780	5785104	578.51
	1.782	6579969	658.00
	1.777	7290274	729.03
300 s	1.778	5328480	532.85
	1.785	5091711	509.17
	1.780	5260832	526.08

(8) Recoveries from recipient surface following contact with adulterated porous surfaces (force):

Force	RT (min)	Peak Area	Peak Area ($x10^4$)
10 N	1.773	1946076	194.61
	1.781	1827692	182.77
	1.777	2250492	225.05
60 N	1.775	2402701	240.27
	1.784	2487261	248.73
	1.780	1354155	135.42
120 N	1.782	2402701	240.27
	1.773	1962988	196.30
	1.777	1996812	199.68
240 N	1.774	1692396	169.24
	1.778	1878428	187.84
	1.770	1793868	179.39

(9) Recoveries from recipient surface following contact with adulterated non-porous surfaces (force):

Force	RT (min)	Peak Area	Peak Area ($x10^4$)
10 N	1.785	6174081	617.41
	1.781	6140257	614.03
	1.789	6613793	661.38
60 N	1.779	6579969	658.00
	1.784	6343201	634.32
	1.780	6275553	627.56
120 N	1.787	7341010	734.10
	1.779	7053506	705.35
	1.784	6664529	666.45
240 N	1.781	6326289	632.63
	1.788	7256450	725.64
	1.783	6512321	651.23

(10) Recoveries from recipient surface following contact with adulterated porous surfaces (rotation):

Rotation	RT (min)	Peak Area	Peak Area ($x10^4$)
90°	1.783	3366686	336.67
	1.775	3214477	321.45
	1.780	4161550	416.16
180°	1.777	3332862	333.29
	1.781	3603454	360.35
	1.776	4246111	424.61
270°	1.782	3518894	351.89
	1.780	4195375	419.54
	1.778	3823310	382.33
360°	1.781	3671102	367.11
	1.783	4415231	441.52
	1.779	4060078	406.01

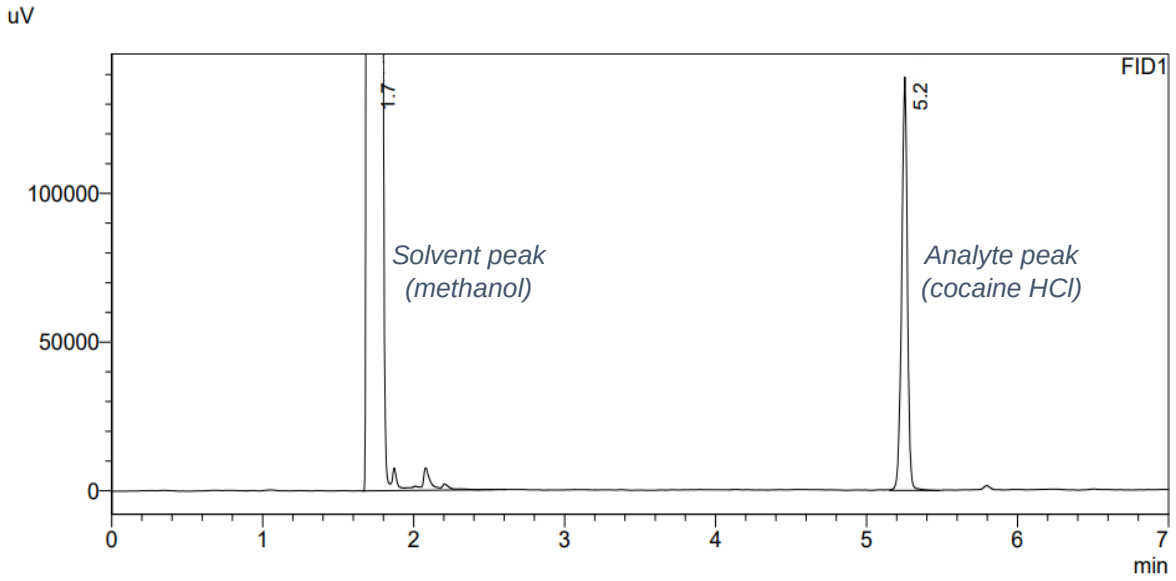
(11) Recoveries from recipient surface following contact with adulterated non-porous surfaces (rotation):

Rotation	RT (min)	Peak Area	Peak Area ($x10^4$)
90°	1.791	8237347	823.73
	1.786	9099860	909.99
	1.793	8389555	838.96
180°	1.795	8507939	850.79
	1.790	8406467	840.65
	1.788	9049124	904.91
270°	1.792	9624132	962.41
	1.796	8186611	818.66
	1.787	8947652	894.77
360°	1.789	8660147	866.01
	1.794	9471924	947.19
	1.792	8795444	879.54

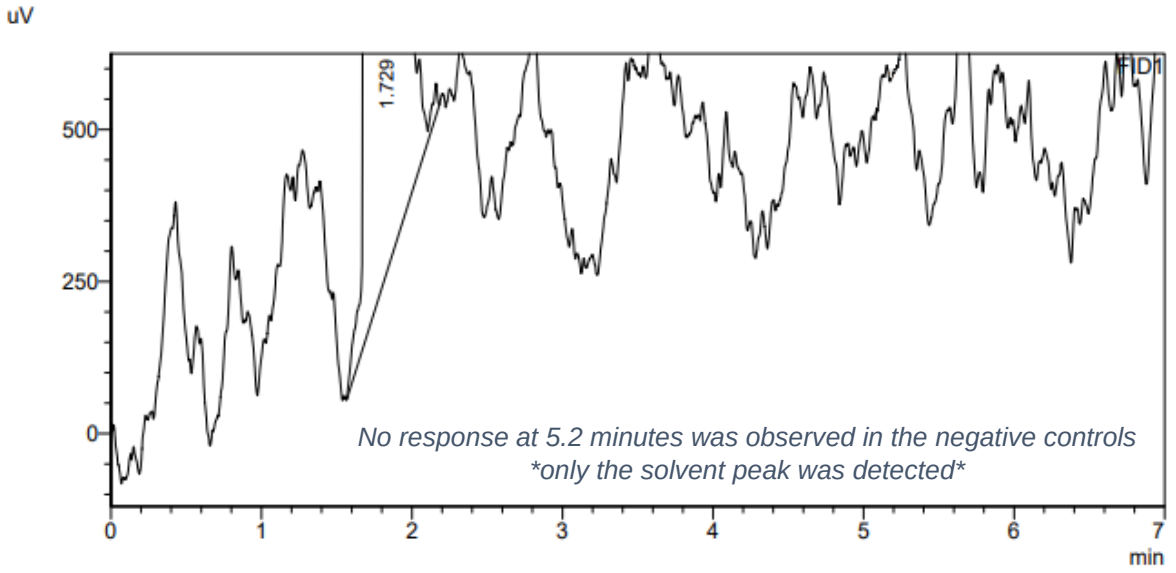
APPENDIX B

Data from GC-FID Analyses

Exemplar chromatogram showing the response of the instrumentation to a standard solution of cocaine HCl (1,000 mg/L):



Exemplar chromatogram showing the response of the instrumentation to a surface wash extract of the chamois leather (negative control):



(Chromatograms acquired using a Shimadzu Nexis GC-2030 with a FID)

Operating conditions:

Detector	FID
Column	SH-Rxi™-5ms (30 m, 0.25 mm, 0.25 µm)
Carrier gas	Helium 1.14 mL/min
Injector temperature	280°C
Detector temperature	280°C
Oven temperature	250°C
Injection volume	2 µL
Split ratio	25:1
Run time	7 min

Shimadzu Nexis GC-2030 with AOC-20i auto injector:

(1) Calibration curve:

Calibrator	RT (min)	Peak Area
1 mg/L	5.211	1297
	5.215	2192
	5.221	1296
10 mg/L	5.228	3097
	5.222	2736
	5.229	1651
25 mg/L	5.220	6182
	5.231	5509
	5.234	6446
50 mg/L	5.243	8764
	5.230	9871
	5.220	8556
100 mg/L	5.249	26078
	5.241	27902
	5.227	26240
250 mg/L	5.219	80004
	5.217	76054
	5.219	81029
500 mg/L	5.233	167635
	5.230	165772
	5.240	167035
750 mg/L	5.221	255390
	5.227	307108
	5.249	259771
1,000 mg/L	5.239	365845
	5.244	397534
	5.237	365574

(2) Negative controls (solvent washes of unadulterated banknotes following cleaning with isopropyl alcohol):

Negative Control	Response at RT = 5.2 min	Peak Area
Paper 1	ND	-
Paper 2	ND	-
Polymer 1	ND	-
Polymer 2	ND	-

ND = 'not detected' (no measurable response recorded between 4.2 and 6.2 min)

(3) Chamois leather recoveries:

Recovery Test Sample #	RT (min)	Peak Area
1	5.235	65851
2	5.212	69560
3	5.215	74319
4	5.236	70235
5	5.243	65588

(4) Recoveries from recipient surface following contact with adulterated paper and polymer banknotes (time):

	Paper Banknotes		Polymer Banknotes	
Time	RT (min)	Peak Area	RT (min)	Peak Area
2 s	5.214	23150	5.248	89321
	5.224	31127	5.225	76741
	5.243	40883	5.210	82980
	5.226	20287	5.243	90080
	5.223	17423	5.222	86150
60 s	5.218	32048	5.214	91673
	5.241	42505	5.236	76230
	5.236	28088	5.220	89218
	5.224	29798	5.235	85332
	5.223	37673	5.238	92900
300 s	5.217	36139	5.214	79911
	5.247	38340	5.239	92286
	5.241	27855	5.248	88298
	5.220	43347	5.213	81548
	5.240	35934	5.229	95252

(5) Recoveries from recipient surface following contact with adulterated paper and polymer banknotes (force):

	Paper Banknotes		Polymer Banknotes	
Force	**RT (min)**	**Peak Area**	**RT (min)**	**Peak Area**
10 N	5.237	24071	5.212	79298
	5.216	37673	5.234	73264
	5.224	42060	5.221	81957
	5.210	15378	5.219	75105
	5.239	34400	5.236	80525
120 N	5.217	43261	5.223	83184
	5.236	28468	5.240	92491
	5.225	36241	5.235	95283
	5.237	20696	5.222	99752
	5.222	38491	5.217	89627
240 N	5.222	46511	5.215	97502
	5.226	34196	5.208	100980
	5.215	28264	5.229	93309
	5.234	40025	5.230	112945
	5.218	33275	5.228	102616

(6) Recoveries from recipient surface following contact with adulterated paper and polymer banknotes (rotation):

	Paper Banknotes		Polymer Banknotes	
Rotation	**RT (min)**	**Peak Area**	**RT (min)**	**Peak Area**
120°	5.225	44423	5.204	109980
	5.229	56900	5.219	103434
	5.206	63752	5.208	115400
	5.234	41764	5.227	107423
	5.206	54241	5.203	112639
240°	5.222	48923	5.211	100264
	5.219	70707	5.207	116218
	5.218	61400	5.223	108752
	5.235	81548	5.232	110900
	5.204	74184	5.217	103127
360°	5.205	83389	5.219	103434
	5.232	67230	5.209	105070
	5.220	49127	5.218	117036
	5.211	77772	5.221	102923
	5.223	81364	5.218	112025

(7) Gelatine-based hydrogel recoveries:

	Paper Banknotes		Polymer Banknotes	
Recovery Test Sample #	**RT (min)**	**Peak Area**	**RT (min)**	**Peak Area**
1	5.234	21785	5.211	130825
2	5.210	32089	5.243	140380
3	5.227	23996	5.211	149598
4	5.213	34600	5.226	132586
5	5.219	30253	5.210	139519

(8) Recoveries of cocaine HCl from paper and polymer banknotes following handling:s

	Paper Banknotes		Polymer Banknotes	
Number of times handled	**RT (min)**	**Peak Area**	**RT (min)**	**Peak Area**
10	5.230	75470	5.229	36259
	5.214	77339	5.238	38306
	5.241	62429	5.243	40865
	5.221	69786	5.234	45935
	5.225	81883	5.218	42377
20	5.234	80230	5.221	34771
	5.232	66973	5.214	33933
	5.238	73602	5.224	30886
	5.241	71477	5.228	33049
	5.219	67819	5.223	36190
30	5.237	62429	5.207	32026
	5.225	80644	5.233	30700
	5.227	68645	5.235	27956
	5.243	72205	5.228	25816
	5.214	69156	5.236	23839
40	5.241	59735	5.237	24211
	5.223	78578	5.234	23304
	5.242	72992	5.243	27235
	5.210	67052	5.237	22653
	5.206	68134	5.229	21676
50	5.236	74231	5.234	23350
	5.216	69058	5.230	20443
	5.217	78165	5.225	24257
	5.229	65871	5.240	22118
	5.217	73621	5.214	18047

(9) Recoveries of cocaine HCl from billfold wallets after housing adulterated banknotes:

	Sample	RT (min)	Peak Area
Billfold wallets housing adulterated paper notes	1	5.220	12567
	2	5.243	5822
	3	5.236	9532
	4	5.226	3686
	5	5.224	8445
Billfold wallets housing adulterated polymer notes	1	5.244	33738
	2	5.233	28679
	3	5.209	38459
	4	5.212	26094
	5	5.235	32164