

Submitted to the Division of Postgraduate Research of Kingston University,

London

An Investigation into inorganic gunshot residue (IGSR) using Scanning Electron Microscopy (SEM) and Energy

Dispersive X-Ray (EDX)

This thesis is submitted in partial fulfilment of the requirements of Kingston University

for the degree of:

Masters by research in Forensic and Archaeological Sciences

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July 2021

Table of Contents

Acknowledgements	4
Abstract	5
Keywords	6
Chapter 1: Introduction	7
1.1 Firearms	8
1.2 Firearm Mechanisms	9
1.3 Gunshot Residue Background	11
1.4 Organic gunshot residue and Inorganic gunshot residue	12
1.5 GSR in the Environment	14
1.6 Deposition	16
1.7 Transfer and Prevalence	20
1.8 Collection and Analysis Techniques	26
1.9 Scanning Electron Microscope and Energy Dispersive X-Ray Spectrometry	27
1.10 Aims and hypotheses	31
Chapter 2: Materials and Methods	33
2.1 Instruments and laboratory equipment	34
2.2 Study One: the transfer of GSR particles	
2.2.1 Sample preparation and collection	35
2.3 Study Two: differing ammunition types	
2.3.1 Sample preparation and collection	
2.4 Study Three: distance and dispersion	40
2.4.1 Sample preparation and collection	40
2.5 Analysis	41
Chapter 3: Results and Discussion	44

3.1 Particle distribution; study 145
3.1.1 GSR particle distribution, number and type; study 148
3.1.2 Study one discussion50
3.2 Particle analysis; study 255
3.2.1 GSR particle analysis, number and type; study 256
3.2.2 Three component GSR particle sizing; study 261
3.2.3 Study two discussion62
3.3 Particle analysis; study 366
3.3.1 GSR particle analysis, number and type; study 368
3.3.2 Three component GSR particle sizing; study 372
3.3.3 Study three discussion77
Chapter 4: Conclusions and Future Research
4.1 Conclusions
4.2 Future Research
References
Appendix98

Acknowledgements

I would like to firstly thank my family and friends for their help and support throughout this writing process and the chaos of the last year, I wouldn't have got this far without any of you.

I'd also like to thank my supervisors Dr Robert McHardy and Dr Brian Rooney, for

their knowledge and guidance throughout the last several months.

Lastly, I would like to thank Richard Giddens for his help in the lab and all the advice

he gave.

I am so grateful for everyone that's helped me on this journey.

Abstract

Gunshot residue (GSR) is the debris that is expressed from a firearm when it has been discharged. A series of experiments were carried out following a range of shooting and transfer studies using SEM/EDX to detect inorganic gunshot residue. The initial investigation focused on the transfer of GSR particles on clothing from the firer of a Ruger semi-automatic rifle, using CCI standard velocity .22" long rifle round nose cartridges. Following discharge of the firearm, person to person contact was initiated with a non-firearm user not present in the vicinity at the time of the shooting. This resulted in the transfer of GSR from the shooter to the test subject, but with a reduced likelihood of three component particles (barium, antimony, lead) being detected. The total number of particles consistent with GSR transferred ranged from 112 to 302. Out of those particles, only 2 to 14 were three component GSR particles.

Further experiments carried out at the ballistics and forensics investigation unit at An Garda Siochana, investigated the composition of GSR in a firearm that discharged combinations of Wolf and Fiocchi 9 mm ammunition. From the particle compositions found it was possible to identify which ammunition had been fired last due to the percentage of SbBa particles present.

The distance GSR travelled from several different calibre firearms was assessed by examining targets over ranges of 2-10 metres. These experiments suggest that particles consistent with GSR can travel up to 10 metres from the firearm but characteristic three component particles only travelled 10 metres from a 12-gauge shotgun. Conversely, most three-component GSR particles from the other firearms only travelled up to 3-4 metres. All ammunition types had at least one three-component particle at 4 metres, but the highest levels were detected at 2 metres.

The analysis of evidence from the three studies provides useful information for which observation should be considered. Study one highlighted how important the transfer of GSR can be, the evidence suggests that significant one and two component particles can be transferred directly from a firer to a recipient. If large amounts of GSR particles are directly transferred, then it can be assumed indirect contact or presence when a weapon was discharged would also lead to the recovery of GSR particles.

Study two allows a link between ammunition and weapon to be established, by looking at the chemical composition of the particles from both cartridge case and gun barrel we can identify component elements within the particles. The analysis suggests that it's possible to distinguish between types of ammunition, which would provide important evidential information when recovery of GSR has been carried out.

Study three provides evidence that suggest GSR particles can travel up to 10 metres from a firearm though typically three-component GSR particles will only travel to around 3 metres. These results directly link to study one, and relate to the transfer of GSR, proving that being in the vicinity of the discharge of a weapon can lead to the presence of those particles. Together the results from these studies can assist with the interpretation of GSR evidence.

Keywords: Gunshot Residue, Scanning Electron Microscope, Energy Dispersive X-Ray Analysis, Forensic Investigation, Range of firing, Transfer

Chapter 1: Introduction

1.1 Firearms

A firearm is a mechanism which is mounted in an ergonomically designed frame from which a projectile exits through a tube (White, 2016). In the UK, the availability of firearms is restricted, with in depth controls and criminal checks required for an individual to own a firearm. Due to the relatively low ownership of guns by UK residents, alongside different laws depending on firearm type, some firearm types are prohibited in the UK, compared with countries where firearms are readily available such as the United States of America, firearm related homicides are statistically low in comparison. In the year between March 2018 and March 2019, there were 9,787 firearms related offences in England and Wales and in 51% of those cases the firearm had been discharged. The types of weapons used in those cases can be seen in Figure 1 (Office for National Statistics, 2020), where 45% of cases involved an imitation or air gun firearm, these fire small pellets at a lower velocity. In comparison, gun deaths in the United States of America totalled 38,355 in 2019 and 14,414 of these were homicides involved a firearm (BBC, 2021).



Figure 1: Chart displaying offences recorded by the police where firearms were reported to have been used, by type of weapon in England and Wales, in March 2019 (Office for National Statistics, 2020).

1.2 Firearm Mechanisms

A round of ammunition is made up of the bullet, cartridge, primer, and propellant. Often erroneously referred to as a bullet, the round is comprised of a cartridge case, primer, propellant, and a projectile. The types of ammunition that are commonly encountered are shotgun, pistol, revolver, and rifle. Typically, cartridges made for rifles are made from brass and the bullet is found in the neck of the cartridge. These components can be seen in Figure 2 (White, 2016; Wallace, 2017).



Figure 2: Rimfire, centrefire and shotgun round cross sections showing typically where the primer, propellant and bullet are within a cartridge (Wallace, 2017).

Shotgun ammunition differs from other cartridges. This ammunition is usually comprised of shot pellets, shotshell casing, primer, powder, charge, and wadding. (Schwoeble and Exline, 2000). The cartridge can provide a huge amount of information, most importantly its headstamp, which is usually an identifying mark for the firm that made it.

Ballistics is the science of propulsion, flight, and impact of projectiles. There are three main fields of ballistics: internal, external, and terminal ballistics. Internal ballistics focuses on the characteristics of the projectile while it is still inside a firearm. External ballistics look at what happens when the projectile exits the firearm. Terminal ballistics cover what happens when the projectile hits something (White, 2016). When a firearm is discharged there are impressions made on the bullet called rifling marks and striations. These markings help link the cartridge to a particular type of firearm by direct comparison (Sutton, Trueman and Moran, 2017).

1.3 Gunshot Residue

Gunshot Residue (GSR), Firearms Discharge Residue (FDR) or Cartridge Discharge Residue (CDR) is the residue which forms when a firearm has been discharged (Blakey et al., 2018). This residue is made up of burnt and unburnt spherical and irregular particles from the primer compound (Saverio Romolo and Margot, 2001). It may also include material from the cartridge, the projectile, and the firearm as well as organic GSR from the propellant (French, Morgan and Davy, 2014). The round contains several different components. These include (a) a projectile, which is generally made from lead alloy and is often jacketed with gilding metal, (b) the cartridge case, which is typically made from brass, (c) the propellant which contains mostly nitrocellulose and usually other components such as nitroglycerine and lastly (d) the primer cup that contains a primary explosive, typically a type of fuel and an oxidant (Brozek-Mucha, 2011). GSR is primarily particulate matter, but it can also be characterised as volatile and gaseous (Maitre et al., 2017). The GSR residue is a result of a firearm discharge that occurs when the firing pin hits the shock and impact sensitive primer, leading to detonation. This primer then ignites the propellant inside the cartridge and the result is an ejection of the particulates (Bell and Seitzinger, 2016). There is an increase in temperature and pressure, followed by a rapid expansion of the gases once the projectile has been released, which in turn results in the rapid cooling of the metallic vapours to create condensed droplets (Brozek-Mucha, 2007). When discharging a firearm, the internal barrel temperature can reach approximately 1500-3600°C with a concurrent pressure of 1400-40,000 psi, though

this will vary depending on the firearm and ammunition type used (Coumbaros *et al.*, 2001). Many inorganic GSR particles formed from the primer have a spherical morphology and generally have an average diameter that ranges between 0.5 μ m and 100 μ m (Ali *et al.*, 2016). GSR particles are created when the gases created by firing condense, this can be observed in the surface and interior of the particles (Coumbaros *et al.*, 2001). These particles are commonly analysed for forensic purposes using a scanning electron microscope (SEM) coupled with energy dispersive X-ray spectroscopy (EDX) (Costa *et al.*, 2016).

1.4 Organic gunshot residue and Inorganic gunshot residue

Inorganic GSR (IGSR) originate from the primer. The inorganic particles for sinoxid ammunition are primarily composed of lead (Pb), barium (Ba) and antimony (Sb), though they do contain some organic components, these are not usually looked for when using SEM/EDX due to the sample typically being carbon coated (Sarvas *et al.*, 2009). As per the American Society for Testing and Materials (ASTM) guidelines these are the elements that must be present to allow a characteristic identification of GSR from sinoxid ammunition (Dalby, Butler and Birkett, 2010, Ali *et al.*, 2016). The lead, barium and antimony that originates from the primer tends to come in the form of lead styphnate, barium nitrate and antimony trisulphide (Luten *et al.*, 2018). Particles that contain just two components (Pb/Sb, Pb/Ba, Sb/Ba) are identified as being consistent with GSR (Brozek-Mucha, 2017). Particles that contain just one element are named as indicative of GSR. This practice is consistent in many forensic laboratories (Brozek-Mucha, 2009). It should be noted that some primers do not contain one or even two of the components typically associated with GSR, which

can and has previously resulted in false negatives in cases. As a result, forensic examiners are recommended to utilise a case-by-case approach wherever possible (Charles, Nys and Geusens, 2011; Gallidabino, M.*et al.*, 2015). UK forensic providers commonly use a classification system for GSR. The current system has been in place since the 2000's and involves 9 types of classification, as seen in table 1.

Table 1: A list of the composition of the 9 types of classification for GSR (Shaw,2020).

Туре	Classification
Type 1	Lead, barium, antimony
Type 2	Lead, barium, antimony, aluminium
Туре 3	Lead, barium, antimony, tin
Туре 4	Lead, barium, calcium, silicon
Type 5	Lead, barium calcium, silicon, tin
Туре 6	Mercury, antimony, barium
Туре 7	Mercury, antimony, barium, tin
Туре 8	Barium, lead
Туре 9	Titanium, zinc

Organic GSR (OGSR) residue originates from the propellant. OGSR is generally formed from explosive materials which come from unburnt propellant, while the IGSR is formed from a mixture of the primer, projectile, and the cartridge case (Bueno and Lednev, 2014). While IGSR typically contains lead, barium, antimony and possibly zinc, aluminium, copper, and iron; there are hundreds of organic compounds that could potentially be related to OGSR though it usually consists of nitrocellulose, nitroglycerin, diphenylamine, dinitrotoluene, resorcinol, cresol and trinitrotoluene (Gandy *et al.*, 2018; Goudsmits *et al.*, 2019). It is possible to detect OGSR from propellant powder or spent cases and methods to detect the organic compounds have been previously researched, such as, GC-MS, IMS and UPLC-MS; however, a global standard method to collect, extract and analyse OGSR has not yet been identified (Goudsmits *et al.*, 2019).

Inorganic GSR is detected using an analytical method called IMS which is based on the identification of particulates that have a specific size and morphology and which also include lead, barium, and antimony (Arndt *et al.*, 2012). Forensic laboratories employ methods of analysis which focus on the detection of IGSR utilising SEM-EDX (Goudsmits *et al.*, 2015). Due to the introduction of lead-free and nontoxic cartridges in recent years, research is now beginning to focus more on OGSR particles (Khandasammy *et al.*, 2019). Metal-free or specifically lead-free cartridges have been designed to minimise shooters exposure to any toxic-metals and aimed at those using firing ranges due to the high quantity of lead typically found in those areas. However, the effectiveness of the GSR classification remains highly dependent on the ammunition involved (Heringer and Ranville, 2018).

1.5 GSR in the Environment

One of the key challenges in interpreting GSR data is the attribution of GSR to an individual, while also assessing the probability of random or innocent recovery of GSR particles in the wider environment (Lucas *et al.*, 2016). The confirmatory identification of these particles can establish whether a firearm has been discharged

or if the suspect has been in the presence of a weapon being discharged (Garofano *et al.*, 1999). It is suggested that a Bayesian approach should be used for evaluating GSR evidence. For this, two factors should be considered. Firstly, the number of GSR particles expected and secondly the probability of random GSR particles occurring (Biedermann *et al.*, 2010). Firearm laws and firearm availability is considerably different across the world. This leads to the importance of interpreting the discovery of GSR in certain circumstances so that the risk of contamination can be assessed. (Brozek-Mucha, 2014).

GSR can be looked at as a population of particles, but critically a minimum criterion for what can be classed as a positive recovery is also required (Hannigan et al., 2015). The Federal Bureau of Investigation (FBI) laboratory required a minimum of three Pb/Sb/Ba particles for a population to be confirmed with the addition of other GSR consistent particles (Wright et al., 2006). However, the FBI no longer accept GSR cases as they put the resources they had to do so toward fighting terrorism. This is similar to the position of several UK forensic service providers. The Forensic Science Service (FSS) guidelines from 2006 recommended not putting significance on a low level of GSR, which they defined as one to three particles. Following on from the privatisation, most UK forensic service providers continued to adopt the same cautious guidelines (Forensic Science Service, 2006). Where most will consider a finding of three or fewer GSR particles inconclusive, though there is currently not enough published work to provide a robust justification of that cut-off level. In comparison, the US Army Criminal Investigation Laboratory requires a minimum of four Pb/Sb/Ba particles for a population to be confirmed, however this is due the area being a military setting and already having high background levels of

GSR present. For the purpose of research, a case-by-case laboratory criterion should be followed; on average laboratories class a minimum of three, three component particles a high enough level for a positive result (Hannigan *et al.*, 2015). More experiments on levels of GSR in the general environment and the transfer to non-firearm users are critically important for the correct interpretation of GSR results. When GSR evidence is presented in court it must be understood that there is always a possibility a positive GSR result could be collected from a person unrelated to the incident. A main concern is false positives from contact with the police, due to the potential for officers to have a high background level of GSR present on their clothing or within police vehicles (Lucas *et al.*, 2019). Ultimately the decision to enter any GSR evidence in a trial remains the choice of the courts (Shaw, 2016).

1.6 Deposition

Following the discharge of a firearm, the residues created by firing the weapon are then deposited on the shooter's clothing, skin, and the firearm itself (Kara *et al.*, 2015). GSR has a half-life of approximately 1 hour (Brozek-Mucha, 2011) in terms of persistence on the skin of a shooter, though this does depend on the activity of the shooter after firing a weapon. GSR evidence can be lost through handwashing and movement. The material of the clothing worn can also affect evidence as there are several materials like silk or leather which will not retain GSR as well as materials like wool or cotton, due to the fibres present in the clothing. Research by Arndt *et al.* (2012) suggested that the ideal time to take samples is within 4 hours, after which the particles present will decrease rapidly. Typically, GSR evidence is collected from the hands of a suspect, which is usually where the highest concentration of GSR will be after the discharge of a firearm. However, if the collection is not taken within 4

hours of discharge, then samples should be collected from the clothing, hair, and face as an alternative as GSR has been shown to persist for longer in these areas (Zadora and Brozek-Mucha, 2003). Arresting a shooter directly after the discharge of a firearm is not a common occurrence and due to the rapid loss of GSR particles from the hands the remaining residue is likely to consist of only a few picograms of GSR, Zeichner does not give how many picograms of GSR this would be, probably due to the high variability caused by differing times and activities between firing and arrest, however it could be inferred that it would be fewer than the 100+ particles found on close range targets and gun barrels (Zeichner, 2003).

The initial deposition of GSR is known as primary transfer, which occurs as a result of firing the firearm. From this, the secondary transfer of the particles from one place to another is possible (Brozek-Mucha, 2014). GSR 'patterns', otherwise known as the typical spread of GSR after firing, will vary significantly between different firearm-ammunition combinations. This can be due to the weight of the bullet, calibre, powder type and composition of the ammunition along with the barrel length and loading method (Gagliano-Candela *et al.*, 2008). Distribution of GSR particles will be influenced by the distance from the shooter and the time after shooting along with the activity of the shooter (Ditrich, 2012). Even when the same type of ammunition is used in the same firearm there can still be variations in the composition of the GSR. Factors that influence this variation include the history of the firearm i.e. its previous usage, the chemical composition of the primer and gunpowder, and physical conditions such as the external the temperature at the time of shooting as well as the temperature of the chamber barrel (Rijnders, Stamouli and Bolck, 2010). When a three-component particle of IGSR is found it is believed it occurred by one of three

ways, the subject in question used a firearm, the subject in question was in close proximity when a firearm was used, or that the subject in question came into contact with someone or something that previously had GSR on it. However, the absence of GSR does not mean that a subject was not exposed to GSR. Factors like the weather, time, washing of hands and clothes or other activity could result in the complete loss of any GSR particles (Patterson, 2014).

The most common method of sampling GSR consists of using aluminium stubs topped with carbon coated tape. The stubs can be mounted directly into the SEM to be examined (Charles, Lannoy and Geusens, 2013). Aluminium stubs with adhesive tape or tabs are favoured over liquids due to the liquid being weaker in its adhesion, and liquid alone cannot be put inside the SEM. The liquid commonly used for GSR analysis is petroleum ether. Particle retention is also superior on stubs compared to liquids as the adhesive layer of liquid is thin which means it has less of a surface area, while particle collection using stubs is quicker and more efficient to use in the field (Reid et al., 2010). IGSR is best detected using an SEM coupled with EDX. This technique is non-destructive while allowing chemical and morphological identification to take place. Other techniques like atomic absorption spectroscopy (AAS) and neutron activation analysis (NAA) can also be used and may be the preferred choice depending on the quantity of GSR present (Chohra et al., 2015). Certain brands of 0.22 " calibre ammunition do not always contain all three characteristic components of GSR and may only contain lead and barium, or simply just lead. These particles are more likely to have more refined chemical details and thereby requiring SEM/EDX analysis (Coumbaros et al., 2001).

Clean range ammunition is a type of ammunition that does not contain heavy metals and is jacketed to prevent the release of lead, so as to minimise the shooter's exposure to harmful chemicals. In cases where clean range ammunition has been used methods like capillary electrophoresis, liquid chromatography mass spectrometry (LCMS) and X-ray fluorescence can be used to assist in differentiating between the ammunition types (Costa *et al.*, 2016). Some findings have highlighted that the components of GSR are regularly found in firework particles and vehicle brake linings, though they can be distinguished from one another as fireworks contain magnesium and particles from brake linings contain high amounts of iron and sulphur (Grima, M. *et al.*, 2011). It has also been proposed that investigating OGSR may be more useful than originally thought though the methods for identifying the organic particles are not regularly used in casework laboratories (Dalby and Birkett, 2010).

More recently, studies have investigated other methods of analysing IGSR. Currently inductively coupled plasma-mass spectrometry (ICP-MS), inductively coupled plasma-optical emission spectroscopy (ICP-OES) and laser induced breakdown spectroscopy (LIBS) have been reported as fast analysis techniques in the detection of IGSR (Tarifa and Almirall, 2015). There is a limited scope of studies regarding the dispersion distances of GSR. Schwoeble and Exline (2000) investigated how the GSR plume is dependent on the type of firearm being fired, CCI, FED, REM and WIN ammunition was used. Fojtasek *et al.* (2003) measured GSR distribution at floor level and found none further than 6 m in an open environment and 10 m in a closed environment, using 9 mm Luger Sellier and Bellot ammunition. In contrast Gerard *et al.* (2011) found GSR at 18 m from the muzzle though this is thought to be

associated with transport on the bullet. In these experiments less than 10 GSR particles were found at distances of 10 m and more, the ammunition used for their study was 9 mm luger FMJ ammunition by Sellier & Bellot. Hofer, Graf and Christen (2017) used chemiluminescence to look for OGSR at various distances and found levels dropped to nothing by 3 m, different ammunition types were used in this experiment, GECO 9 mm luger, RUAG 9 mm luger, Fiocchi 9 mm luger and S&B 9 mmm luger, while Zapata *et al.* (2018) used multispectral imaging to find an exponential decrease of GSR from 10 cm to 2.2 m shooting distance. For their experiments 9 x 19 semi-jacketed hollow point ammunition by Sellier & Bellot was used.

Brozek-Mucha (2009) reported the fraction of Sb particles increasing from 10 cm to 1 m from the discharge while the number of particles larger than 4.5 µm increased with distance from the gun. This does not agree with Gerard *et al.* (2011) who found fewer particles larger than 8 µm further from the discharge. If a large and small GSR particle left the muzzle of a gun with the same velocity, then it would be expected that the larger particle would travel the farthest distance due to having higher momentum from its greater mass. There are other factors like surface area, morphology and collisions between particles being emitted from the gun that will affect the distance the particles travel.

1.7 Transfer and Prevalence

The transfer and prevalence of GSR has not been extensively researched and is not well defined. However, there are several significant publications that have detailed some key findings on this subject. French and Morgan (2015) noted that GSR can be

transferred when contact is made with a surface upon which GSR is present or by being in the vicinity when a weapon is discharged. This research was conducted by creating transfer and deposition scenarios to mimic firstly; a scene in which a subject who wasn't present at the moment of firearm discharge made contact with an individual who was present. This was performed by having a shooter discharge rounds and then shake hands with someone who had not been present when the rounds had been discharged and a second scenario in which someone was standing a metre behind the shooter during firearm discharge. In both scenarios a SIG Sauer p226 self-loading pistol was used and five rounds of 9 mm Luger 95 grain jacketed soft point 9P1 ammunition were fired. The authors discovered that direct contact by shaking hands with a shooter resulted in significant GSR deposit, during the three runs carried out between 200 and 647 GSR particles were recovered. While the second scenario, an individual standing a metre behind the shooter, was found to have high amount of GSR present, the number of particles found on the bystander were 21, 36 and 28 in runs 1, 2 and 3 respectively. After the contact by secondary transfer 9 to 29 GSR particles were recovered during the three runs. Finally, the particles recovered from the tertiary transfer ranged from 12 to 22 GSR particles for the three runs. This simulation demonstrated that direct transfer and secondary/tertiary transfer of GSR is a viable possibility and that in each of the scenarios GSR particles could be recovered from the third participant in the transfer chain. By contrast an earlier paper by Hannigan et al. (2015) surveyed clothing for background levels of GSR following non-firearms offences, this research took place in Ireland. They found that the detection of GSR on clothing is more common than the detection on the skin but still a rare occurrence. 100 clothing garments were submitted into examination in relation to non-firearm offences, the cuffs of each

garment were sampled with a 12.5 mm carbon coated stub. Out of the 100 clothing garments that were sampled 98 of them did not contain the three component Pb/Sb/Ba GSR particles. One singular round Pb/Sb/Ba particle was found on one garment and a second garment contained two Pb/Sb/Ba particles and 10 GSR consistent particles. Hannigan et al. (2015) continued to investigate the occurrence of residue types and carried out a discharged ammunition survey. This involved sampling 100 discharged rounds that had previously been collected from crime scenes. Small foam tipped swabs were used to sample each interior of the discharged rounds. The foam tip was then rolled onto a 12.5 mm carbon coated stub. The following five residue types were found from the results of the ammunition survey: Pb/Sb/Ba, Pb/Sb/Ba/AI, Pb/Sb/Ba/Sn, Pb/Ba/Sn and Pb/Ba. From these findings the authors concluded that the presence of GSR on clothing is uncommon. Significantly all the clothing items tested in this study had been removed from suspects who had been transported in police cars and had then subsequently been inside police stations. From the clothing survey it was concluded that the chance of finding at least one 2-component GSR particle or two 2-component particles on clothing is 0.01. It was also concluded that the probability of finding one or two 3component GSR particles on clothing by chance is 0.02 and that finding more than two 3-component GSR particles is much less than 0.02. This means that while the chance of finding 3-compnent particles is possible, it is uncommon, though this information can allow the evaluation of the evidence and how significant it is. However, the research carried out by Hannigan et al. (2015) differs from the other research mentioned, as this research was based on two different surveys and focused more on the prevalence of GSR. The first survey was on pieces of clothing which had no relation to any shooting events, while the second survey was based on

testing actual discharged ammunition. While the research still makes for an important contribution to the field in terms of background GSR levels it does not include or replicate any sort of real-life event or case scenario event.

A previous study by Charles and Geusens (2012) led to the evaluation of the overall contamination of GSR during the arrest of a suspect. The risk of contamination in this scenario was evaluated as high. The authors conducted two scenarios, one a high contamination and the other a low contamination. The low contamination scenario consisted of police officers wearing regular civilian clothes but also having their gun and handcuffs. They proceeded to load their weapon and carry out an arrest by laying down the arrestee, handcuffing and frisking them. Both the hands of the officers and the hands of the targets were sampled. In this scenario, less than 4 Pb/Sb/Ba particles were found on the hands of police officers and an average of 1 Pb/Sb/Ba particle was recovered from the targets. In the high contamination scenario, police officers were again dressed in civilian clothes and had their gun and handcuffs but also had a bulletproof vest, a technical vest, and gloves. They carried out the same arrest as the previous scenario. This study suggested the level of contamination was higher on suspects during the frisking process and that the highest source of contamination came from the gloves of the police officers involved. An average of 66 Pb/Sb/Ba particles were found on the officers' gloves, between 3 and 6 Pb/Sb/Ba particles were found on the officers' hands and between 7 and 22 Pb/Sb/Ba particles were found on the officer's vests. A recent study carried out by Blakey et al. (2019) focused on vehicles that had the potential to be the source of secondary or even tertiary GSR transfer. The authors noted that there is potential for police facilities, personnel, and vehicles to act as the source of GSR contamination.

Blakey *et al.* (2019) began their experiment by taking 170 samples from seven recreational shooters vehicles. The areas that were sampled were based on the most probable locations GSR would be transferred to based on contact. Characteristic GSR was discovered in all the vehicles sampled, which suggests that the interior of motor vehicles are environments that can facilitate the direct transfer and persistence of GSR particles. These findings indicate that GSR found within a vehicle may not be directly related to a firearm being discharged within it but could in fact be due to secondary transfer. In all seven vehicles Pb/Sb/Ba particles were recovered ranging in numbers from 12 particles to 816. Police personnel and recreational shooters are both regularly exposed to higher levels of GSR than typical civilians, therefore could be a major contamination factor, but little was known about how much their vehicles could be a subsequent source of contamination.

Ali *et al.* (2016) and Berk *et al.* (2007) carried out similar research using police vehicles and stations. Ali *et al.* (2016) sampled different areas of a Pittsburgh police station including the interview room and areas of a holding cell. They also simulated an arrest involving a police vehicle, where after a set amount of time in the vehicle the arrestees were sampled. Out of all the samples taken only one characteristic GSR particle was found on the interview desk within the police station. Berk *et al.* (2007) also examined samples from vehicles and Chicago based detention centres. No particles were found in the marked squad cars, marked squad rolls or detective cars; however, 2 Pb/Sb/Ba particles were found in tactical cars, 34 Pb/SB/Ba particles were found on table type surfaces and 20 Pb/Sb/Ba were recovered from the restraining bars. Both studies found that there is potential for secondary transfer of both IGSR and OGSR from a police vehicle or from a police station onto a

suspect; however, the potential for this occurrence is still quite small and not of great importance due to the small number of particles which are likely to be found. Berk et al. (2007) note that it's important for a baseline to be created for the amount of GSR present in a police vehicle or police station, so that any number of particles above this baseline can be contributed to tertiary or secondary transfer. Lucas et al. (2019) also proposed that although secondary transfer of GSR is possible, it should be a minor factor when evaluating any GSR evidence. The authors investigated GSR persistence on the hands of police officers who were carrying but not discharging a weapon. They investigated the background level of GSR on an officer's hands, specifically if an increased background concentration of GSR particles could be transferred to a person of interest. The authors also created a simulation to assess what amount of GSR could be transferred during an arrest. They found that in total, 788 Pb/Sb/Ba particles were recovered after an officer had handled a firearm following hand washing, and that 252 Pb/Sb/Ba particles were found on the officers when they had washed hands after handling a firearm. The data indicated that a very low amount of characteristic GSR was found on the officers during the first part of the assessment. The average frequency of an officer having at least one three characteristic or consistent particles was 7.9% from a police population of 76 compared to the random population of 289 tested subjects. Therefore, they determined that the likelihood of secondary transfer from an officer to person of interest during an arrest although possible was not a significant issue for consideration in casework settings. Despite the experiment varying quite differently in the way it was carried out to the others, the results gathered seem to follow the consensus that a person can be contaminated by secondary transfer they do

however state that further prevalence studies should be carried out on a much larger scale.

1.8 Collection and Analysis Techniques

As previously stated, the use of adhesive stubs is the preferred method for collection of GSR although the make and model of the stubs vary from laboratory to laboratory. Lucas et al. (2016) used SEM pin stubs at a standard size of 12.5 mm, which were held in closed plastic vials and were either covered with transparent double sided adhesive tape or double-sided carbon adhesive tape and analysed their samples using a CamScan Apollo SEM with Genesis EDS system and a Zeiss Evo 50 SEM with Oxford EDS System. Cook (2016), Brozek-Mucha (2011) and Blakey et al. (2019) carried out their research using similar methods. Cook (2016) used slightly smaller 12 mm standard aluminium pin collection stubs, which had a carbon adhesive tape and for analysis also used a Zeiss Evo 50 SEM with Qemscan BSD, Oxford instruments X-MayN 80 mm 2 silicon drift detector and Inca GSR-analysing software; Brozek-Mucha (2011) also used aluminium stubs with a conductive carbon adhesive tab. The aluminium stubs were then coated with a conductive layer of carbon using a SCD 050 sputter, BALTECH and were then examined using a JSM-5800, JEOL SEM coupled with an INCA energy X-ray spectrometer, Oxford Instruments Ltd. This is the same technique of collection and analysis used by Brozek-Mucha in a previous study dating to 2011. Blakey et al. (2019) used 12.5 mm SEM stubs with a carbon tab, these stubs were then coated with a conductive layer of carbon using Quorum Technologies Q150T ED Rotary-Pumped Carbon Coater. The stubs were analysed using a FEI Quanta200 ESEM, with automatic search from INCA GSR software.

In comparison Manganelli, Weyermann and Gassner (2019) used a similar method of collection; 12 mm carbon tabs mounted onto 12.5 mm aluminium stubs which were held in plastic vials with screw caps. A different method of analysis due to them focusing on OGSR was carried out using an Agilent Infinity 1290 ultra-high performance liquid chromatography (UHPLC) from Agilent Technologies. The use of aluminium stubs with carbon coated tabs is a seemingly reliable method for the collection of GSR samples.

1.9 Scanning Electron Microscope and Energy Dispersive X-Ray

The SEM was first introduced as a method for detecting IGSR in 1974 and since then it has become an internationally accepted method for GSR analysis (Taudte et al., 2014). GSR particles are commonly analysed using an SEM coupled with EDX, as this method allows the analysis of both the morphology and chemical composition of the particles (Ali et al., 2016). The use of the SEM is thought to be superior to other methods of analysis for GSR due to it being able to characterise individual particles morphologically and elementally (Lebiedzik and Johnson, 2000). The SEM is composed of an electron column, a specimen chamber, and a computer control system (UI-Hamid, 2018). Within the SEM, a beam of electrons is accelerated down a column by a high anode potential. The electrons are produced typically from a hot metallic, electron rich filament, typically a tungsten filament but this is not exclusive. The filament choice will influence the spot size. Typically, three electromagnets are then used to focus the beam on the sample. Incident electrons will then cause electrons to be released from the sample due to elastic and inelastic scattering events that happen at the samples surface. Backscattered electrons are the result of elastic collisions of the incident electrons, whereas secondary electrons are released

from the surface atoms following inelastic collisions. Inelastic collisions will also produce X-rays characteristic of the atoms in the samples surface. The emitted backscattered electrons, as seen in Figure 3, allow GSR to be located due to heavy metals present and the characteristic X-rays identify the chemical composition of the particle (Wallace, 2008).



Figure 3: An image showing the electron beam focusing on an image and the backscattered electrons being emitted from the object (Robertson, 2013).

An SEM has a depth of field which is 200 times greater than that of an optical microscope, which gives it incredibly high resolution and magnification potential (Saverio Romolo and Margot, 2001). The SEM must be kept under a high vacuum during use to enable the electrons to pass through the column without being affected by air molecules. The electron column is generally under a high vacuum of 4-10 Pa, while the specimen chamber is usually under a vacuum of 3-10 Pa (UI-Hamid, 2018).

For the SEM to create a backscattered image, the electron beam scans over the samples surface in a raster pattern. Each electron which is scattered from the sample is detected in each position by the electron detector. Depending on the intensity of the electron signal, this is displayed as a brightness on the screen. The higher the atomic number, the brighter the sample will appear. When the monitors scan is matched with the scan of the electron beam the monitor can then display an image which will represent the sample surface's morphology. Following the creation of this image the X-ray microanalysis technique can be used. When analysing the image formed by the SEM the electron beam can be made to stay in a fixed position over an area that may be of interest. When the electron beam penetrates this area, it excites X-rays from the whole excitation volume. The size of the volume is determined by two things: the accelerating voltage and the mean atomic number of the sample.

For the purpose of this study, X-ray spectra were collected from single areas chosen on each of the analysed particles. EDX allows the chemical classes of particles to be defined, this is based on the elements present (Phermpornsagul *et al.*, 2020). The Xray energies within these spectra determine the presence or absence of lead, barium, antimony, and any other relevant elements. The beam of electrons is focused onto the sample causing electrons in the sample to be ejected. Higher energy electrons in the atoms of the sample will then drop down to fill the vacated energy levels resulting in the simultaneous emission of an electromagnetic wave with an energy corresponding to the energy gap the electron has dropped down. This wave will have the wavelengths of X-rays, as seen in figure 4, and these

wavelengths are characteristic of the elements present. EDX is used to analyse the X-rays to assign the appropriate elements (Ebnesajjad and Ebnesajjad, 2014).



Figure 4: A schematic which describes the EDX spectroscopy (Colpan, 2018)

One major disadvantage of the SEM is the time taken to analyse even one singular stub. Therefore, the relative sample area that can be sampled on an SEM stub and the small size of particles within the area of the stub requires a large volume of evidence to be collected for examination. Due to this, the majority of SEMs are used alongside an automated search software, which allows different search criteria to be input and which generates an image of each particle so that the morphology can be examined (Dona-Fernandez *et al.*, 2018). The automated SEM system is otherwise known as computer-controlled scanning electron microscopy (CCSEM) (Schwoeble and Exline, 2000).

1.10 Aims and Hypotheses

Study 1

The aim of study 1 was to determine the extent of GSR transfer to clothing following contact between a shooter and a test subject. This is to be measured by counting the numbers of particles consistent with and characteristic of GSR on the test subject's t-shirt.

The hypothesis is that following close contact between the shooter and the test subject, high levels of one, two and three component GSR particles will be detected on the clothing of the test subject.

Study 2

The aim of study 2 was to distinguish between two types of ammunition when discharged from a 9mm parabellum calibre Steyr M9 semi-automatic pistol, according to the composition of the GSR particles produced. The study also aimed to determine the possibility of identifying which ammunition was fired last.

The null hypothesis for this study is that the composition of the two ammunition types is indistinguishable.

 H_0 = It is not possible to distinguish between the composition of GSR produced by the two different types of ammunition.

 H_1 = It is possible to distinguish between the composition of GSR produced by the two different types of ammunition.

<u>Study 3</u>

The aim of study 3 is to determine whether the calibre of firearm and ammunition affects the distance GSR travels by measuring the number of GSR particles on targets set at different distances.

The null hypothesis for this study is that there will be no significant difference in the distance GSR travels with different calibre firearms and ammunition. $H_0 = GSR$ will travel a consistent distance regardless of firearm and ammunition used.

 $H_1 = GSR$ from shotgun ammunition will travel further than lower calibre ammunition.

Overarching Aim

The overarching aim of this research was to investigate the transference of GSR to the clothing of an individual who had not been in direct contact with a firearm. In addition to these contact experiments, this study will also determine the potential distance GSR travels from a firearm and delineate if GSR can be matched to a specific brand of ammunition; thereby linking it to a specific shooting incident. Furthermore, experiments on the distance of GSR travelled from separate types of firemarms will also be conducted, to ascertain the extent of the deposition area of GSR at the scene of a shooting and therefore identify the extent of potential sources for transfer.

Chapter 2: Materials and

<u>Methods</u>

2.1 Instruments and laboratory equipment

The instruments and equipment used within the studies are as follows, the initial sampling was carried out using 12 mm diameter aluminium Cambridge pin stubs with carbon coated sticky tabs from Agar.

Further examination was carried out using a Scanning Electron Microscope Zeiss Evo 50 SEM with associated Oxford Instruments Inca Analytical Suite utilising X-act EDS Detector. The primary opening parameters are: 20 kV accelerating voltage with a 2 nA beam current.

2.2 Study one: the transfer of GSR particles

The first experiment was designed to determine if GSR could be transferred from a shooter to a test subjects clothing and if it could be transferred, how much GSR would be present. A volunteer was used as the test subject and close body to body contact in the form of a hug was carried out to create the transfer, the volunteer being the recipient. The clothing worn by the recipient was Primark own brand black t-shirts made from 65% polyester and 35% cotton, purchased from Primark. The firearm used for this study was a Ruger 10/22 LR semi-automatic rifle, used alongside CCI standard velocity .22 " long rifle round nose cartridges. This was then followed by analysis of the clothing worn to measure the amount of GSR present.

2.2.1 Sample preparation and collection



Figure 5: Photograph showing a semi-automatic ruger (gun 28) which was the firearm used in study one, property of Surbiton Rifle Club.

The transfer of GSR particles study was investigated at Surbiton Postal Rifle Club. On arrival at the club both participants thoroughly washed their hands and changed into clean, previously unworn clothing. The shooter of the firearm proceeded to enter the range and fire 20 CCI standard velocity .22 " long rifle round nose cartridges from a ruger semi-automatic rim-fire rifle as seen in figure 5. The rifle was positioned on the right-hand shoulder of the shooter, the barrel of the rifle was held with the left hand and the trigger was pulled with the right hand. Once this was completed, the shooter exited the range and came into contact with the test subject in body-to-body contact for 5 seconds. The test subject removed the t-shirt where it was folded and placed in a brown paper evidence bag. Following this, both the shooter and test subject thoroughly washed their hands and changed into clean clothing. These actions were repeated until five contaminated t-shirts had been collected from the test subject. One t-shirt that had been worn by the test subject prior to the experiment and that had not come into contact with the shooter had been placed in an evidence bag to be used as a control.

Laboratory analysis of the samples utilised labelled SEM stubs as seen in figure 6, which coincided with the side and number of the top the sample was taken from. A sample of the clean washed bench where the t-shirts were sampled was also taken as a control, the bench was cleaned with water and diluted soap. Evidence bags containing the contaminated t-shirts were placed on a separate bench. Brown paper was placed on the bench and the first evidence bag was cut open. Clothing was removed from the bag with a gloved hand and placed on the brown paper. The t-shirt was sampled in four sections (front left, front right, back left, back right) using aluminium 12 mm Cambridge pin stubs with carbon coated sticky tabs. The brown paper was replaced, and the bench cleaned for each tested t-shirt. In total 25 samples from study one were collected for SEM for analysis.

2.3 Study two: differing ammunition types

The second study carried out by the Garda Siochana was to determine if it is possible to measure the extent of GSR mixing when different ammunition is fired
from the same gun and to also see how much GSR carries over when two different ammunition types are used in the same firing apparatus.



Figure 6: An image showing one of the aluminium carbon coated stubs which are placed inside the SEM and used in all three experiments.

2.3.1 Sample preparation and collection

The second study was carried out by the Garda Siochana in their ballistics and forensic investigation section, at the Garda National Technical Bureau, An Garda Siochana Headquarters, Phoenix Park. The discharge weapon in this experiment was a 9 mm Parabellum 'Steyr M9' semi-automatic pistol. Alongside the discharge weapon, two different types of ammunition were used, these were Italian manufactured 'Fiocchi' 9 mm Parabellum full metal jacket and Russian made 'Wolf' 9 mm Parabellum, full metal jacket rounds. The barrel of the pistol was cleaned to try and ensure no fouling would occur, and a dry control swab was taken of the barrel interior. The gun was then loaded with three rounds of the Fiocchi ammunition, these rounds were discharged, and a swab was taken of the barrel interior. Following this, the barrel was cleaned once again. Three more rounds of Fiocchi ammunition were discharged from the barrel followed by one round of the Wolf ammunition and the barrel interior was swabbed again. Once these discharges had taken place a swab from the discharged Fiocchi cartridge case and discharged Wolf cartridge case were taken.

The final stage of the experiment involved once again extensively cleaning the barrel, then three more rounds of Fiocchi ammunition were discharged from 1.5 m onto a paper target. One round of Wolf ammunition was then discharged from the Fiocchi fouled barrel into a separate target also from 1.5 m away.

All swabs and paper targets were appropriately packaged into evidence bags and sent to Kingston University. Aluminium SEM stubs were then labelled to match each swab or paper target. An initial control was taken from the clean bench, the bench was cleaned with water and diluted soap. All the received evidence bags were placed on a bench separate to the one being used for sampling. Using clean scissors, brown paper was placed on the bench and cut to the required length.



Figure 7: An image showing one of the five packaged samples sent by the Garda Siochana, the samples are barrel swabs. The image shows the control Fiocchi swab.

The scissors were then cleaned again, and the first evidence bag was cut open. The swab was removed from the evidence bag and placed onto the brown paper, once the swab was removed from its collection tube, it was rolled 20 times onto the sticky tab covering the aluminium SEM stub so that any particles would be transferred to the stub. The same process was carried out for the other four swabs received, one of these bagged swabs can be seen in figure 7. Following this, the first paper target was then removed from its evidence bag, and using an aluminium SEM stub the entire target was sampled, this was repeated for the second paper target. The brown paper was replaced, and the bench was washed down in between each swab or paper target. Seven samples were taken and placed into a stub box to be taken to the SEM for analysis, due to the sheer number of particles on these samples 100

particles were analysed using the SEM, these were chosen by searching the sample surface using a grid pattern and selecting particles that were well separated and by covering the whole sample area.

2.4 Study three: distance and dispersion

The third study carried out by the Garda Siochana was to determine how far GSR will travel in the direction the gun was pointed in when fired. Four different types of ammunition and four different firearms were used against five different target distances.

2.4.1 Sample preparation and collection

The distance and dispersion study was carried out by the Garda Siochana in their ballistics and forensic investigation section.

Table 2: Showing the firearm used and the corresponding ammunition used in the experiment.

Firearm Used	Corresponding Ammunition	
.22 " Long Rifle calibre Winchester	.22 " LR calibre hollow point	
lever action rifle	ammunition by Winchester	
9 mm Parabellum calibre Steyr M9	9 mm Para calibre full metal jacket	
semi-automatic pistol	ammunition by Fiocchi	
.223 " Remington calibre CZ 527 bolt	.223 " Rem calibre full metal jacket	
action rifle	ammunition by Hornady	
12-gauge Winchester semi-automatic	12-gauge shotgun ammunition by Eley	
Shotgun		

Five separate brown paper targets were used for each calibre firearm and large pieces of white cleaning tissue were attached to each target as this would create better retention capabilities for the GSR due to the tissue acting like clothing and retaining GSR particles like clothing fibres. A blank target was prepared before any firearm was discharged. A target for each calibre firearm was fired at from different distances, those distances were 2 metres, 3 metres, 4 metres, 5 metres and 10 metres. An authorised person not involved in the discharge and who was not present during the firing was responsible for retrieving and packaging each target. Each target was hung up from a string target line, this line was cleaned after each target was collected. The firing range used in this experiment was a 25 metre indoor range at the Garda Headquarters, Dublin. No repeats were carried out.

2.5 Analysis

Prior to analysis all stubs were carbon coated using an Agar Auto Carbon Coater (figure 8). This was to prevent build-up of electric charge on the samples when under the electron beam of the microscope. The analysis of the samples was carried out using SEM/EDX analysis and was completed on a Zeiss Evo 50 SEM with associated Oxford Instruments Inca Analytical Suite utilising X-Act EDS Detector as seen in figure 9.

The aluminium carbon coated stubs were inserted into the sample chamber, the SEM then generates a beam of incident electrons in the electron chamber which is above the sample chamber. These electrons are produced by a thermal emission source, in this case, a heated tungsten filament. The electrons are then focused into a small beam in the SEM column by electromagnetic lenses. The beam is then

positioned and focused on the sample surface by scanning coils which are close to the end of the column, the beam then scans over the sample surface in a raster pattern, which creates the SEM image.

For this research a manual search of the sample areas was carried out, this involved using the joystick, magnification and focusing tools on the Zeiss SEM's keyboard. Using the joystick, the electron beam was moved across the samples surface in a grid pattern to ensure all areas of the sample were viewed. The magnification tool was used to magnify the sample to 450x, and the focus tool was used to gain a clearer image of the area. The areas of interest were pinpointed by several deciding factors, these included the shape of the particle, the general size of the particle and the brightness of the particle. Once an area of interest was noted, preferably of a reasonably large spherical size with high brightness, the beam was focused within the centre of the particle. At this stage the EDX part of the analysis was used, once the beam was focused the adjoining station was used alongside the SEM to create an X-ray which determines what elemental material the particle consisted of. This involved 'running' the system to gain peaks of the elements involved. The higher the peak on the x-ray the more concentrated an element is within the particle. From the information the x-ray gave it could be seen whether or not the elements present were those consistent with GSR. If the elements present were consistent with GSR it was at this point that the screens were frozen and screenshots were taken of the x-rays alongside backscattered electron images.



Figure 8: An image showing the Agar Auto Carbon Coater used in all three experiments to give the aluminium SEM stubs the carbon coating that allows the GSR to be seen, based at Kingston University.



Figure 9: An image showing the Zeiss Evo 50 SEM with associated Oxford Instruments Inca Analytical Suite utilising X-Act EDS Detector used in all three experiments, based at Kingston University.

Chapter 3:

Results and Discussion

3.1 Particle distribution; Study 1

The expectation from the study is that several three-component GSR particles will be found on the t-shirts, having transferred from the shooter's hands and clothing. It is likely that the higher number of recovered particles will be found on the front right and the back left and right of these t-shirts as that is where the shooters hands and arms encountered the material.

As expected, the quality controls and bench sample were clear, and no particles were recovered from these samples. This shows there was no contamination at the site of the study, Surbiton Rifle Club, and no contamination occurred during sample retrieval in the laboratory.



Figure 10: Graph showing the total number of one and two component particles analysed in study one Vs the sample area where they were recovered. Graph includes results from all five of the t-shirts from the recipient used in the study.





A higher level of one and two component particles were expected to be seen on the front right and back left of t-shirt one. However, the front right of the t-shirt revealed very few particles, and the back left of the t-shirt revealed no particles at all, this can be seen in figure 10. This could be due to either low deposition on the firer or the type of controlled contact between the firer and the test subject, in this study the deposition on the firer was not tested however for future research into this topic the amount of deposition could be tested to allow for better comparison. In terms of the three-component GSR particles, these results differ from what was expected as the three-component GSR particles were found on the front left of the t-shirt, see figure 11. However, this could be due to where the gun was held when the firearm was discharged. The firearm was placed on the firers right hand shoulder the firers left

hand was holding the barrel of the rifle and it is common for empty cartridge casings to exit the firearm at speed within a range of different directions, this could mean that GSR found on the recipient could potentially have come from the cartridge making contact with the firer. See appendix a: results, table A.

T-shirt two was the only one to clearly show the expected higher number of particles on the back compared to the front for both one and two component particles and three component particles. The majority of the one and two component particles recovered from t-shirt two were found on the back left and back right of the t-shirt, followed by a smaller number on the front left. The lowest number of particles were recovered from the front right of the t-shirt, this can be seen in figure 10. The areas where the higher numbers of particles were recovered from are those expected if the shooters arms were the main vector of transfer, as this is where the arms of the shooter would have touched the recipients t-shirt in the controlled contact. The arms of the firer were placed on the recipient as naturally as possible while keeping the position the same for each t-shirt worn. The three-component GSR particle recovery results follow the hypothesis as the highest number of three-component GSR particles were found on the back right and back left of the t-shirt, see figure 11. It was expected that many of the three-component particles would be found on the back left of the t-shirt as that is where the firer's shooting hand would have made contact. See appendix a: results, table B.

The other T-shirts do not follow the expectation of higher levels on the back left. The third t-shirt used in the study has a higher particle count on the back compared to the front, but there is a slightly higher particle count on the front left as can be seen in

figure 10. This could be due to where the firearm was held and cartridges exiting the firearm. The three-component particles again show a higher particle count on the front left of the t-shirt as can be seen in figure 11. See appendix a: results, table C.

The fourth t-shirt in the study contains a much higher one and two component particle count on the back right than other t-shirts as seen in figure 10. Higher numbers of three-component particles were found on the front right and back right of the t-shirt as seen in figure 11. This is in line with results from the second t-shirt. See appendix a: results, table D.

The fifth and final t-shirt from the study shows a higher particle count on the front and back of the t-shirt. Figure 11 shows that three-component particles were recovered from all four samples taken from the t-shirt, with the higher particle count on the front left. See appendix a: results, table E.

3.1.1 GSR Particle distribution, number, and type; study 1

The expectation from the study was that most of the samples will have high numbers of lead, followed by other one and two component Sb, Ba/Pb, Sb/Pb, Ba with the hope that several three-component Pb, Sb, Ba particles will be found alongside those.

The results from t-shirt one shows many Pb and PbBa particles were recovered, with lead being the particle that appeared the most, specifically on the front left and back right of the t-shirt where 29 Pb particles were recovered in each instance. Alongside the Pb particles high levels of PbBa particles were also recovered, the higher

numbers of those were also found on the front left and back right of the t-shirt. Alongside high numbers of Pb and PbBa particles there were PbSb and SbBa particles found alongside two Pb, Sb, Ba particles. See appendix a: results, table F.

The results from the second t-shirt also show a high level of Pb particles, the larger number of particles found were from the front left where 20 particles were recovered followed by the back left and back right of the t-shirt where 25 particles were recovered from both areas. Alongside the Pb particles found, the second most common particle recovered from t-shirt two were the PbSb particles. The larger number of these particles were also found on the front left, back left and back right of the t-shirt. In total 7 Pb, Sb, Ba particles were recovered from t-shirt two, these particles were recovered from the back left and back right of the t-shirt. See appendix a: results, table G and figures a1-a2.

The third t-shirt gave a very similar pattern of results as the first t-shirt, a high number of Pb particles were recovered from the front left, back left and back right of the t-shirt. This was followed by several PbBa particles, the highest numbers of which were recovered from the front left and back right of the t-shirt. From the third t-shirt 9 Pb, Sb, Ba particles were recovered in total. These were found on the front left, front right and back left of the t-shirt. See appendix a: results, table H and figures a3-a4.

The fourth and fifth t-shirts had a much higher particle count than the first three tshirts, with a total of 295 particles being recovered from T-shirt four and 302 from Tshirt five, compared with 112 from T-shirt one, 168 from T-shirt two, and 179 from T-

shirt three. High numbers of Pb, PbSb and PbBa particles were recovered from Tshirts four and five from the front left, back left and back right of the t-shirt. A particularly high number of lead particles were recovered from the back right of t-shirt four where 77 particles were found. In total 7 Pb, Sb, Ba particles were recovered from T-shirt four, being found in each area. In total 14 Pb, Sb, Ba particles were recovered from t-shirt five with the highest number having been recovered from the front left of the t-shirt. See appendix a: results, table I and figures a5-a6 for T-shirt four and appendix a: results, table J and figures a7-a8 for T-shirt five.

A high level of Pb particles was common across all t-shirts. Followed by PbBa particles. The recovery of those particles were from the front left and back right, this was a common trend typical for all the t-shirts examined. A second rising trend is the numbers of Pb and PbBa particles recovered from each of the t-shirts, with t-shirt one and to the particle numbers are at a steady level between 20-30, from t-shirt 3 there is an increase of those particles to between 25 and 36. The particles retrieved from the front right and black left of the fourth t-shirt had a similar rise, with particles staying between 27-35. Finally the fifth t-shirt had a rise in particles on the same area, with particle numbers at 35-39. The numbers of Pb and PbBa particles found on the back left and front right of all five t-shirts shows a gradual trend in the rise in numbers.

3.1.2 Study one discussion

Studies carried out by Patterson (2014) and Gagliano-Candela, Colucci and Napoli (2008) show that the absence of GSR does not necessarily mean that it was not present but there are other factors like weather, time and the washing of clothes and

hands that can result in the loss of particles and that the spread of GSR will vary significantly with different firearms and ammunition. Currently UK forensic providers do not put any significance on finding three or fewer three-component GSR particles (White, 2016), however, two-component particles are not factored into this and the whole population of particles found is not considered. Hanningan *et al.*, (2015) support the idea that it is important to find a minimum criterion for what can be classed as a positive result, yet their research again only focuses on the three-component GSR particles.

The results gathered from the transfer study follows the original hypothesis, that someone who comes into direct physical contact with a firearm user who has recently fired a gun will find themselves with one, two and three-component particles present on the clothing they were wearing at the time of contact. This scenario produced a high particle count for one or two-component particles though somewhat lower for three component particles. It is expected that encountering a firearm user would transfer three-component GSR particles, yet this research showed that if only the three component particles are searched for a range of 2 to 14 particles were found even following direct contact very shortly after a firearm discharge. However, large numbers of indicative particles were found. These ranged from 108 to 281, however because one and two component particles can be produced by non-firearms sources they are not as useful in a casework context, ultimately, they are just less definitive (White, 2016).

This study was carried out with transfer conditions which should maximise the level of transfer of GSR including three-component particles. In a casework scenario it is

extremely unlikely the contact and recovery of the T-shirts would occur on such a short timescale, so it would be expected that the three-component particle count would be lower. If an innocent person with no relation to firearms were to pick up GSR by chance, the contact is more likely to be made several minutes or hours after a gun is fired and arrests and recovery of clothing after even more time. It is highly likely in those circumstances that particles would drop off the clothing of the firer creating a loss of GSR. The rate at which it would be lost from clothing would be highly dependent on the activity of the person wearing the clothing (Brozek-Mucha, 2011). Therefore, this study implies that there is a relatively low chance of picking up more than a few GSR particles from contact with a member of a gun club. Further work on persistence is needed to more thoroughly investigate how much may be transferred from a firer after different time gaps.

The current work can be compared to the study carried out by French and Morgan (2015), in which they looked at the transfer of GSR after contact. The contact they made in the study was simply a handshake and overall they found higher levels of three component transfer. As previously discussed, in a casework scenario it is more likely to find GSR on clothing than hands due to the increased persistence on clothes. Therefore, this study aimed to investigate the sampling where GSR is more commonly found in casework.

Differences between the French and Morgan (2015) experiments compared to the current work include the total size of sample area, the type of surface and the specific gun and ammunition used. The lower levels of three component particles in the current work may be due to using .22 " ammunition fired from a rifle rather than 9

mm. The greater calibre of the 9mm ammunition means more propellant and therefore more primer to ignite it, so there would be an expectation of more three component GSR particles arising from it. Also, the use of a long-barrelled firearm in a rifle means that the firers hands are further away from the muzzle which is a major point of the emission of GSR. Therefore, someone firing a handgun may be expected to have higher levels of GSR on their hands and possibly also clothing, than someone firing a rifle. For the current work the use of a rifle with .22 " ammunition was chosen because this reflects the predominant type in legal gun clubs and therefore the study follows what would be expected in the UK public. This is due to the legality of firearms within the UK, the legal guns allowed in the country; rifles, shotguns, muzzle-loading pistols, and handguns, all of which require a firearms licence. Therefore a person who picks up GSR without being connected to a crime but through contact with a gun club member is far more likely to be exposed to GSR from .22 " ammunition fired from a rifle.

It is not uncommon for the transfer of three-component GSR to be low (Charles, S. & Guesens, N. 2011) more significant is the much higher level of two-component particles. However, because two-component particles cannot be used for definitive interpretation alone (White, 2016), it is advisable to look at the whole population of particles that have been recovered as its unlikely that such a high count of two-component particles would occur randomly. The high content seen here, in addition to only a few three-component GSR particles would indicate that such particles had likely come from a firearm discharge.

From figures 10 and 11 there is some indication that higher particle counts occurred on the back left and back right of the t-shirts. This would follow expectations as the shooter's hands and arms were pressed against the back of the recipient, and both the shooter's hands and arms would have been the closest to the firearm at the time of discharge, therefore encountering a high number of particles. However, the overall distribution is not consistently clearly defined as higher on the back than front with some cases of high levels on the front left. An important factor that may have influenced this is the firearm used in this study, a ruger semi-automatic rifle. This meant that the used cartridges exited the firearm in different directions, which likely added to the spread of particles over the firers clothing. The importance of the positioning of the ejection port in determining the dispersion of GSR has been shown by Schwoeble and Exline (2000).

The implications for casework depend on the numbers of GSR particles found. It can be concluded that if a high level of three-component GSR particles, defined as three particles by Hannigan et al. (2015) are found, then it is more likely that that sample is from a person who has fired a gun rather than someone it has been transferred to. Further, the very high levels of indicative particles show that they cannot be relied on as evidence of exposure to the discharge of a gun. The study was aimed to find out if alternate explanations for the recovery of GSR being transferred in the general environment is possible, other than direct contact with a firearm or firearm user. As with all studies there are potential errors which can occur and can be the reason for certain results. With study one the initial contact and firing of the weapon occurred at Surbiton Rifle Club. This is a location which is used by members daily meaning it is a highly contaminated location. Although after each contact scenario had taken place

both the shooter and the recipient changed clothing and thoroughly washed the hands it is still possible that particles in the area, in the air or on the skin could have ended up on the clothing, alongside particles in the air, particles on the arm could have ended up on the clothing. The firer wore long sleeves during firing and transfer however the arms were not washed on either donor or recipient between each transfer.

3.2 Particle analysis; study 2

The expectation from this study is that it will be possible to distinguish between the composition of GSR produced by two different ammunition types. This study used a 9 mm parabellum calibre Steyr M9 semi-automatic pistol alongside Fiocchi ammunition and Wolf ammunition. It is expected that if there is a difference between the two ammunition types that this will be seen in the relative amounts of the component elements within those particles. Further, when the gun was fired with one ammunition followed by the other, it follows the hypothesis that it would be possible to distinguish between the composition of GSR produced by different ammunitions and that the particles on the target would largely reflect the composition of the last ammunition fired.

The initial bench control swabs taken from the laboratory were clear, meaning no contamination occurred. Where possible up to 100 particles were analysed from the six samples, both swabs and targets. The control wolf cartridge swab being the only sample where this was not possible as only 99 particles could be identified. See appendix a: results, table K.



3.2.1 GSR particle analysis, number, and type; study 2

Figure 12: Graph showing the total number of particles Vs the type of particle recovered from the cartridge swabs. Graph includes results from the two swabs, one taken from the Fiocchi cartridge, and one taken from the Wolf cartridge.



Figure 13: Graph showing the total number of particles recovered Vs the type of particle recovered. Graph includes results from two swabs and two targets used in the study.

From figure 12 and 13 the results suggest that the three-component particles were more common in the Fiocchi ammunition and that SbBa particles were proportionally higher in the Wolf cartridge case and both the gun barrel and target after the Wolf ammunition was fired. See appendix a: results, table L and figures a9-a12. To confirm this a statistical analysis was performed.

Principal component analysis (PCA) was used for unsupervised data analysis to explore any potential pattern in the separation of the samples. A scores plot (figure 14) shows which samples differ, whilst a loadings plot (figure 15) gives information on how the samples differ.

Principal components (PCs) are generally described as being orthogonal (at right angles) to each other and as such do not describe overlapping change within a data set. The first PC accounts for the largest amount variance within a data set and subsequent PCs account for decreasing amounts of variance.

PC1 scores, which account for 89% of variance in the data, can be found in figure 14, running from left to right (up and down shows PC2) in the following order (actual values are in the table within figure 14):

- Target after firing 1x round wolf from Fiocchi fouled barrel
- Swab of barrel after 1x round wolf from Fiocchi fouled barrel
- Swab from Wolf cartridge case
- Swab from Fiocchi cartridge case
- Target after firing 3x rounds Fiocchi
- Swab of barrel after 3x rounds of Fiocchi

The last two are close together on the horizontal axis and the three samples associated with the Fiocchi ammunition can be seen to the right while those from the Wolf are to the left. This shows a distinction between the samples where the Fiocchi ammunition was fired last and those where the Wolf ammunition was fired last.



Figure 14: Scores plot for particles composition variation showing PC1 on the x axis and PC 2 on the y axis. T1WF foul = sample from target following firing 1 round of Wolf after 3 rounds of Fiocchi; SD3F1W = sample from barrel following firing 1 round of Wolf after 3 rounds of Fiocchi; SCWc = sample from Wolf cartridge case; SCFc = sample from Fiocchi cartridge case; T3F1.5 = sample from target after firing Fiocchi; SDF = sample from barrel after firing Fiocchi.

The loadings plot for PC1 describes which particles contribute to the differences between the samples associated with Fiocchi and Wolf ammunition. A decrease in Sb,Ba particles and an increase in Pb, Sb, Ba particles in the Fiocchi associated samples compared to the Wolf are shown to be the main contributors (figure 14). Pb only; Ba only; Sb only; Pb,Sb and Pb,Ba particles show little difference in levels between the Fiocchi and Wolf associated samples being close to 0 on the loadings plot.



Figure 15: Loadings plot for PC1. This clearly shows that the cause of the difference between Fiocchi and Wolf associated samples is a relative lack of Sb,Ba particles and relative increase in Pb,Ba,Sb particles in the Fiocchi associated samples.

There were no consistent trends for PC2 (vertical axis figure 14) or PC3 (horizontal axis figure 15) when comparing the manufacture of the ammunition (Wolf vs Fiocchi) or origin of sample (cartridge case, gun barrel swab or target)

Finally, PC4 (vertical) in the second scores plot (figure 16) accounts for less than 1% of variation, but shows a weak cluster of cartridge case swabs, targets, and barrel swabs, thus indicating that the origin of the sample may have a very minor effect on composition.



Figure 16: Scores plot for variation in particle composition showing PC3 on the x axis and PC 4 on the y axis. A weak effect can be seen by clustering of cartridge case samples (circled red), target samples (circled green) and barrel swabs (circled orange) in the P4 axis. T1WF foul = sample from target following firing 1 round of Wolf after 3 rounds of Fiocchi; SD3F1W = sample from barrel following firing 1 round of Wolf after 3 rounds of Fiocchi; SCWc = sample from Wolf cartridge case; SCFc = sample from Fiocchi cartridge case; T3F1.5 = sample from target after firing Fiocchi; SDF = sample from barrel after firing Fiocchi. The software used was 'The Unscrambler' by CAMO software.

3.2.2 Three component GSR particle sizing; study 2

This study allowed the recovery of 167 three component GSR particles. Table 3

displays the varying particle sizes of those recovered particles from the two swabs

and two targets taken.

Table 3: A table showing the measurement of particles found on the four samples

from swabs and targets. Measurement of each particle is shown in μ m.

GSR Particle Size (µm)	discharge three rounds of Fiocchi, swab	discharge three rounds Fiocchi, followed by one round Wolf, swab	three rounds Fiocchi discharged from 1.5 m, target	one round Wolf from Fiocchi fouled barrel, target
0-6.99	15	8	31	17
7-12.99	29	20	17	4
13-18.99	13	6	2	0
19-24.99	2	1	1	0
25-29.99+	0	1	0	0
Mean particle size (µm)	10.32	10.35	7.40	6.10
Smallest particle size (µm)	4.09	3.80	3.18	3.72
Largest particle size (µm)	24.89	34.06	23.94	12.18
Standard Deviation (σ)	4.31	5.71	3.69	2.00

From table 3 it can be seen that the particles analysed from the barrel swabs have a greater size on average in comparison to the particles analysed from the targets, meaning that fewer of the larger particles travelled a far enough distance to have met the targets. Both size and air resistance effect how far the particles travelled. The

table also shows that there is no clear difference in particle size between the Fiocchi ammunition and the Wolf ammunition.

3.2.3 Study two discussion

The differing ammunition type study carried out for this research was designed to see if it is possible to distinguish between two different types of ammunition when the same gun is used. The two types of ammunition used in this study were Italian manufactured Fiocchi 9 mm Parabellum calibre rounds of ammunition and Russian manufactured Wolf 9 mm Parabellum calibre rounds of ammunition. They were used with the same 9 mm Parabellum calibre Steyr M9 semi-automatic pistol.

The results show that both the Fiocchi and Wolf ammunition produced Pb, Sb, Ba particles with no additional aluminium, tin or calcium/silicon meaning they are within the Type 1 classification bracket for GSR (White, 2016). Due to both types of ammunition being within the same classification they are almost indistinguishable from each other. However, from the results in figures 12 and 13 it can be seen that the Fiocchi ammunition contains relatively more three-component GSR and fewer Sb,Ba two component particles per 100 particles than the Wolf ammunition. The statistical analysis shows that if enough particles are recovered, it may be possible to distinguish between the GSR of two ammunitions within the same classification by considering the whole population of particles rather than just the three component particles. However, it should be noted that in general casework the numbers of GSR particles found may not be enough to perform reliable analysis. While the distinction is clear looking at 100 particle samples, it would be rare to find so many particles on a suspect. Testing carried out by Charles, S. & Geusens, N. (2011) was completed 5

minutes after a simulated arrest, typically in a real case the crime scene investigators who would carry out the sampling would do so 1-2 hours after arrest. This time period would lead to loss of a high number of GSR particles.

Schwoeble and Exline (2000) argue that it is not possible to match GSR to a specific manufacturer. Among other reasons they point out that it is possible that a manufacturer may buy in primers from different sources on occasion leading to variations. While this is true, in casework where GSR is being compared to a swab from a gun or a cartridge case, it has been long established that GSR can be differentiated according to the types listed in table 1 (Shaw, 2020). These results suggest that further discrimination may be possible within those types if the numbers of particles are large enough and if 2-component particles are included in the analysis.

The particle size analysis shows no clear distinction between the GSR produced by the Wolf and Fiocchi ammunition. Perhaps this is unsurprising given both are 9mm parabellum so one would expect similar amounts of primer in the same calibre ammunition. Therefore the GSR is being produced under similar conditions.



Figure 17: Photograph showing a target after three rounds of Fiocchi ammunition were fired directly at said target, sent to Kingston University by the Garda Siochana.

The sample taken from the fouled barrel of the gun and the samples of the target after a shot had been fired from the foul barrel investigated the capacity to identify the ammunition of the last shot fired from a gun, despite it having been previously used with ammunition from another manufacturer. Some carry over of the GSR remaining in the gun barrel from previous shots would be expected to be found, but it was not known how much of a factor this would be. A swab directly from the barrel was taken alongside a paper target at which the shots had been fired. The last shot fired was Wolf ammunition while the gun barrel had Fiocchi GSR from previous shots in it. The swab sample has a higher presence of antimony and barium in comparison to the target sample. However, both Wolf ammunition samples still have a much higher presence of all Sb and Ba particles and much lower Pb, Sb, Ba particles in comparison to the Fiocchi barrel swab and the Fiocchi target sample. Despite the similarity in the classification type according to Shaw (2020) of the ammunition used in this study, it is possible to tell that Wolf was the ammunition fired last even though it was being fired from a fouled barrel. This is due to the high presence of Ba,Sb particles and it is possible to speculate that the Wolf ammunition contains more antimony sulphide or lower levels of lead compounds, which would explain the difference between the Fiocchi and Wolf ammunition. This chemical variation would provide a possible explanation for the ability to differentiate between the two ammunition types. The results indicate that the majority of the GSR deposited in the gun barrel and on the target when firing a gun comes from the ammunition rather than old GSR that has remained in the barrel from previous shots. In order to further clarify the amount of GSR originating from the previous fouling of the gun barrel, it would be better to repeat the experiment with more clearly differentiated ammunition types from the classification scheme, for example one with additional tin or aluminium that was not present in the other.

Each sample from the differing ammunition type study had 100 random particles recovered. This was due to the high volume of particles present. It is possible that this affects the type of particle analysed due to unconscious bias in the selection of particles from which to obtain spectra. The Garda Siochana ballistic team performed the firearm discharges for this research and all evidence was packaged, sealed, and delivered to Kingston University as FSR guidelines for maintenance of exhibit continuity.

However, if this study was to be repeated it would be advised to have a comparison of just Wolf ammunition versus the Fiocchi ammunition, as well as firing the Wolf ammunition three times before the Fiocchi ammunition to see if there is a true effect. It would also be advised to test for antimony sulphide in both ammunitions to confirm its presence. Repeats of the study would provide more results and allow for any patterns or trends to be seen, this could further support the reliability of the studies. The use of an automated SEM would improve the particle selection as it would remove any unconscious bias. Additionally, it would be advised to not outsource evidence but to have it completed on site, to control and minimise the potential for contamination, and to confirm that all studies were carried out in the correct manner. However, all control samples from the studies carried out by the Garda Siochana were clear and contained no particles so there is no concern about contamination.

3.3 Particle analysis; study 3

Three particles were recovered from the control target which was not fired at. Despite precautions this shows a low level of contamination on the firing range as could be expected when carrying out such studies in areas where very large numbers of rounds have been fired. The finding of a relatively low level of contamination will not significantly affect the findings because much larger populations of particles were recovered.



Figure 18: Graph showing the total number of one and two component particles analysed in study three Vs the distance at which they were fired. Graph includes results from all four ammunition types, 9 mm, .22 " LR, .223 " and 12-gauge.



Figure 19: Graph showing the total number of three-component GSR particles analysed in study three Vs the distance at which they were fired. Graph includes results from all four ammunition types, 9 mm, .22 " LR, .223 " and 12-gauge.

3.3.1 GSR particle analysis, number, and type; study 3

The expectation for these results is that there should be a reduction in the number of particles found the further away the target is. Therefore, the targets at 10-metres should show the least number of particles. There is an expectation that the particles will start to reduce, and the numbers will drop before the 3-metre target (White, 2016). This study will help determine whether the calibre of a firearm and ammunition affects the distance GSR travels. It is expected that the shotgun would project GSR further due to its higher power ammunition. Projectiles and particles emitted from the muzzle of a gun will have gained kinetic energy from the high pressure in the barrel caused by the burning of the propellant. This results in them leaving the muzzle with high velocity. Air resistance acts as a force against their progress due to collisions between particles in the air and those coming from the gun. The law of conservation of momentum (momentum = mass x velocity) means that it will take many more collisions with the relatively low mass air particles to reduce the velocity of a high mass projectile fired from a gun, compared to reducing the velocity of a small, low mass GSR particle. It can be seen that a bullet or pellet fired from a gun travel much further than the smoke it emits from the barrel, and it would be expected that inorganic GSR particles, due to their small size and hence low mass, would similarly only travel a few metres. Larger GSR particles of a higher mass would potentially go further than those of lower mass. However, other factors than momentum also play a part, such as particle size, morphology (aerodynamics) and collisions between GSR particles. Maitre et al. (2021) carried out a study on pellet distribution. They found that as the distance of flight of the pellets increase the pellets spread further from each other. There are phases which the pellets go through at the time of firing, phase 1; where the pellets are enclosed in the shotgun

wad as it exits the firearm, phase 2; where the wad is affected by air resistance and begins to open and finally phase 3; where the pellets begin to disperse at approximately 1.5 m. Maitre *et al.* (2004) state that as the distance of the pellets from the shotgun increase the pellets spread further apart, creating a pellet distribution area.

Overall, the results for three component GSR (figure 19) drop off from around the 20+ level to less than 10 when a range of 2 metres is exceeded. This follows the expected trend but the results for the 12-gauge shotgun are a notable exception. It is likely that the extensive damage caused to the targets by this firearm, especially at close range, has affected the recovery of GSR particles. The results for one and two component particles (figure 18) are less clear and show some unexpected variations contrary to expectations, particularly with the shotgun, but also the 9 mm calibre at 3 metres.

The 9 mm parabellum ammunition produced a high amount of PbSb particles, these amounts were at their highest on the 2 metre and 3 metres targets with totals of 43 followed by 248. Pb particles were also found to have higher levels at the first two targets, 2 and 3 metres, with totals of 22 followed by 69. From all the particles recovered from the targets the totals did follow expected hypothesis that GSR will travel a consistent distance regardless of firearm and ammunition used and begin to drop off at the 3-metre point. See appendix a: results, table Q and figure a13-a15.

The .22 " LR ammunition produced high levels of Pb particles, 157 of which were recovered from the 2-metre target, the Pb particles did decrease down at the 10-metre point but was lowest at the 3-metre target. PbBa particles were also common

from this ammunition, 44 particles were recovered from the 2-metre target, and they decreased, again the lowest amount recovered was at the 3-metre target. In total 23 PbSbBa particles were recovered from the 2-metre target and these particles rapidly decreased down to 0 at the 10-metre target. See appendix a: results, table R and figure a16-a18.

The .223 " REM calibre ammunition had lower amounts of particles recovered, however overall, still followed the hypothesis that the GSR particles will travel a consistent distance regardless of which firearm or ammunition is used, also that particles will be at their lowest at the 10-metre target. Ba particles appeared the most at the 2-metre target, followed by PbBa and PbSb particles, which totalled 19 and 26 particles at the 2-metre target. In total 19 PbSbBa particles were recovered from the 2-metre target and then the particles decrease the further the distance to 5 particles at 3 metres and one particle at 4 and 5 metres. See appendix a: results, table S and figure a19-20

There were unexpected variations of particle numbers from the 12-guage ammunition. The PbSb particle numbers were much higher than any threecomponent GSR particles, 160 particles alone were recovered from the 2-metre target. The Pb particles are also very high, the highest number of particles collected were from the 2-metre target at 225, which could be originating from the lead pellets from the shotgun cartridge hitting the targets at the time of discharge. See appendix a: results, table T and figures a21-22.





Figure 20(a-e): Image (a) showing the 2 metre and image (b) 3 metre targets of the 12-gauge shotgun ammunition by Eley. Image (c) showing the 4 metre and image (d) 5 metre targets of the 12-gauge shotgun ammunition by Eley. Image (e) showing the 10-metre target of the 12gauge shotgun ammunition by Eley.

3.3.2 Three component GSR particle sizing; study 3

Tables 4-7 show the numbers of particles of different sizes detected at varying distances from firearm to target. They correspond to 9 mm, .22 ", .223 " and 12-gauge calibres respectively. Figure 21 shows a plot of particle size against distance travelled for the different calibres.
Table 4: A table showing the measurement of particles found on the samples from

the 9 mm para-ammunition by Fiocchi targets, and the distance at which the

ammunition was fired.

9 mm para-a	9 mm para-ammunition by Fiocchi				
GSR	2 metres	3 metres	4 metres	5 metres	10 metres
Particle					
Size (µm)					
0-6.99	5	0	0	0	0
7-12.99	9	0	4	4	3
13-18.99	4	0	1	2	0
19-24.99	2	0	0	0	0
25-29.99+	2	0	0	0	0
	-			-	
Average	12.52	N/A	10.65	10.54	9.45
particle					
size (µm)					
Smallest	3.85	N/A	7.71	7.12	7.85
particle					
size (µm)					
Largest	29.15	N/A	16.55	14.46	10.87
particle					
size (µm)					
Standard	7.31	N/A	3.69	3.01	1.52
Deviation					
(σ)					

Table 5: A table showing the measurement of particles found on the samples from

 the .22 " LR calibre ammunition by Winchester targets, and the distance at which the

 ammunition was fired.

.22 " LR cali	bre ammuniti	ion by Winch	ester		
GSR	2 metres	3 metres	4 metres	5 metres	10 metres
Particle					
Size (µm)					
0.0.00	2		0	4	0
0-6.99	3	2	U	1	0
7-12.99	15	4	1	0	0
13-18.99	4	0	0	1	0
19-24.99	0	0	0	0	0
25-29.99+	0	0	0	0	0
				-	-
Average particle size (um)	9.83	7.96	7.67	10.10	N/A
Size (µm)					
Smallest particle size (µm)	6.20	6.44	7.67	5.82	N/A
Largest particle size (µm)	17.80	9.94	7.67	14.37	N/A
Standard Deviation (σ)	2.75	1.33	0	6.04	N/A

Table 6: A table showing the measurement of particles found on the samples from

 the .223 " rem calibre ammunition by Hornady targets, and the distance at which the

 ammunition was fired.

.223 " rem ca	.223 " rem calibre ammunition by Hornady				
GSR	2 metres	3 metres	4 metres	5 metres	10 metres
Particle				away	
Size (µm)				-	
0.0.00		0	0		
0-6.99	1	0	0	0	0
7-12.99	8	2	1	1	0
13-18.99	4	1	0	0	0
19-24.99	1	0	0	0	0
25-29.99+	5	2	0	0	0
Average	17.27	19.92	7.72	12.01	N/A
particle					
size (µm)					
Smallest	4.03	7.97	7.72	12.01	N/A
particle					
size (µm)					
Largest	37 15	30.79	7 72	12.01	Ν/Δ
narticle	57.15	50.75	1.12	12.01	
sizo (um)					
5126 (µ11)					
Standard	10.48	10.35	0	0	N/A
Deviation					
(σ)					
				1	

Table 7: A table showing the measurement of particles found on the samples from

 the 12-Gauge shotgun ammunition by Eley targets, and the distance at which the

 ammunition was fired

12-gauge sh	otgun ammu	nition by Eley	1	12-gauge shotgun ammunition by Eley				
GSR	2 metres	3 metres	4 metres	5 metres	10 metres			
Particle								
Size (µm)								
0.0.00		-			0			
0-6.99	1	5	0	0	2			
7-12.99	0	2	5	0	2			
13-18.99	0	0	6	1	0			
19-24.99	0	0	0	0	0			
25-29.99+	1	0	0	0	0			
Average	74.77	6.39	12.78	13.17	7.45			
particle size (µm)								
Smallest	3.15	3.10	9.21	13.17	5.15			
particle								
Size (µIII)								
Largest	146.40	12.83	16.94	13.17	9.30			
particle								
size (µm)								
Standard	101.20	3.48	2.86	0	1.93			
Deviation (σ)								
(-)								

Tables 4-7 show a variation in the particle sizes for each of the ammunitions used in the study. The GSR from the .22 " LR ammunition had the smallest average particle sizes overall and the 12-gauge ammunition had the largest average particle sizes. Though from table 7, the averages in this case were high due to larger particles travelling a shorter distance, bringing up the overall average. Both the .22 " LR and the .223 " ammunition had no particles found at the farthest distance (10 m), compared to the 9 mm and 12-gauge who have particles of a fairly similar size at the largest distance 10.87 μ m from the 9 mm and 9.303 μ m from the 12-gauge.



Figure 21: A chart showing the average GSR particle size of those recovered from study three VS the distance from the target at which they were fired. Graph includes results from all four ammunition types, 9 mm, .22 " LR, .223 " and 12-gauge.

3.3.3 Study three discussion

The distance and dispersion study was designed to see how far three-component GSR particles will travel depending on the type of firearm used. The hypothesis for this study was that most GSR particles will travel a few metres due to velocity and mass but would start to drop off and not be present on the targets further away (White, 2016), A study carried out by De Forest *et al.* (2004) found that after testing GSR from eight separate sources the average velocity was 500-600 ft per second, though some particles in the testing had higher velocities. The evidence found showed that particles experience rapid rates of deceleration which explained why

fewer particles were found at the larger distances. The results closely support this hypothesis in three of the ammunition types used, those being; 9 mm coupled with the Steyr M9 semi-automatic pistol, this pistol was designed in 1999, the idea behind the design was to create a firearm that was easy to carry and conceal. The M9 uses 9x19 mm or 9x21 mm cartridges, the muzzle velocity for this firearm is 300 – 460 m/s. The .22 " LR coupled with .22 " Long rifle Winchester lever action, this firearm was designed and produced in 1972 under model 9422 before being discontinued in 2005, the muzzle velocity for this firearm is 341-346 m/s. Standard models used .22 short, .22 long and .22 long rifle ammunition, it was designed to be a reliable and easy firearm to use. Followed by the .223 " coupled with .223" Remington calibre bolt action rifle, this firearm was sold on the market from 1960 onwards and is a smallbore rifle originating from the Czech Republic, the muzzle velocity for this firearm is 955 m/s. There are several different calibres for this firearm, the standard being .17 Remington, .204 Remington and .223 Remington. Finally the 12-gauge shotgun ammunition by Eley couple with a 12-gauge Winchester semi-automatic Shotgun. The muzzle velocity for this firearm is 1,098 m/s.

The highest number of three-component GSR particles found with these ammunition types were found within the first 2 metres and steadily drop off the further away the targets are, between 3 and 10 metres. There is of course potential for low levels to be found at greater distances as has been reported in other studies (Fojtasek *et al.*, 2003; Gerard *et al.*, 2011). There were several GSR particles found between 4 and 10 metres in agreement with Brozek-Mucha (2009) and Fojtasek *et al.* (2003). Because these are relatively low levels there could be several reasons for this like contamination in the SEM stub box, contamination when at the firing range at the

time of target collection or contamination during sampling. However, it is quite possible that a smaller number of particles do travel over this range as in the previous studies. The possibility of GSR travelling on the projectile must also be considered (Blakey *et al.* 2018; Gerard *et al.* 2011) though samples from targets avoided bullet holes in this study.

The results from the 12-gauge shotgun vary in comparison to the other ammunition used. There was an expectation that the GSR may travel further because the larger amounts of propellent in the ammunition may produce higher velocity GSR particles leaving the muzzle, due to higher combustion pressure. The plume distributions of different firearms had previously been studied by Schwoeble and Exline (2000) but they did not study the GSR on targets. With the shotgun ammunition in this study, additional aluminium was present in the three component particles. The presence of additional aluminium as a fuel in the primer affected the range of particle compositions produced with large numbers of Ba, AI particles formed (White, 2016), so numbers of characteristic lead-barium-antimony particles may be correspondingly reduced. GSR with additional aluminium was observed to condense into significant numbers of Ba. Al and Pb. Sb particles which may have led to a reduced number of Pb, Ba, Sb, Al particles where all key components (Pb, Sb, Al) are present, this could be the reason for the lower numbers of particles found in the 3-component category. The results in table T show that the 12-gauge contained a higher level of Pb, Sb and Sb, Ba particles and a much higher lead count than any of the other ammunitions used. The likely reason for the lead count being so high could be due to the lead from the pellets within the ammunition transferring to the targets. This can be compared to the findings of Gerard *et al.* (2011) of GSR at much greater distances and it being associated with the projectiles. The difference between jacketed and

unjacketed projectiles is therefore a factor. Another potential reason for this could be due to the shotgun wadding. At short distances this wadding would exit the firearm at speed due to velocity and mass and could hit the targets which would have an unpredictable effect on particle numbers. The excessive damage to targets caused by the shotgun pellets could also be a major factor as to why the results are not clear. As can be seen in figure 20e the photo of the 12-gauge shotgun target at 10 metres clearly shows how much of the ammunition and excess pellets has torn through the target, making it a much harder area to sample. Overall, the shotgun results do not provide a reliable pattern but, this research does give a suggestion that larger numbers of shotgun GSR particles may travel further, due to the higher combustion pressure occurring at the time of firing. According to Boltzmann distribution as molecules become smaller the velocity increases, when gunshot residue is expelled, it experiences high pressure and extreme temperatures, to stabilise themselves the particles form into as small a surface as they can and solidy when cooled, which creates microstructures (Brozek-Mucha, 2007; Kara, İ. 2022). However damaged targets, shotgun wadding, and damaged target sampling could have all affected these samples.

Four different firearms were used for this study and were as follows, .22 " Long Rifle calibre Winchester lever action rifle, 9 mm Parabellum calibre Steyr M9 semiautomatic pistol, .223" Remington calibre CZ 527 bolt action rifle and a 12-gauge Winchester semi-automatic shotgun. The firearms used all vary in certain ways, pistols are small, lightweight, and preferred for close-range shooting, these are favoured by law enforcement. Rifles are a larger firearm which have space for a higher quantity of ammunition. They provide better accuracy and are often used for

target practice. Finally, shotguns, these are the most powerful out of the firearms used. Shotguns use shells rather than single bullets. Shotgun shells contain small pellets which when shot spread out over a large area. Shotguns are favoured typically for hunting.

The chart in figure 21 shows the size of GSR particles against the distance they travelled for each ammunition type alongside the standard deviation, shown as error bars. There was one very large particle from the 12-gauge shotgun on the 2-metre target that skews the average. Overall, there does not appear to be a significant change in size of particles over distance travelled. This compares with Gerard *et al.* (2011) finding fewer large particles at greater distances and Brozek-Mucha (2009) finding more, though the latter was examining ranges up to only 1 metre.

The results from the control sample show that three Pb particles were found on the target. This could mean that contamination occurred either at the firing range, during sampling or with the SEM stub box however, measures in place including separate people retrieving the target to the person who fired the gun, extensive cleaning when sampling took place and the stub being kept separately from any other contaminated stub were all preventative and no other measures could have been implemented to stop this from happening. It must be accepted that low levels of contamination are almost impossible to entirely rule out, as recognised in the way GSR results are reported in casework (White, 2016)

Chapter 4: Conclusions

and Future Research

4.1 Conclusions

This research investigated if there are alternate explanations for GSR being transferred in the general environment other than direct involvement with firearms. It is important to consider the source of any GSR evidence, as interpretation of whether a firearm has been discharged by an individual, if they have encountered a firearm user or if they have merely been in the vicinity at the time of a firearm discharge is critical in court (Blakey *et al.* 2018). The three studies carried out relate to the transfer, deposition, and identification of GSR and its recovery on individuals. The results from each of the studies provide their own information on whether GSR can be recovered and matched to its source which could help with any future research into the topic. There is currently not enough published data on background levels of GSR in the environment or the transfer from the environment to a subject, so the expert is left to concede that they cannot rule out a single particle or low level having come from adventitious contamination in the general environment (White, 2016).

The transfer study, study one, was designed as a step in assessing how realistic that possibility is, given the most likely area to be contaminated is a person's clothing and the most likely legitimate source, excluding arrest by armed police officers, is from a gun club member. The aim of this study was to determine the extent of GSR transfer to clothing after contact had been made between a doner and a recipient. Overall, only relatively little three component GSR was found to be transferred, a total of 39 particles over the five t-shirts tested, when the short timescales involved are considered. Therefore, this study found transfer from a gun club member an improbable though not a completely inconceivable source of high levels of GSR contamination. However, the hypothesis stated that following close contact between

the donor and recipient, high levels of one, two and three component GSR particles would be detected on the donor's clothing, though the 39 particles found are not a small amount, given the time scale between firing and the transfer it would be expected to find higher levels of GSR.

The second study demonstrated that superficially similar GSR populations could be distinguished and matched to different ammunition sources by considering the whole population of particles rather than just the three component GSR. This shows potential for identifying the source of GSR given sufficient numbers of particles are found and a control sample such as a cartridge case or gun barrel is available. However, this is not always the evidence that can be retrieved, but an ideal to what information could be looked at if the evidence was present. The aim for the second study was to distinguish between two differing ammunition types when both had been discharged from a 9 mm parabellum calibre Steyr M9 semi-automatic pistol, and to determine which ammunition had been fired last. The results from this study show that it is possible to discern between the two ammunition types used. While it was possible to discern between the two ammunition types, in general case work this may not be possible due to the smaller amount of GSR that is likely to be available in comparison to the 100 particles examined within this study. The second part of this study was to determine which ammunition had been fired last, it was possible to do so due to the higher levels of antimony sulphide in the Wolf ammunition along with lower levels of lead compounds. This chemical variation is where the main difference between the Wolf and Fiocchi ammunition is. The third study tested how close a person must be to the discharge of a firearm to become contaminated with GSR. The aim was to determine whether or not the calibre of firearm and ammunition time effects the distance the GSR will travel. The

study found that while a few particles may travel further, the majority of three component GSR, 73% of the particles identified in this study, will only travel less than 3 metres, in agreement with current interpretation (White, 2016). However, shotguns may be an important exception to this generalisation as 63% of the three component GSR particles from the shotgun travelled further than 3 metres.

The identification of GSR particles originating from firearms rather than other environmental sources has become a significant issue for the presentation of GSR evidence in the criminal justice system. In particular, attributing an explanation of the origin and deposition of the particles remains contentious. The findings of this study indicate that GSR deposited from a shotgun can be distributed over a wider area than that of a handgun. In addition, the risk of GSR deposition on the clothing of an individual who had not previously had direct contact or been in the vicinity of a firearm incident is possible although it requires significant physical contact or interaction. Furthermore, differentiation and attribution of different types of ammunition to specific GSR particles has been achieved in a limited number of experimental models. These findings all contribute further knowledge to the research of GSR identification, persistence and distribution, and provide invaluable insight for casework examiners in assessing the origin and mechanism of GSR deposition.

4.2 Future Research

This research was carried out on a small scale therefore it would be recommended that further research is carried out on a larger scale and repeated to gain a better understanding of particle population. As can be seen from the transfer study, even when closely repeating a firing and transfer scenario, a wide range of number of

GSR particles was recovered so more repeats are necessary to increase the statistical significance. Ideally if the studies carried out here were repeated there would also be an environmental GSR study alongside, as this would allow better understanding of the standard amount of GSR in areas of the general environment, i.e., public transportation, vehicles and publicly shared locations like shops and seating which allows a 'base' level to be set. It would also be useful to know how much GSR was on the donor, to again set a base level for the transfer, this way it could be seen if significant transfer had taken place.

Another improvement in analysis for the first and third studies would be to use an SEM with an attached automated system. The SEM used for this research did not have an automated system which leaves room for human error, particles could easily be missed even when being meticulous.

Significant results could be gained with more in-depth research and larger sample groups. Future court cases could benefit from more knowledge surrounding twocomponent GSR particles alongside three-component GSR particles. Results which include the whole population of particles can completely change how they are interpreted, though those results should not be used to say that someone has fired a gun. Certain occupations like construction workers have a high risk of having large numbers of metallic particles on them including particles which are indicative of GSR. Therefore, caution is also required when considering one and two-component particles (White, 2016).

For any further research into differing ammunition types, it would be advised to try multiple different ammunition types to see if this is true across all classifications of GSR type. For any further research carried out on the topic of distance and dispersion it would be advisable to set up secondary targets surrounding the initial target so that the full spread of GSR can be sampled, it would allow the researcher to confirm GSR travels without dealing with the issues faced in this research regarding the shotgun studies. Alongside different ammunition types further research into differing particle shape and size could be carried out, there is a typical standard when looking at GSR evidence of spherical particles of a similar average size, this is not always necessarily the case for every GSR particle however and if an automated system is set to recognise certain attributes then important data may be missed.

There is room for improvement where research methods are concerned, very few methods are used for researching OGSR and a global standard to carry out such research has not yet been identified. The materials used when investigating the evidence are typically carbon coat stubs as they have superior retention in comparison to other materials like liquids, further research to test out different sampling materials that may be quicker and more efficient in the field could be considered.

There is also a lot of value at looking at the whole population of particles recovered from a sample, specifically two-component particles, even with primary or secondary transfer samples. Court cases will mostly focus on three-component particles, but they aren't the only particles that should be considered.

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Appendix A: Results

Table A: A table showing the number of one and two component particles recovered

 alongside any three-component GSR particles in total from the four samples taken

 from the recipients t-shirt, t-shirt one.

Sample	Total number of one and two component particles analysed	Total number of three- component GSR particles analysed
T-shirt one, front left	49	2
T-shirt one, front right	12	0
T-shirt one, back left	0	0
T-shirt one, back right	49	0

Table B: A table showing the number of one and two component particles recovered

alongside any three-component GSR particles in total from the four samples taken

from t-shirt two.

Sample	Total number of one and two component particles analysed	Total number of three- component GSR particles analysed
T-shirt two, front left	36	0
T-shirt two, front right	3	0
T-shirt two, back left	48	2
T-shirt two, back right	74	5

Table C: A table showing the number of one and two component particles recovered

in total from the four samples taken from t-shirt three.

Sample	Total number of one and two component particles analysed	Total number of three- component GSR particles analysed
T-shirt three, front left	68	5
T-shirt three, front right	19	2
T-shirt three, back left	37	2
T-shirt three, back right	46	0

Table D: A table showing the number of one and two component particles recovered

Sample	Total number of one and two component particles analysed	Total number of three- component GSR particles analysed
T-shirt four, front left	54	1
T-shirt four, front right	20	3
T-shirt four, back left	56	1
T-shirt four, back right	158	2

in total from the four samples taken from t-shirt four.

Table E: A table showing the number of one and two component particles recovered

in total from the four samples taken from t-shirt five.

Sample	Total number of one and two component particles analysed	Total number of three- component GSR particles analysed
T-shirt five, front left	75	7
T-shirt five, front right	44	1
T-shirt five, back left	82	3
T-shirt five, back right	87	3

Table F: A table showing the number and type of one, two and three-component

GSR particles recovered from t-shirt one.

	T-Shirt one, number and type of particles				
Particle Type	T-shirt one, front left	T-shirt one, front right	T-shirt one, back left	T-shirt one, back right	
Pb	29	2	0	29	
Sb	0	0	0	0	
Ва	9	0	0	0	
Pb, Sb	6	0	0	4	
Pb, Ba	4	1	0	15	
Sb, Ba	1	0	0	1	
Pb, Sb, Ba	2	0	0	0	

Table G: A table showing the number and type of one, two and three-component GSR particles recovered from t-shirt two.

	T-Shirt two, number and type of particles				
Particle Type	T-shirt two, front left	T-shirt two, front right	T-shirt two, back left	T-shirt two, back right	
Pb	20	1	25	25	
Sb	1	0	0	0	
Ва	1	0	4	7	
Pb, Sb	10	1	11	12	
Pb, Ba	4	1	6	25	
Sb, Ba	0	0	0	0	
Pb, Sb, Ba	0	0	2	5	

Table H: A table showing the number and type of one, two and three-component

GSR particles recovered from t-shirt three.

	T-Shirt three, number and type of particles				
Particle Type	T-shirt three, front left	T-shirt three front right	T-shirt three, back left	T-shirt three, back right	
Pb	38	11	25	26	
Sb	0	0	0	1	
Ва	7	1	2	1	
Pb, Sb	10	4	5	4	
Pb, Ba	8	1	3	14	
Sb, Ba	0	0	0	0	
Pb, Sb, Ba	5	2	2	0	

Table I: A table showing the number and type of one, two and three-componentGSR particles recovered from t-shirt four.

	T-Shirt four, number and type of particles					
Particle Type	T-shirt four, front left	T-shirt four, front right	T-shirt four, back left	T-shirt four, back right		
Pb	27	7	35	77		
Sb	0	0	0	0		
Ва	3	3	2	12		
Pb, Sb	10	2	6	32		
Pb, Ba	13	5	12	35		
Sb, Ba	0	0	0	0		
Pb, Sb, Ba	1	3	1	2		

Table J: A table showing the number and type of one, two and three-component

GSR particles	recovered	from	t-shirt five.
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	T-Shirt five, number and type of particles					
Particle Type	T-shirt five, front left	T-shirt five, front right	T-shirt five, back left	T-shirt five, back right		
Pb	39	22	35	33		
Sb	0	0	0	1		
Ва	4	4	3	4		
Pb, Sb	11	7	12	15		
Pb, Ba	14	10	29	31		
Sb, Ba	0	0	0	0		
Pb, Sb, Ba	7	1	3	3		

Table K: A table showing the number of particles recovered in total from the swabs

and targets of control ammunition samples.

Sample	Total number of all particles analysed	Total number of three-component GSR particles analysed
bench control, swab	0	0
gun barrel control sample, swab	0	0
control Fiocchi cartridge, swab	100	52
control Wolf cartridge, swab	99	38
discharge three rounds of Fiocchi, swab	100	59
discharge three rounds Fiocchi, followed by one rounds Wolf, swab	100	36
three rounds Fiocchi discharged from 1.5 m, target	100	51
One round Wolf from Fiocchi fouled barrel, target	100	21

Table L: A table showing the number and type of one, two and three GSR particles

recovered from the swabs and targets of control ammunition samples.

	Study 2, n	umber, and	l type of part	icles		
Particle Type	swab, Control Fiocchi cartridge	swab, Control Wolf cartridge	swab, Discharge 3x rounds of Fiocchi	swab, discharge 3x rounds Fiocchi, followed by 1x round Wolf	target, 3x rounds Fiocchi discharged from 1.5m	target, 1x round wolf from Fiocchi fouled barrel
Pb	0	3	0	1	9	3
Sb	1	4	2	2	0	0
Ва	6	7	8	6	5	4
Pb, Sb	1	5	3	4	11	5
Pb, Ba	1	5	2	4	1	1
Sb, Ba	39	37	27	47	23	66
Pb, Sb, Ba	52	38	59	36	51	21

Table M: A table showing the total number of particles recovered from the 9 mm

Sample	Total number of one and two component particles analysed	Total number of three- component GSR particles analysed
9 mm para calibre ammunition	99	22
by Fiocchi at 2 m		
9 mm para calibre ammunition	321	0
by Fiocchi at 3 m		
9 mm para calibre ammunition	45	5
by Fiocchi at 4 m		
9 mm para calibre ammunition	28	6
by Fiocchi at 5 m		
9 mm para calibre ammunition	10	3
by Fiocchi at 10 m		

para-ammunition by Fiocchi.

Table N: A table showing the total number of particles recovered from the .22 " LR

calibre ammunition by Winchester.

Sample	Total number of one and two component particles analysed	Total number of three- component GSR particles analysed
.22 " LR calibre ammunition by Winchester at 2 m	248	23
.22 " LR calibre ammunition by Winchester at 3 m	44	6
.22 " LR calibre ammunition by Winchester at 4 m	20	1
.22 " LR calibre ammunition by Winchester at 5 m	51	2
.22 " LR calibre ammunition by Winchester at 10 m	24	0

Table O: A table showing the total number of particles recovered from the .223 " rem

Sample	Total number of one and two component particles analysed	Total number of three- component GSR particles analysed
.223 " rem ammunition by	133	19
Hornady at 2 m		
.223 " rem ammunition by	43	5
Hornady at 3 m		
.223 " rem ammunition by	26	1
Hornady at 4 m		
.223 " rem ammunition by	49	1
Hornady at 5 m		
.223 " rem ammunition by	20	0
Hornady at 10 m		

calibre ammunition by Hornady.

Table P: A table showing the total number of particles recovered from the 12-gauge

shotgun ammunition by Eley.

Sample	Total number of one and two component particles analysed	Total number of three-component GSR particles analysed
12-gauge shotgun ammunition by Eley at 2 m	392	1
12-gauge shotgun ammunition by Eley at 3 m	233	8
12-gauge shotgun ammunition by Eley at 4 m	111	10
12-gauge shotgun ammunition by Eley at 5 m	185	1
12-gauge shotgun ammunition by Eley at 10 m	157	4

Table Q: A table showing the number and type of three-component GSR particles

	Number and type of 9 mm para-ammunition particles					
Particle Type	shot from 2 metres	shot from 3 metres	shot from 4 metres	shot from 5 metres	shot from 10 metres	
	away	away	away	away	away	
Pb	22	69	18	9	2	
Sb	0	0	0	0	0	
Ва	4	3	3	5	2	
Pb, Sb	43	248	9	4	2	
Pb, Ba	6	1	6	0	0	
Sb, Ba	2	0	4	4	1	
Pb, Sb, Ba	22	0	5	6	3	

recovered from the 9 mm para-ammunition targets.

Table R: A table showing the number and type of three-component GSR particles

recovered from the .22 " LR calibre ammunition targets.

	Number and type of .22 " LR ammunition particles					
Particle Type	shot from 2 metres away	shot from 3 metres away	shot from 4 metres away	shot from 5 metres away	shot from 10 metres away	
Pb	157	16	9	18	12	
Sb	0	0	1	2	0	
Ва	2	7	3	6	8	
Pb, Sb	12	4	2	12	3	
Pb, Ba	44	11	4	11	1	
Sb, Ba	1	0	0	0	0	
Pb, Sb, Ba	23	6	1	2	0	

Table S: A table showing the number and type of three-component GSR particles

 recovered from the .223 " rem calibre ammunition targets.

	Number and type of .223 " rem calibre ammunition particles					
Particle Type	shot from 2 metres away	shot from 3 metres away	shot from 4 metres away	shot from 5 metres away	shot from 10 metres away	
Pb	21	12	11	16	16	
Sb	0	0	0	3	0	
Ва	46	8	4	10	1	
Pb, Sb	19	11	7	9	1	
Pb, Ba	26	4	1	9	1	
Sb, Ba	2	3	2	1	1	
Pb, Sb, Ba	19	5	1	1	0	

Table T: A table showing the number and type of three-component GSR particles

recovered from the 12-gauge shotgun ammunition targets.

	Number and type of 12-gauge shotgun ammunition particles				
Particle Type	shot from 2 metres away	shot from 3 metres away	shot from 4 metres away	shot from 5 metres away	shot from 10 metres away
Pb	225	120	11	67	86
Sb	0	0	0	0	4
Ва	4	9	59	10	10
Pb, Sb	160	89	12	104	46
Pb, Ba	1	3	7	1	3
Sb, Ba	1	4	12	2	4
Pb, Sb, Ba	1	8	10	1	4





Figure a1: T-shirt two back left. A backscattered electron image showing a threecomponent GSR particle present on the back left of t-shirt two. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 μm



Figure a2: T-shirt two back left. EDX spectra showing a three-component GSR particle on the back left of t-shirt two, electron image shown in figure a1. Trace AI is present along with the Pb, Ba and Sb.





Figure a3: T-shirt three back left. Backscattered electron image showing a threecomponent GSR particle present on the back left of t-shirt three. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 μ m.



Figure a4: T-shirt three back left. EDX spectra showing a three-component GSR particle present on the back left of t-shirt three. Electron image of the particle is shown in figure a3. Pb, Ba and Sb are the only major peaks.




Figure a4: T-shirt four back right. Backscattered electron image showing a threecomponent GSR particle found on the back right of t-shirt four. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 µm.



Figure a6: T-shirt four back right. EDX spectra showing a three-component GSR particle present on the back right of t-shirt four. Electron image for this particle can be seen in figure a4. Trace Cu likely from the cartridge case can be seen along with the major Pb, Ba and Sb peaks.





Figure a7: T-shirt five front left. Backscattered electron image showing a threecomponent GSR particle present on the front left of t-shirt five. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 μm.



Figure a8: T-shirt five front left. EDX spectra showing a three-component GSR particle present on the front left of t-shirt five. Electron image for this particle can be seen in figure a7. Trace Cu likely from the cartridge case can be seen along with the major Pb, Ba and Sb peaks. The As and Co are mislabels as can be seen by there absence in some areas labelled.



Figure a9: Swab taken from barrel after 3x rounds of Fiocchi ammunition were discharged. Image contains background material from the swab. Backscattered electron image showing a three-component GSR particle measuring at 16.78 μm found on the swab. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 μm.



Figure a10: Swab taken from barrel after 3x rounds of Fiocchi ammunition were discharged. EDX spectra showing a three-component GSR particle found on the swab. Electron image can be seen in figure a9. Pb, Ba and Sb are the only major peaks present. There may be some S as expected when the Sb peak is high.



50 µm	EHT = 20.00 kV
	WD = 9.5 mm

Figure a11: Swab taken from barrel after 3x rounds of Fiocchi ammunition were discharged followed by 1x round of Wolf ammunition. Electron image showing a three-component GSR particle measuring at 18.59 μ m found on the swab. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 μ m.



Figure a12: Swab taken from barrel after 3x rounds of Fiocchi ammunition were discharged followed by 1x round of Wolf ammunition. Spectra showing a three-component GSR particle found on the swab. Electron image of this particle can be seen in figure a11. The major peaks are Pb, Ba and Sb with some S as expected given the high Sb peak. There is trace K which is not expected in GSR and may be from the background substrate.



50 µm	EHT = 20.00 k ⁴
	WD = 9.5 mm

Figure a13: 9 mm para-ammunition by Fiocchi shot from 2 metres away. Backscattered electron image showing a three-component GSR particle measuring at 10.02µm found on the sample. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 µm.

kV



Figure a14: 9 mm para-ammunition by Fiocchi shot from 2 metres away. EDX spectra showing a three-component GSR particle found on the sample. Electron image for this particle can be seen in figure a13. Pb, Ba and Sb are the major peaks. There may be some S as expected with the high Sb peak. As is a mislabel as can be seen by its absence in some labelled areas



Figure a15: 9 mm para-ammunition by Fiocchi shot from 3 metres away. EDX spectra showing a two-component PbSb particle found on the sample. Pb and Sb are the only major peaks present.





Figure a16: .22 " LR calibre ammunition by Winchester shot from 5 metres away. Backscattered electron image showing a three-component GSR particle measuring 5.823 μ m, found on the sample. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 μ m.



Figure a17: .22 " LR calibre ammunition by Winchester shot from 5 metres away. EDX spectra showing a three-component GSR particle found on the sample. Electron image of this particle can be seen in figure a16. Pb, Ba and Sb are the major peaks. Some S is present as expected with a high Sb peak. There appears the be trace Na,Cl which may be a contaminant on the background substrate. The As is a mislabel as can be seen by its absence from some labelled areas.



Figure a18: .22 " LR calibre ammunition by Winchester shot from 10 metres away. EDX spectra showing a two component PbSb particle found on the swab. Pb and Sb are the only significant peaks present.



50 μm EHT = 20.00 kV WD = 9.5 mm

Figure a19: .223 " rem calibre ammunition by Hornady fired from 5 metres away. Backscattered electron image of a three-component GSR particle measuring 12.01 μ m, found on the sample. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 μ m.



Figure a20: .223 " rem calibre ammunition by Hornady fired from 5 metres away. EDX spectra showing a three-component GSR particle found on the sample. Electron image of this particle can be seen in figure a19. As well as the major Pb, Ba and Sb peaks there is significant Ca and Al present. This is not a combination expected in GSR and may be due to the surrounding particles. The As and Kr are mislabels.





Figure a21: 12-Gauge shotgun ammunition by Eley shot from 3 metres away. Backscattered electron image showing a three-component GSR particle measuring 4.102 μ m, found on the sample. Electron high tension: 20 Kv, Working distance, 9.5 mm, 20 μ m.



Figure a22: 12-Gauge shotgun ammunition by Eley shot from 3 metres away. EDX spectra showing a three-component GSR particle with additional AI found on the sample. Electron image for this particle can be seen in figure a21. Note the higher levels of Ba and AI associated with shotgun ammunition. There is some Ca which may come from surrounding debris.