# The relevance of gunshot residues in forensic science

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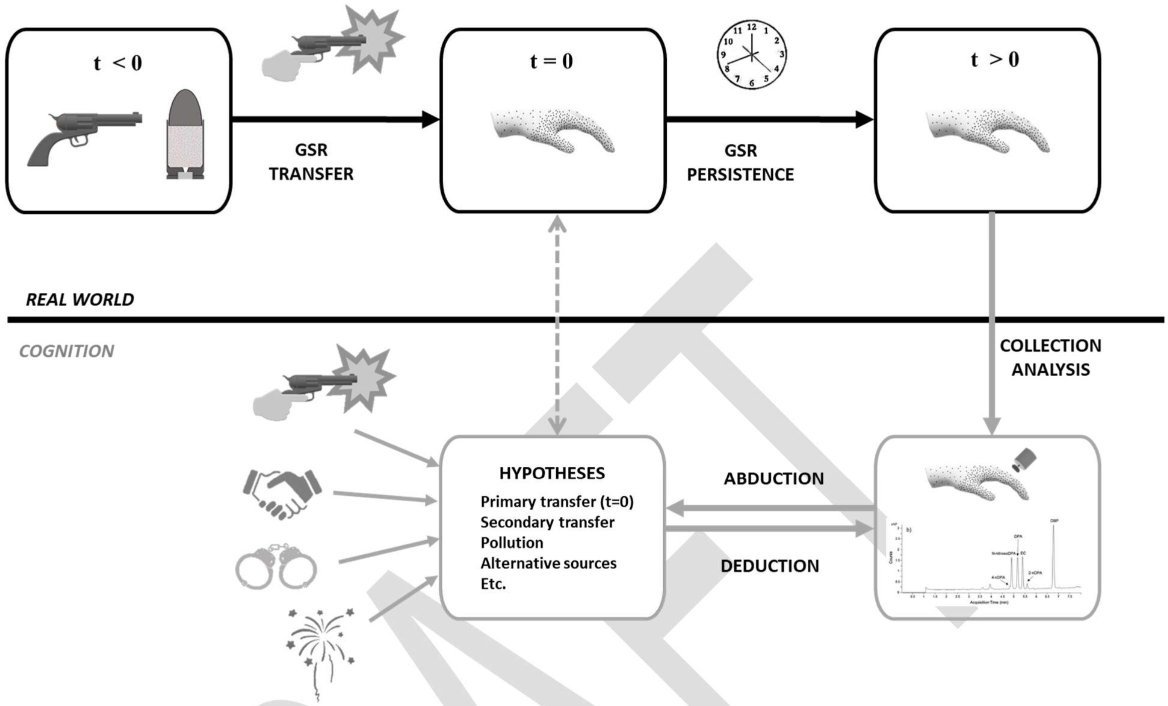
## Abstract

Gunshot residues (GSR) are routinely exploited by forensic scientists in the investigation of firearm-related events. While many new techniques are daily reported in the literature for the analysis of GSR, there is still a significant lack of data on the transfer, persistence, and prevalence of GSR. Such fundamental knowledge is essential to fully exploit the information potential of GSR for investigation or in Court. This paper provides an overview of the relevant questions related to GSR, more particularly to infer about the trace's origin (i.e., is it from a firearm discharge?) and the activity that caused transfer (e.g., primary, secondary or subsequent transfer). GSR production and composition will be briefly described, considering both inorganic and organic components. Then, the available knowledge about the primary transfer, the secondary transfer and the persistence of GSR will be outlined, as well as the prevalence (background level) of the targeted elements and/or compounds in the environment, more particularly on the hands of people unrelated to firearm incidents. Finally, the methods developed for the collection, analysis and interpretation of GSR will be discussed. A holistic approach combining fundamental forensic science knowledge about GSR transfer, persistence and prevalence together with other available information is discussed as a path forward to increase the relevance and value of the GSR trace in practice.

## Graphical Abstract




Gunshot residue is a relevant trace in the investigation of firearm related events. While much research has been carried out to develop detection and analysis methods, the focus needs now to shift from means to purposes. More knowledge on the trace characteristics, how it transfers, persists and the prevalence in the environment, will lead to a more reliable evaluation from a forensic rather than analytical perspective.

**Keywords:** Forensic science, firearm discharge residue, GSR, trace, transfer, persistence,

prevalence, background level, detection, analysis, interpretation.

# 1. Introduction

Firearm related incidents can lead to the transfer of many traces, related or not to the discharge of the firearm(s). While DNA, fingermarks or fibres can be transferred and detected on firearms and related materials (e.g., bullets, cartridge cases), specific traces are typically encountered during the investigation of such events such as striae and deformation of bullets, impact holes, spent cartridge cases and gunshot residues (Charles et al., 2010; Charles et al., 2020; Maitre et al., 2017; Mattijssen et al., 2016). These traces contribute to the inference of bullet trajectories, number, distance and time of the shooting, as well as material characterisation (e.g., gunshot residue) and association (e.g., between bullet, cartridge case and firearm; between the individuals involved and the firearm or the shooting event) (Brozek-Mucha, 2017; Mattijssen et al., 2016; Meng & Caddy, 1997; Nichols, 2018; Niewoehner, 2005; Romolo & Margot, 2001). This overview focuses more particularly on the relevance of gunshot residues (GSR)[1] also referred as firearm discharge residue (FDR) or cartridge discharge residue (CDR), in the forensic investigation of firearm related incidents.

GSR are produced during the discharge of a firearm and transfer on surrounding people and surfaces. The results of their qualitative and quantitative analysis have the potential to help answer different types of questions such as (Pitts & Lewis, 2019; Wallace, 2008; Wolten et al., 1977): Who handled or discharged a firearm? Who was present when a firearm was discharged? What firearm and ammunition were discharged? Where or when was a firearm discharged?

One of the main purposes of GSR detection and analysis is to evaluate **if the trace's origin is a firearm discharge (source level) and what type of activity caused the transfer (activity level)**. It is important to differentiate these questions as GSR can be transferred directly due to a firearm discharge (primary transfer) or indirectly through secondary or a higher level transfer (Charles & Geusens, 2012; Ditrich, 2012; French & Morgan, 2015; French et al., 2014; Gassner et al., 2019; Gassner & Weyermann, 2020; Hofstetter et al., 2017; Krishnan, 1977; Maitre et al., 2019; Manganelli et al., 2019)

Thus, in practice the following questions are generally more particularly addressed:

[1] The acronym GSR will be used in this article.

- Are GSR target components detected (e.g., specific inorganic elements or organic compounds)?
- How many different target components are detected?
- Where and when are they detected?
- In what quantity are they detected?

To answer these questions, forensic scientists need to acquire fundamental knowledge on the GSR trace (i.e., transfer, persistence, prevalence) and develop methodologies to detect, collect, analyse and interpret the trace in a forensic casework context. This paper aims at outlining available knowledge on the GSR trace, focusing more particularly on the residue transferred on shooter vs non-shooters (rather than on targets or surrounding objects). GSR detection and analysis are also described, and the possibilities and limitations of current approaches will be discussed, suggesting a path forward for the development of forensic GSR research.

## 2. GSR production and composition

GSR refers to all material (e.g., particles, elements, compounds) generated and transferred during or immediately after the discharge of a firearm. When the trigger is pulled, the firing pin strikes the primer of the cartridge, producing a burst of flame that ignites the smokeless powder. During the combustion of the powder, a large quantity of gas is produced and contributes to the rapid increase of temperature and pressure (Tenney, 1953; Thornton, 1994; Zeichner, 2009). Under these extreme conditions, the bullet is pushed out of the muzzle and vapours (also called “plume”) composed of gas as well as burnt and unburnt particles are released through all available openings of the firearm (Schwoeble & Exline, 2000). These gas and particles known as GSR are transferred on nearby surfaces including the firearm, the hands, clothes, face and hair of the shooter and bystanders. They can also be found on the target and surrounding surfaces (Blakey et al., 2018). The residues originate from the primer, the cartridge case, the projectile (i.e., the bullet), the propulsive charge (also called smokeless powder) and the weapon including lubricants and cleaning products (see Figure 1).

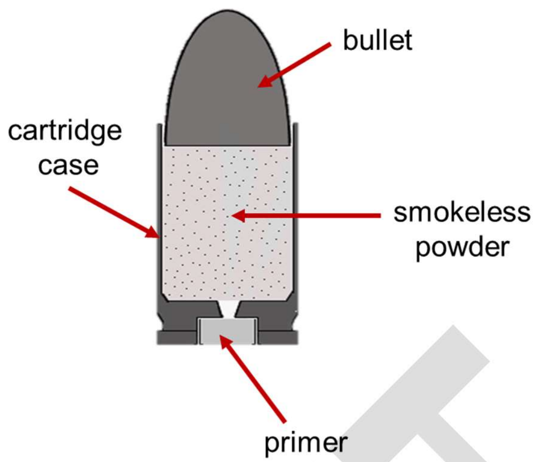



**Figure 1** - Different parts of ammunition contributing to the GSR trace.

Two main types of GSR are usually differentiated for methodological purposes as different methods are generally used to detect and/or analyse them: inorganic and organic GSR (Blakey et al., 2018; Dalby et al., 2010; Feeney et al., 2020; Romolo & Margot, 2001; Wallace, 2008; Wolten et al., 1977).

### *2.1. Inorganic GSR (IGSR)*

IGSR originate mainly from the primer mixture (see Table 1). They are formed after the vaporisation and condensation of heavy metals elements contained in the primer mixture (e.g., lead, antimony, tin, copper and barium) (Schwoeble & Exline, 2000). The weapon and the cartridge case also produce IGSR through surface wrenching but generally in smaller proportions (Gallidabino & Weyermann, 2020; Wolten et al., 1979). The primer is a mixture of components having different roles such as the primary explosives, the sensitisers or the pyrotechnic system. These components are generally rich in heavy metals elements. The composition of the primer has changed over time. Mercury fulminate was initially used as the primary explosive together with potassium chlorate (oxidant), antimony trisulfide (flash development), and glass powder (friction agent). These primers were replaced by the Sinoxid®-type primer (in the early 1950s for the US military), a mixture of lead styphnate (primary explosive), barium nitrate and antimony sulfide (pyrotechnic system), to avoid corrosion of the firearms (Brede et al., 1996; Wallace, 2008). Sinoxid® type primers have the advantages of high resistance to corrosion and good chemical stability. They produce heavy-metal particles, and some of them have an elemental combination that is considered characteristic of GSR (e.g., PbBaSb) (Hagel & Redecker, 1986; ASTM

International, 2020; Niewoehner et al., 2003; SWGGSR, 2011). While these primers are still predominantly used nowadays, toxic elements (lead, barium and antimony) are released in the environment during the discharge (Brede et al., 1996). Such toxicity is particularly an issue for indoor shooting activities and other similar situations. As a result, "heavy metal-free" primer or ammunition (e.g., Sintox®, Action 4, PEP II) were developed using less toxic elements such as diazole (primary explosive), zinc peroxides and titanium chlorides (pyrotechnic system) instead of lead, barium and antimony, leading to the production of different consistent particles such as TiZn and Sr (Charpentier & Desrochers, 2000; Gunaratnam & Himberg, 1994; Oommen & Pierce, 2006). Some police services also use modified heavy-metal free ammunition containing tagging elements[2] such as gadolinium, gallium and samarium (Donghi et al., 2019; Niewoehner et al., 2006). Glass powder is also added to the primer mixture as a fractionator. Thus, glass particles can be encrusted in the IGSR particles (Seyfang, Lucas, Popelka-Filcoff, et al., 2019; Seyfang, Lucas, Redman, et al., 2019).



| Primer mixture | | |
|---|---|---|
| **Ammunition type** | **Sinoxid-type** | **Heavy metal-free** |
| **Role** | **Compounds** | |
| **Initiator** | Lead styphnate | Diazole<br>(2-diazo-4,6-dinitrophenol) |
| **Sensitiser** | Tetrazene | Tetrazene |
| **Oxidiser** | Barium nitrate<br>Lead peroxide<br>Lead dioxide | Zinc peroxide |
| **Fuel** | Antimony sulfide<br>Calcium silicate<br>Metal powder (Magnesium, Titanium, Zirconium or Aluminium) | Titanium chloride |
| **Friction agent** | Glass powder | Glass powder |
| **Tagging elements** | - | Gadolinium (III) oxide<br>Gallium-copper-tin<br>Samarium oxide<br>Titanium oxide |

**Table 1** – Example of primer mixture compositions. The main targets components of current methods are PbBaSb, TiZnGd, GaCuSn particles as they are considered characteristic of IGSR. Other particles (such as PbBa, PbSb, PbBaCaSi, BaCaSi, BaAl, BaSb, Pb, Ba and Sb) are also targeted by the analysis but are considered to be less specific (ASTM International, 2020), TiZn and Sr, were suggested as consistent with heavy-metal free ammunitions.

[2] Tagging elements are added for better traceability of GSR (more particularly for GSR produced by police ammunitions).

### *2.2. Organic GSR (OGSR)*

OGSR are mainly from the smokeless powder (see Table 2) that is composed of primary explosives, stabilisers, plasticisers, gelatinisers, sensitisers, and flash inhibitors (Chang et al., 2013; Feeney et al., 2020; Goudsmits et al., 2015). During ammunition storage, reactions with stabilisers and sensitisers may occur (Dong et al., 2020; Laza et al., 2007). For example, the stabiliser diphenylamine (DPA) is known to capture the NOx produced by the degradation of NC and NG through the chemical reaction of N-nitrosation, C-nitration, and N-denitrosation, causing the production of compounds such as *N*-Nitrosodiphenylamine (*N*-nDPA), 2-nitrodiphenylamine (2-nDPA), 4-nitrodiphenylamine (4-nDPA), 2,4-dinitrodiphenylamine (2,4-DNDPA) and 4-nitrosodiphenylamine (4-NODPA) (Espinoza & Thornton, 1994; Folly & Mäder, 2004; Laza et al., 2007; Tenney, 1953). Although, 2-nDPA and 4-nDPA are depletion products, they are also reported in the literature as stabilisers added to the powder (Goudsmits et al., 2016; Joshi et al., 2011).

| **Smokeless powder** | |
|---|---|
| **Role** | **Compounds** |
| **Energy carriers** | Nitrocellulose (NC)<br>Nitroglycerin (NG)<br>Nitroguanidine (NQ) |
| **Stabilisers** | Diphenylamine (DPA)<br>2-nitrodiphenylamine (2-nDPA)<br>4-nitrodiphenylamine (4-nDPA)<br>*N-Nitrosodiphenylamine(N-nDPA) **<br>*4-nitrosodiphenylamine (4-NODPA) **<br>*2,4-dinitrodiphenylamine (2,4-DNDPA) **<br>Ethylcentralite (EC)<br>Methylcentralite (MC)<br>Akardite I to III (AK-I to AK-III) |
| **Sensitisers** | 2,4,6-trinitrotoluene (TNT)<br>Pentaerythritol tetranitrate (PETN) |
| **Flash inhibitor** | Dinitrotoluene (2,3-, 2.4-, 2.6-, 3,4-DNT)<br>2 to 4-nitrotoluene (2 to 4-NT) |
| **Plasticisers/Gelatinisers** | Dibutylphthalate (DBP)<br>Dimethylephthalate (DMP)<br>Diethylphthalate (DEP)<br>Triacetin |

**Table 2** – Example of smokeless powder composition. The target components of analytical methods are mainly the unburnt stabilizers. The primary explosives and flash inhibitors are less often targeted by the analysis. *Compounds marked by an asterisk (*) are degradation products of DPA and TNT, respectively.*

After the discharge of a firearm, unburnt components of the smokeless powder including degradation products can be found in GSR due to incomplete ignition, vaporisation and condensation of the smokeless powder. Combustion products are also produced during the discharge through pyrolysis, pyrosynthesis processes and/or stoichiometric combustion reaction. For instance, benzonitrile and polycyclic aromatic hydrocarbons (PAHs) are reported in the literature as combustion products frequently detected in the spent cartridge and firearm muzzle (Ase et al., 1985; Cropek et al., 2001; Gallidabino et al., 2015; Gallidabino & Weyermann, 2020). Several authors also reported the development of luminescent markers that could be added to the ammunition to facilitate GSR detection (Destefani et al., 2014; Lucena et al., 2013; Weber et al., 2014). While this type of ammunition is not available on the market yet, OGSR are known to produce luminescence that can easily be visualised using optical methods (Hofer et al., 2017; Hofer & Wyss, 2017). Some researchers are also aiming at replacing current stabilisers (e.g., diphenylamine, akardite-II) with environment-friendlier molecules such as curcumin, 2,3,5-trimethylphenol, alpha-ionone and alpha-tocopherol (Dejeaifve et al., 2018; Dejeaifve et al., 2020).

## 3. The GSR trace

The description above focused on GSR from a material and composition viewpoint. This section will expand the discussion on GSR as a forensic trace. A forensic trace can be defined as *the vestige or mark of a presence, an existence or an action of someone or something in a location or space that did not belong to that space initially* (Margot, 2017). The GSR trace is the vestige of an action (i.e., the firearm discharge) that was transferred on one or several individuals (i.e., shooter, bystander, victim), and surrounding surfaces (i.e., crime scene). However, the targeted GSR component can also be transferred through other actions. Thus, the relevance of a GSR trace in a forensic science context is directly linked to the following criteria:

- The trace must be transferred in sufficient quantity (transfer $t = 0$)
- The trace must be persistent (persistence $t > 0$)
- The trace must be specific (low prevalence in the environment)
- The trace must be easily and reliably detectable and quantifiable (see section 4 below)

If these criteria are at least partially fulfilled, inferences about the source of the trace and the activity that has led to the transfer can be made through a hypothetico-deductive process. This process may be more or less mathematically formalised depending on the purpose and jurisdiction (ANZPAA-NFIS, 2017; ENFSI, 2010; Ribaux & Caneppele, 2017; Roux et al., 2012).

### 3.1. Transfer

#### *3.1.1. Primary transfer*

Primary transfer occurs during or immediately after a firearm discharge. GSR is generally transferred on the hands of the shooter, mainly on the upper parts of the thumb and index (see Figure 2) (Brozek-Mucha, 2011; Chohra et al., 2019; Hofstetter et al., 2017; Wolten et al., 1977). Residues can also be transferred to other body parts and clothes: on the forearms, arms, chest, trousers, pockets, face, or hairs of a shooter and by-standers (Andrasko & Pettersson, 1991; Hofstetter et al., 2017; Krishnan, 1977; Merli et al., 2016; Zeichner & Levin, 1995). Some surfaces, such as the inside of the ears or the nostrils were also reported as good receivers (Chavez Reyes et al., 2018; Dobarceranu, 2020a, 2020b).

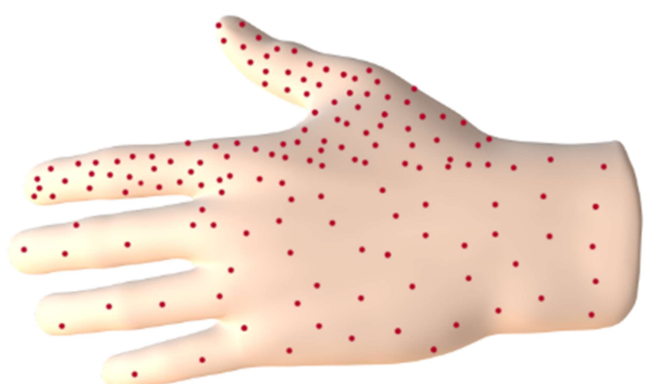


**Figure 2** - GSR is generally transferred in the V-shaped part of the hand between the thumb and the index finger (Meng & Caddy, 1997). GSR are mostly transferred on the upper part of the hand(s) holding the firearm but can also be transferred on the palm(s) as GSR are also present on the firearms.

Only a few studies, using a variety of different analytical methods, reported the relative quantities of I- and OGSR transferred just after the discharge. GSR transfer is influenced by several factors such as donor (firearm/ammunition) - receptor (receiving surface) affinity and the environment (indoor/outdoor) (Chohra et al., 2019; Ditrich, 2012; Moran & Bell, 2013, 2014). The type of firearm has a significant influence on the plume formation and the distribution of the GSR produced (Schwoeble & Exline, 2000). Studies have reported a higher concentration of GSR on the side corresponding to the ejection port of the firearm (Blakey et al., 2018; Fojtášek et al., 2003; Gerard et al., 2011; Hofstetter et al., 2017). For example, the position of the ejection port of a semi-automatic/automatic pistol, the different openings of the cylinder of revolvers, and the barrel length influence the direction and distribution of the transfer GSR (Ditrich, 2012). The type of ammunition used also influences the quantity, composition and particle size of the produced GSR (Campbell, 2018; Chohra et al., 2019; Gallidabino et al., 2015; Gassner et al., 2016; Rijnders et al., 2010; Zeichner et al., 1991). Schwoeble and Exline (2000) noted that smaller particles are generated with revolvers than with semi-automatic/automatic pistols. Furthermore, the transfer is also known to vary significantly between shots of the same firearm and ammunition (Hofstetter et al., 2017; Lopez-Lopez et al., 2013). External environmental factors, such as airflow (e.g., wind), humidity, rain, and distance (of bystanders, target and surrounding surfaces) to the firearms also play a role in GSR primary transfer. In general, the closer a receiving surface is to the plume, the higher is the GSR transfer. In fact, OGSR were observed to transfer in higher average quantities on the hands, gloves, and forearms close to the discharge than on the upper clothes of shooters (Hofstetter et al., 2017). Gerard et al. (2011) observed larger quantities of IGSR near the projectile path and on the right side (side of the ejection port) of the bullet path. This indicates that a person near the projectile or bullet path may be exposed to GSR. The GSR distribution and quantity on the target(s) is also influenced by the distance between the firearm and impacted surface and can thus be used to estimate the shooting distance (Bartsch et al., 1996; Marty et al., 2002; Zeichner & Glattstein, 2002). Some GSR are additionally carried out with the bullet and can be found at entrance (and sometimes also at exit) holes (Brown et al., 1999; Merli et al., 2019).

**Case study 1 –** The potential of GSR to infer about activities

GSR localisation and quantification can be useful in the investigation of suspicious deaths (see Figure 3). In a case known to the authors, IGSR were found in high amounts on the right hands of two dead people. The lifeless bodies were found lying together on the ground. The first person was holding the firearm with his right hand (very high quantities of IGSR were detected). The second person was holding the right forearm of the first person with her right hand (lower, but still relatively high quantity of IGSR was detected). This second person was placed partially under the first person. The lifeless body of a third person was also found close to the others (no GSR detected on the hands). The investigation and detected traces allowed to formulate the proposition that the first person was the shooter. Bullet holes and projectiles were used to reconstruct trajectories, sequence, and the number of discharges. It was inferred that the first person shot several times before the second person reached him and grabbed his right arm. The collected information (including IGSR) enabled to formulate the proposition that the person holding the firearm shot the other two persons in the head (first the third person at close range and then the second person point-blank in the head), before shooting himself also in the head.



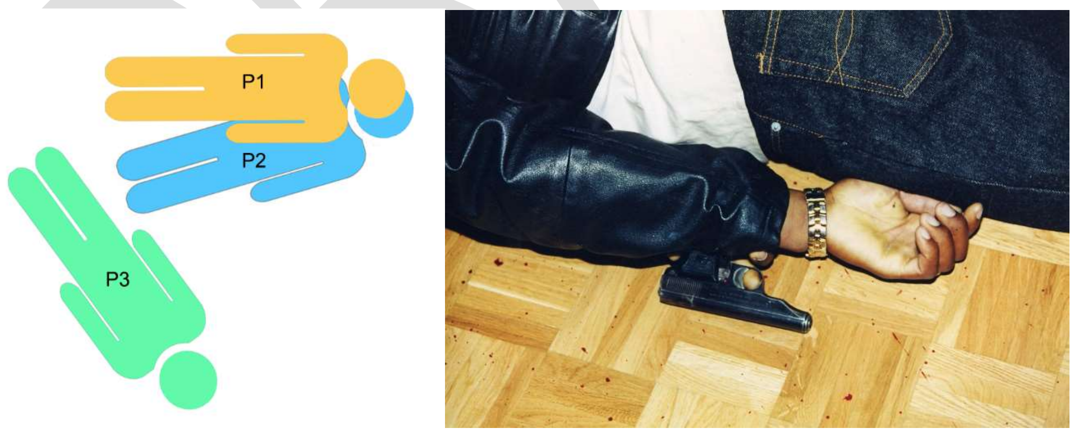



**Figure 3** – Case example 1: schematic representation of the three lifeless bodies as found during crime scene investigation (left) and the first person holding the firearm with his right hand (right).

### *3.1.2. Secondary (and subsequent) transfer*

Following the primary transfer, GSR may be transferred to another surface (receptor) through a secondary transfer. In this case, there is no direct contact between the original donor (firearm/ammunition) and the receiving surface. Subsequent transfer (tertiary, quaternary, …) can further occur, even if they may be difficult to trace back to the original GSR source (see also prevalence section).

The word contamination is often used to describe the consequence of the secondary (or subsequent) transfer. It is important to differentiate the contaminations that are due to careless or poor practice and should be avoided (e.g., caused by an investigator or a respondent at the scene) and those that are part of the scene background and cannot be avoided because they occurred before the scene was secured (e.g., contamination caused by environmental conditions, scavengers or simply the context). Contaminations that are avoidable and caused by poor practice can be referred to as "pollution" (Margot, 2017; Schwendener et al., 2016).

Unavoidable contaminations can occur through different processes linked to primary and subsequent transfers of GSR or GSR alike traces. Primary or secondary transfer of GSR can be due to legitimate activities such as sport shooting, hunting or firearms manufacturing facilities (Gerard et al., 2013; Lucas, Brown, et al., 2016). However, a person of interest (POI) may also come into contact with GSR before the investigation at the crime scene when discovering an incident (e.g., touching a victim to check for vital signs, displacing a firearm or passing through the GSR cloud a few minutes after the shot) (Fojtášek et al., 2003; Luten et al., 2018). Finally, a person may also come into contact with GSR alike traces from other sources and activities unrelated to firearms (see prevalence section below for more details).

Secondary transfer of IGSR was reported from a shooter to a POI after a handshake or the handling of a firearm recently discharged (Brozek-Mucha, 2014; French et al., 2014). French et al. (2014) observed that, in general, a higher number of particles were transferred after a handshake (i.e., just after the discharge occurred) than after the handling of a firearm. A tertiary transfer was also observed in a study by French and Morgan (2015) after a series of handshaking. Secondary transfer of IGSR was reported from police officers to a POI during arrest (Lucas et al., 2019). The scenario involved physical contact for 5 minutes. In most experiments, characteristic particles were transferred to the hands of the POI (average value of 4 characteristic particles). Charles and Geusens (2012) studied the transfer of GSR from police officers and duty firearms to a POI. The scenarios did not involve shooting, but manipulation and loading of the service weapon. While the transfer of GSR did not occur every time, it occurred frequently when the police officer proceeding to the arrest was highly contaminated with GSR. In fact, up to 13 characteristic particles were found on the hands of the arrested person under the high pollution scenario (i.e., police officers were allowed to have their equipment; bulletproof vest, technical vest, and gloves). The gloves were identified as the main source of GSR transfer (average value of 66 characteristic particles on the gloves after the discharge). Moreover, on average, slightly more particles were found on the clothes (vest) in comparison to the hands of the POI.

Preliminary studies of secondary transfer using ion mobility spectrometry (IMS), detected no OGSR after handshaking (Arndt et al., 2012). Ali et al. (2016) also investigated the transfer of OGSR in a police vehicle using liquid chromatography coupled to tandem mass spectrometry (LC-MS/MS). The POI was handcuffed and placed 10 minutes inside the vehicle. Two specimens (of 32) were positive for ethylcentralite (EC). Secondary transfer of OGSR was also reported in two other studies that used LC-MS/MS (Gassner et al., 2019; Maitre et al., 2019). Three scenarios were tested, transfer from the shooter to a POI after a handshake or an arrest as well as transfer during the handling of a firearm recently discharged. While the results obtained in both studies differed in the percentage of positive specimens (i.e., number of polluted POI and relative quantities), both warned about the (sometimes relatively high) risk of pollution for the investigated scenarios. In some instances, more OGSR were found on the non-shooter than on the shooter after contact. The intensity and duration of the contact had an influence on the GSR transfer. However, all scenarios occurred directly after discharge (high risk scenarios) and secondary transfer may significantly decrease some time after the discharge (see also persistence section below).

While pollution occurring during a police intervention can to some extent be controlled and minimized by different measures (e.g., protecting a person's hands before sampling, single-use equipment during the sampling), all contaminations cannot be avoided and must be taken into account in practice. Indeed, police officers proceeding to a complicated arrest may be contaminated for example through shooting training sessions or regular contact with contaminated surfaces (e.g., police vehicles, special unit equipment, and firearms (Berk et al., 2007; Cook, 2016; Gassner & Weyermann, 2020; Gerard et al., 2013; Gialamas et al., 1995). Background samples taken at the scene and from similar materials to those under investigation are, therefore, very important to account for the risk of secondary transfer in the investigation of firearm incidents (Feeney et al., 2021; Lucas et al., 2019; Manganelli et al., 2019; Stamouli et al., 2021). For example, in case study 2, it would have been important to collect background samples from the scene of the shooting (e.g., very close to the bodies, and also further away to evaluate the "overall levels of gunshot residues in the immediate environment" as well as on the people having handled the bodies and in the vehicles used to transport them.

**Case study 2** – The risk of GSR contamination

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| The Bloody Sunday inquiry[3] highlighted that the source of the recovered lead particles on the clothes and hands of the deceased remained undetermined and may have been contaminations rather than indications that they fired or manipulated guns. The following statement by Dr Martin, the forensic scientist in charge of the gunshot residue examination, highlighted the need for background samples to evaluate the risks of contamination of forensic specimens: *I think that contamination is the major issue. When I did the tests and prepared my report for Widgery I was under the impression that 20 to 30 shots had been fired and that the bodies had been transferred in clean conditions to the mortuary. It was only at my cross-examination that I became aware that over 100 shots had been fired, greatly increasing both the overall levels of gunshot residues in the immediate environment and the possibility of fragmentation. In addition, at least some of the bodies had been handled and transported in a way that could have resulted in contamination by gunshot residues.* The concurrent propositions (i.e., discharge of a firearm, ambient contamination due to the number of shootings in the area, secondary transfer due to the handling and transport of the victims) cannot be adequately evaluated without background control samples. |



[3] See more particularly the volume VI of the report of the Bloody Sunday Inquiry and main hearing on day 226: https://webarchive.nationalarchives.gov.uk/20101017060829/http://report.bloody-sunday-inquiry.org/transcripts-index/ (last access: June 2020).

### 3.2. Persistence (t>0)

After transfer, GSR may be lost due to activity (e.g., walking, running, typing or handwashing) and environmental conditions (e.g., wind, rain) (see Figure 4). Only a few persistence studies have been carried out as they are relatively complicated to design. They all showed the need to sample the area of interest (e.g., the hands, face, or forearms of the POI) as quickly as possible given the rapid loss of GSR in the first hours after the transfer (Brozek-Mucha, 2011; Jalanti et al., 1999; Maitre et al., 2018).

The persistence is also affected by the chemical properties of the GSR. IGSR and OGSR are two classes of residues coming from different parts of the ammunition (i.e., primer and smokeless powder). IGSR are very stable metallic particles, that are removed mainly by physical phenomena (i.e., movement and friction). OGSR are organic compounds, that can volatilise or be absorbed on the skin (lipophilic properties) (Moran & Bell, 2013). Moran and Bell (2014) have identified EC, 2-nDPA, and 4-nDPA as more persistent compounds considering their low volatilisation rate and their slow skin permeation.

In suicide cases, higher amounts of GSR are expected to persist on the hands considering the absence of activity of the shooter. However, studies on the subject reported the detection of very few or no characteristic particles on some of the samples taken (Lucas, Cook, et al., 2016). While the persistence can be affected by post-mortem changes, as well as outdoor conditions, studies on the persistence of GSR on decomposing porcine tissue found that GSR are still detected on tissue and skeletonized samples several weeks after a shot (Gibelli et al., 2010; Lagoo et al., 2010). The high variability of the transfer and persistence phenomenon illustrates the difficulty in distinguishing a self-inflicted from a non-self-inflicted wound based on GSR found on the hands of a dead person (Lucas, Cook, et al., 2016; Molina, Castorena, et al., 2007; Molina, Martinez, et al., 2007).

Studies on the persistence of IGSR on the hands of a shooter indicated that most target elements (e.g., Pb, Ba, Sb) were lost during the first hours after a discharge (following an exponential decrease) (Andrasko & Maehly, 1977; Brozek-Mucha, 2011; Jalanti et al., 1999; Kilty, 1975; Nesbitt et al., 1976; Nesbitt et al., 1977; Wolten et al., 1977). Despite the rapid loss, Andrasko and Maehly (1977), Jalanti et al. (1999), and Brozek-Mucha (2011) still reported the detection of a few characteristic particles (PbBaSb) up to 4 hours after the discharge using the Scanning Electron Microscopy coupled to Energy

Dispersive X-Ray Spectrometry (SEM/EDS). Rosenberg and Dockery (2008) were also able to detect Ba up to 5.27 days after six consecutive shots using laser-induced breakdown spectroscopy (LIBS). All research had in common a large variation in particle counts between replicate. Schütz et al. (2001) additionally, observed that the type of ammunition and firearm influenced the amount of IGSR generated. While IGSR persistence on hands is limited due to handwashing or hand sanitizer application (Feeney et al., 2021; Kilty, 1975; Nesbitt et al., 1977), particles could be detected up to 23h on the hair and face as well as in the nose and ears of the shooter (Chavez Reyes et al., 2018; Dobarceranu, 2020a, 2020b; Zeichner & Levin, 1993). In the case of collection on clothes, the recovery of IGSR is influenced by the type of surface (e.g., cotton, leather, wool) (see the section on collection device below) (Charles et al., 2013) and it is important to mention, that brushing and washing can result in a significant loss (Vinokurov et al., 2001).

As for the IGSR, studies showed that OGSR concentrations decrease rapidly in the first hours following the transfer (Northrop, 2001b), but that target compounds can still be detected after 4 hours (Arndt et al., 2012; Gassner et al., 2016; Gassner & Weyermann, 2016; Maitre et al., 2018). After half an hour, Maitre et al. (2018) detected up to 50% of the initial concentration on the hands of a shooter. However, the detected amount decreased to 8-37% after 4 hours depending on the targeted compound and firearm used. In the study of Gassner et al. (2016) less than 1% and 5% of the initial concentrations of ethylcentralite and 2-nitrodiphenylamine respectively were found on the hands of a shooter after 2 hours.

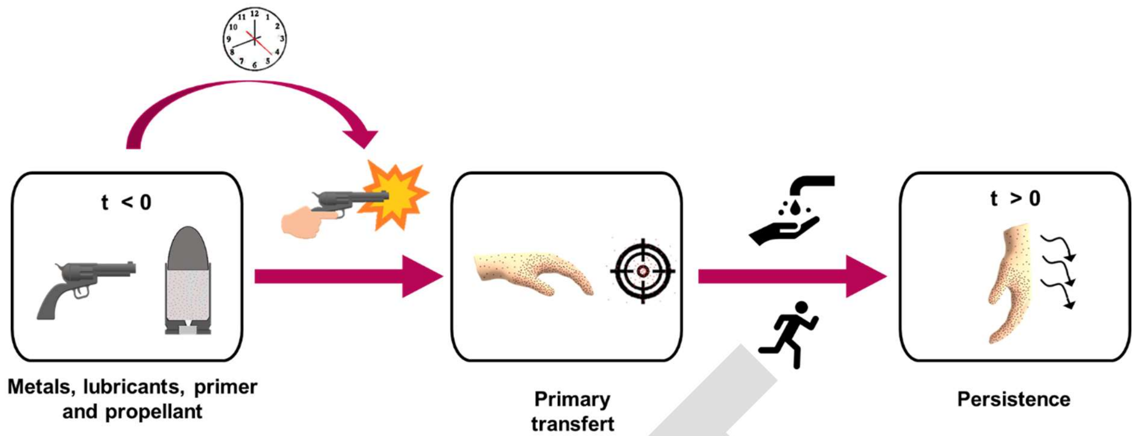


**Figure 4** – The GSR trace detected during the investigation of a firearm discharge is dependent: 1) on the state and composition of the initial materials used (e.g. firearm, ammunition) and on the age of the smokeless powder (e.g. depletion of the stabilisers/sensitisers), 2) on the transfer conditions (e.g., wind, distance to surrounding surfaces, receptor affinities) and 3) on the time, environment and activities between transfer and collection (e.g. rain or handwashing will strongly limit the persistence of GSR on the exposed surfaces).

### 3.3. Prevalence

The prevalence of a trace can be defined by its background distribution in a given population (e.g., on the hands of people unrelated with firearm incidents) or in the environment (e.g., on scenes before the occurrence of a crime) (Berk, 2009a, 2009b; Drzyzga, 2003; Llyod, 1987; Torre et al., 2001). While secondary GSR transfer can explain some background contaminations, other activities also promote the transfer of particles or compounds undifferentiable from I- or OGSR (e.g., welding or fireworks) (Brozek-Mucha, 2015; Mosher et al., 1998). Knowledge of the prevalence of target components in given contexts is essential for correct evaluation of the signification of their presence in case situations. Thus, prevalence studies are necessary knowledge to take into account the risk of legitimate occurrence of targeted components on POI.

The prevalence of IGSR in the civilian population has been studied in various countries and results indicate that a low number of characteristic particles were generally detected. Three characteristics particles (PbBaSb) were found on the hands of one person out of 289 in Australia (Lucas, Brown, et al., 2016). In a Polish study, only one characteristic particle was found on the hands of one person out of

100 persons declaring no contact with firearms (Brozek-Mucha, 2014). A recent international study investigated the prevalence on the hands of more than 1300 persons and found that the mean probability of finding at least one PbBaSb particle for a low-risk population (i.e., general population and car mechanics, n= 752 persons) was 1,7% (Stamouli et al., 2021). More than 10 PbBaSb particles were detected only on the hand of one person of the general population. In all these studies, car mechanics were not found to be more at risk than the general population, while police officers and people owning firearms presented a much higher IGSR prevalence (see also the section about the secondary transfer above).

For OGSR, a study in the US used micellar electrokinetic capillary electrophoresis (MECE) to analyse samples collected from the hands of 100 volunteers, and no positive results were obtained (Northrop (2001a). Another study conducted in the US on the hands of 73 persons from the general population using IMS yielded less than 5% positive results (Bell & Seitzinger, 2016). Two studies in Switzerland investigated the prevalence of OGSR in the civilian population using LC-MS/MS. This instrument is particularly interesting considering the lower limits of detection. In the first study, no OGSR target compounds were found on the hand of 27 people (Hofstetter et al., 2017). In the second study, samples from the hands, forearms, and sleeves of 122 persons were analysed (Manganelli et al., 2019). While one or more OGSR compounds were detected in 18% of the population, only 2 persons had more than four compounds. One person with six OGSR compounds on his hands indicated having discharged a firearm less than 10 minutes before the sampling occurred. However, the other person could not explain the presence of four OGSR compounds on his sleeves. In this prevalence study, 4-nDPA and AK II were the most frequently detected compounds.

One recent study investigated the prevalence of both I- and OGSR on a low-risk (i.e., general population having no contact with firearms) and a high-risk (i.e., police officers, mechanics, agricultural workers, ballistics research personnel) population (Feeney et al., 2021). Low concentrations or concentrations below the detection limits were obtained for IGSR and OGSR in the low-risk population. EC was detected in 42% of these samples. For the high-risk population, higher concentrations were obtained for the EC, Ba, and Pb. These components were detected in approximately 40%, 90%, and 34% of these samples, respectively. Interestingly, the combination of EC, Ba, and Pb was observed in 9% of these samples.

# 4. GSR COLLECTION AND ANALYSIS

The first methodological step starts during the crime (scene) investigation. At this stage, the aim is to find relevant traces on the scene and people of interest following an hypothetico-deductive cycle (see graphical abstract) to reconstruct what happened (Baechler et al., 2020; Ribaux & Caneppele, 2017; Roux et al., 2012).

The following aspects need to be considered to extract relevant information from the GSR trace:

- The *forensic aspects*: knowledge about the trace, including specific case information for example about the competing hypotheses (e.g., suicide vs homicide vs accident) as well as knowledge about the trace transfer, persistence and prevalence are essential to determine where specimen (and background samples) should be collected.
- The *technical aspects:* knowledge about the efficiency of a collection device on encountered surfaces (e.g., skin, hair, fabrics) as well as selection criteria for analytical and data treatment methods (e.g., invasiveness, specificity, reliability and limits of detection) are needed to determine how specimen (and control samples) should be collected and analysed.

While both are important, the forensic aspects (section 3) form the basis upon which the technical aspects (section 4) can be developed (Roux et al., 2021). As a reminder, GSR detection and analysis is considered here mainly to evaluate if the trace's origin is a firearm discharge (source level) and what type of activity provoked the transfer (activity level). Thus, the methodological details will focus on GSR on a person of interest rather than on surrounding surfaces.

### 4.1. Collection

While OGSR particles can be visible to the naked eye (see black spots around the bullet hole in Figure 5), many are latent and need to be collected and analysed in more detail.

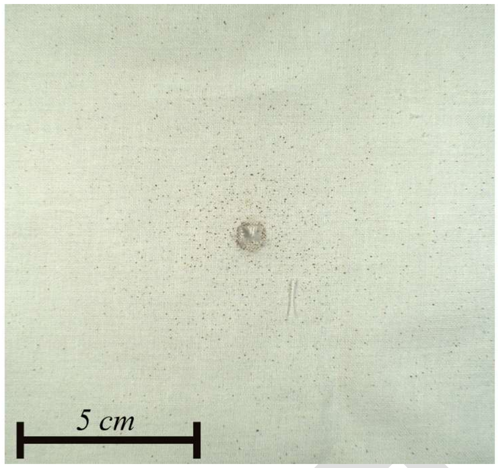


**Figure 5** – Visible GSR particles around a bullet hole on a white cotton tissue. The bullet was shot at 30 cm using a SigSauer P220 pistol (Thun® 9mm Parabellum). This picture was provided by our colleague Julia Fischer.

The collection stage needs to be compatible with the subsequent analysis method. It also requires knowledge about the transfer, persistence, and prevalence of GSR to target relevant areas on a person of interest (see section 3). For example, hands are generally targeted shortly after the discharge as high quantities are generally transferred on the hand(s) holding the firearm (Chohra et al., 2019; Hofstetter et al., 2017). During a discharge, GSR will be transferred both on the palms and back of the hands. If a person has only manipulated a firearm without discharging it, GSR are expected mainly on the palms. However, in practice, the sampling is often performed on the entirety of each hand (back and palm) to ensure sufficient quantities and decrease analysis costs. Moreover, if some time has passed between the investigated discharge event and the trace collection, persistence might be lower on the hands (e.g., if a person of interest has washed his hands in the interval) (Feeney et al., 2021; Kilty, 1975). Thus, other surfaces might also be targeted for collection such as hair, clothes, face, or nostrils (Brozek-Mucha, 2011; Hofstetter et al., 2017). While knowledge about the persistence and prevalence of GSR on those areas is limited (complicating the subsequent interpretation in a forensic context), other issues must also be taken into account as collection efficiency differs on different surfaces such as skin, fabrics, or hair.

The efficiency of collection devices was tested on different areas of interest for IGSR and/or OGSR. The main collection device, routinely used by police services and forensic laboratories, is an aluminum stub mounted with a double-sided carbon adhesive (see Figure 6). This device was reported as the most efficient tested device for the collection of IGSR and OGSR on the skin (DeGaetano et al., 1992; Gassner et al., 2016; Reid et al., 2010; Taudte et al., 2016). A modified stub was also recently proposed to access the nostrils (Chavez Reyes et al., 2018). Stubs can also be used to collect GSR on hair and fabrics, but the saturation of the carbon adhesive may occur for example on wool or cotton fabrics, thus, limiting the collection efficiency (Charles et al., 2013; Zeichner & Levin, 1993). Moreover, the presence of a large number of fibres and skin debris on the stub may complicate the detection of GSR particles with the SEM/EDS (Romolo & Margot, 2001).

**Figure 6** – Collection of GSR using an aluminum stub mounted with a carbon adhesive

Other collection devices were reported in the literature. For example, swabbing was proposed to collect OGSR as the targeted molecules dissolve easily in solvents. However, two studies indicated that swabs were less effective than stubs for the collection of OGSR on the hands (Gassner et al., 2016; Taudte et al., 2016). Swabs were reported to have the same efficiency as stubs to collect IGSR from the hair (Zeichner & Levin, 1993). Swabs were also found suitable for collection in the nasal mucus with the risk of causing nasal bleeding (Aliste & Chavez, 2016; Schwartz & Zona, 1995). An alternative was proposed by blowing noses on handkerchiefs, fabrics, or polymer film (Merli et al., 2016; Schwartz & Zona, 1995). Other devices were also tested to collect GSR such as a fine-toothed comb to collect GSR from the hair (MacCrehan et al., 2003) or vacuuming through a filtration system for collection from clothes including pockets (Andrasko & Pettersson, 1991). GSR particles may also be transferred onto materials such as filter papers, adhesive foils, or photo papers (ENFSI, 2015; Werner et al., 2020).

It is important to have general knowledge about the collection efficiency (i.e., the recovered quantities) to infer about the transferred quantities. However, a firearm discharge is a highly variable event, and it is difficult to reproduce GSR-like particles in controlled conditions. A successive sampling of the same target surface was studied and showed that some GSR were still present after a first sampling (Zeichner & Levin, 1993). Thus, it may be interesting to study and compare recovery rates between devices, but also to evaluate how many iterations are necessary until GSR components can not be detected anymore.

### 4.2. Analysis

The analysis of GSR aims to detect, qualify or quantify targets components (e.g., particles, elements, molecules). Each type of analysis has advantages and disadvantages that will be briefly discussed considering important criteria such as destructivity (i.e., the trace can be reanalysed using the same or a different technique), specificity (i.e., capacity to identify compounds or chemical elements), reliability (i.e., precision and accuracy) and limits of detection (LODs). Other criteria such as availability in forensic laboratories, cost, time, level of complexity (i.e., needed skills), hazards and possible application on the crime scene (e.g., portability) should also be considered when selecting a suitable method in a given forensic context.

- ***Optical methods*** have been found useful to deliver spatial information for the estimation of the shooting distance. In fact, luminescent GSR particles may be visualised in the near-infrared wavelength. Certain organic compounds (e.g., diethyl phthalate, dibutyl phthalate) have been identified as having a strong IR-luminescent (Hofer et al., 2017; Hofer & Wyss, 2017). Moreover, the visualisation of GSR particles on dark or multi-coloured fabrics may be enhanced using digital infrared photography (Bailey, 2007). These optical methods are thus, non-destructive and relatively rapid but, they have limited specificity and relatively high LODs. They can to some extent be carried out directly on the scene.

- ***Chemographic methods*** can be performed on the crime scene, deliver spatial and timely information about the trace (Geusens et al., 2019; Marty et al., 2002). Although they are invasive, some of them can also be carried out in sequence with other methods presenting higher selectivity (Werner et al., 2020). However, chemographic tests are only indicative and have thus, a high risk of false positive (Schwoeble & Exline, 2000). In practice, they are usually implemented to estimate the distance of shooting rather than to detect GSR on the hands of a person of interest. The Sodium Rhodizonate test targeting several inorganic elements (i.e., Pb, Ba, Sr, Cd, Sn, Ag, Hg, Ti, Cu, and Zn), the Rubeanic acid test (or the Dithiooxamide test) targeting copper and nickel, and the Modified Griess test targeting free nitrites, are usually used in practice for GSR analysis (Andreola et al., 2011; Bartsch et al., 1996; Berger et al., 2019; ENFSI, 2015; Lekstrom & Koons, 1986).

- ***Spectrometric instruments*** are the most common techniques for the analysis of IGSR. To date, the method of choice for the detection of IGSR in most forensic laboratories is Scanning Electron Microscopy coupled to Energy Dispersive X-Ray Spectrometry (SEM/EDS) (ASTM International, 2020; White & Owens, 1987; Wolten et al., 1977). It has the advantage over other methods of allowing a quantitative analysis as well as giving detailed information about the composition and morphology of the detected particles. However, SEM/EDS requires a high level of skill, is costly, and requires long analysis times. Neutron Activation Analysis (NAA) and Atomic Absorption Spectrometry (AAS) (Kanstrup et al., 2016; Krishnan, 1974; McFarland & McLain, 1973; Rudzitis et al., 1973) were the first methods proposed for the detection of inorganic elements and are complementary since NAA has a low LOD for barium, antimony, and iron while AAS has a low LOD for lead. However, these two techniques are less specific and therefore, rarely found in forensic laboratories. NAA is also a complex technique that require specific knowledge (Chohra et al., 2019; Stone & Petty, 1974). More recently, Graphite Furnace AAS (GFAAS) has been proposed as a more sensitive and specific alternative to AAS, but this method is also rarely available in forensic laboratories (Aliste & Chavez, 2016; Yuksel et al., 2016). X-Ray Fluorescence (XRF) and Laser-Induced Breakdown Spectroscopy (LIBS) have also been proposed for the elemental mapping of inorganic particles on specimens (e.g., clothes, stubs) (Berendes et al., 2006; Dockery & Goode, 2003; Gong et al., 2022; Latzel et al., 2012; Lopez-Lopez et al., 2017). They can be (trans)portable for direct analysis on the crime scene. However, they do not deliver information on morphology and particle counts. Inductively Coupled Plasma (ICP) coupled with Optical Emission Spectrometry (OES), Atomic Emission

Spectrometry (AES) or mass spectrometry (MS), and Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) are instruments with adequate LODs and specificity for heavy metals elements (e.g., Pb, Sb, Ba, Co, Ni, Zn). ToF-SIMS can additionally detect some organic compounds such as EC, NC and NG (Coumbaros et al., 2001; Mahoney et al., 2006). However, these techniques are also less available in forensic laboratories (Brunjes et al., 2022; Coumbaros et al., 2001; Diaz et al., 2012; Koons, 1998; Szynkowska et al., 2012). Fourier Transform Infrared (FTIR) and Raman spectroscopy were proposed for the analysis of OGSR (i.e., stabilisers and primary explosives) (Lopez-Lopez, Delgado, et al., 2012; Lopez-Lopez, Ferrando, et al., 2012). These instruments are not commonly used in practice but could be integrated as screening techniques considering their rapidity and potential portability. However, they have relatively high detection limits and may still require destructive sample preparation (to concentrate the samples). Desorption Electrospray Ionisation-Mass spectrometry (DESI-MS) was proposed for OGSR analysis as it required minimal specimen preparation and allowed a rapid analysis (Morelato et al., 2012; Zhao et al., 2008). The limit of detection was however deemed too high to be useful in forensic practice.

- ***Separation methods***, particularly when coupled to mass spectrometry (MS), are specific, reliable and have low LODs. While sample preparation is required and invasive, the extracts can generally be stored in a freezer and re-analysed. Gas Chromatography (GC) and Liquid Chromatography (LC) coupled to MS or MS/MS have been proposed for the analysis of OGSR (Laza et al., 2007; Tarifa & Almirall, 2015). Ultra High Performance (UHP)LC-MS presents nowadays the best LODs (sub-nanogram range) (Gassner & Weyermann, 2016; Taudte et al., 2015). GC-MS generally has higher LODs and some thermolabile compounds can be degraded during injection (e.g., NG, *N*-nDPA) (Muller et al., 2007; Zeichner & Eldar, 2004). Capillary electrophoresis is another separation technique that allows the simultaneous analysis of both I and OGSR but has relatively high LODs (Morales & Vazquez, 2004; Northrop, 2001a, 2001b). Such instrumental techniques are generally relatively costly and cannot be easily carried out on the scene. Ion mobility spectrometry is the only portable and quick separation method but is, unfortunately, less specific and reliable than the previous techniques (Arndt et al., 2012; Moran & Bell, 2013). OGSR can be collected on stubs without disturbing the distribution of

IGSR allowing lower invasiveness and the possibility of sequential analysis of both types of residues (Bonnar et al., 2020).

Nowadays, the analysis of IGSR using SEM/EDS is the one commonly applied in forensic laboratories. OGSR analysis is still rarely implemented in practice although a combined analysis of both types of GSR would provide additional information. Several studies have been carried out to develop and evaluate different approaches for the combined detection and/or analysis of I- and OGSR (Bell & Feeney, 2019; Benito et al., 2015; Bonnar et al., 2020; Redoute Minziere et al., 2020; Taudte et al., 2016). In total, three different approaches have been suggested using a combination of different collection and analysis techniques:

- One specimen is collected and divided in half for parallel analysis of I- and OGSR using different techniques (Abrego et al., 2014; Benito et al., 2015; Redoute Minziere et al., 2020). The disadvantage of this approach is that the recovered quantities are approximately divided by half for each type of GSR, with the risk of falling below the limit of detection of the instrument used for the analysis. Instead of dividing one specimen, collection of several specimens in sequence (ENFSI, 2017) or on different areas of interest has also been suggested as an alternative and may present potential if I- and OGSR transfer and persistence mechanisms were sufficiently different. However, current knowledge on the transfer and persistence of GSR does not support this hypothesis (see section 3.1 and 3.2).

- One specimen is collected and analysed in sequence (Gandy et al., 2018; Goudsmits et al., 2019; Morelato et al., 2012; Tarifa & Almirall, 2015; Zeichner & Eldar, 2004). In this approach, a good recovery was generally observed for the type of GSR analysed first, while a loss was observed for the type of GSR analysed subsequently (Bonnar et al., 2020; Redoute Minziere et al., 2020; Taudte et al., 2016). If a priority is given to one type of GSR or if the loss can be minimised, this represents an interesting approach.

- One specimen is collected and analysed simultaneously using a single technique for both types of GSR (Bell & Feeney, 2019; Feeney et al., 2021; Morales & Vazquez, 2004; Ott et al., 2020).

While this is theoretically the best approach (at least in terms of efficiency), none of the analytical techniques tested has adequate LODs for both types of GSR yet.

The selected combined analysis approach should maximise the recovery of both IGSR and OGSR and allow reanalysis of the specimen. According to available data, the sequential analysis of one specimen for both types of residues is the most adequate approach for maximum recovery of GSR. It is even possible to recover OGSR from a stub while preserving the quantity and distribution of the IGSR particles on the stub for subsequent analysis and storage (Bonnar et al., 2020; Redouté Minzière & Weyermann, 2021).

# 5. INTERPRETATION

The GSR trace can contribute to the reconstruction of events (Baechler et al., 2020; Kind, 1994) or it can be used as evidence in a trial (ANZPAA-NFIS, 2017; ENFSI, 2010). While some methods can be directly applied on site (see section 4.2), this remains relatively rare in practice. GSR specimens are often collected by forensic technicians or police investigators and sent to the laboratory for analysis (mainly using SEM-EDX as indicated in section 4.2). Several weeks can occur between the collection and the analysis of the GSR specimens, thus only visible (or easily enhanced) traces can generally be used in the early crime scene investigation. The next sections will describe how GSR can contribute as a clue in the investigation and as a proof in Court.



## 5.1. Investigation (the problem to find)

During the investigation of a firearm related event, the forensic scientists search for relevant traces and contribute to the formulation of hypotheses about the activities and involved persons with the other investigators (Baechler et al., 2020; Kind, 1994). It was proposed to follow an hypothetico-deductive reasoning cycle to find relevant traces (deductive inferences) and formulate hypotheses/propositions[4] about the event (abductive inferences) (Crispino, 2008; Ribaux & Caneppele, 2017; Ribaux & Talbot

[4] More information about the use of the term « hypotheses » and « proposition » in forensic science is given in the Sydney Declaration (Roux et al., 2022).

Wright, 2014; Roux et al., 2012). At this stage, all possible propositions should be considered and tested according to the available knowledge (e.g., case information as well as trace fundamentals from section 3), and when necessary, through case-specific experimentation.

As mentioned, knowledge about the transfer, persistence and prevalence of GSR is very important at this stage, but generally lacking in availability and representativity (Cadola et al., 2021). Indeed, as reported in section 3, it is not possible to collect comprehensive data for all singular cases (Crispino et al., 2021). Many factors will influence the amount of transferred GSR and their subsequent persistence (see Figure 3), such as the type of firearm, the ammunition, the discharge conditions (e.g., number of shots, environmental and weather conditions, …), the targeted zones for collections (e.g., location relative to the firearm, type of surface, orientation of the surface, …) or the activities after the discharge (e.g., handwashing, typing, running, …).

When investigating a suspicious death, four main propositions can be considered: suicide, accident, homicide, or natural death. If we consider only the suicide and homicide, clues about the intervention of a third party can be useful to differentiate these alternatives. In this case, GSR is believed to help, as more GSR are expected on the hand(s) of a shooter than on the hand(s) of a non-shooter. Thus, a high amount of GSR on the dominant hand of the deceased together with a nearby firearm generally supports the hypothesis of suicide rather than a homicide. Estimating the shooting distance is also useful in such cases. However, one has to remain careful, as relatives often discover and touch the deceased before calling the police (i.e., risk of secondary transfer and GSR displacement[5]). Moreover, firearm position and GSR amount can significantly vary even in undisturbed suicide cases (Lucas, Cook, et al., 2016; Molina, Castorena, et al., 2007; Molina, Martinez, et al., 2007; Vachon & Martinez, 2019). Increasing the knowledge on transfer, persistence, and prevalence of GSR, considering also unusual cases, is particularly useful to support the investigation. Such knowledge can then also feed the evaluation of the GSR trace for subsequent stages (e.g., trial).

Once the hypothetico-deductive reasoning cycle has allowed refuting some propositions and ideally strengthen the confidence in one set of mutually exclusive propositions (e.g., homicide vs. suicide), the

[5] In a case known to the authors, a close friend held the deceased when she discovered the lifeless body (before the police arrived on the scene), thus displacing the firearm and contaminating herself with traces (i.e., blood, GSR).

investigation moves to the next stage (Baechler et al., 2020; Kind, 1994). In general, if no clue about the intervention of a third party was gathered (i.e., the suicide proposition is supported rather than the alternative), then the investigation stops at the reconstruction of the event. On the other hand, if the proposition about the intervention of a third party is retained and a person of interest has been identified, the investigation moves to the evaluation stage (Kind, 1994).

### 5.2. Evaluation (the problem to prove)

Following analysis, the forensic laboratories generally submit their report following standardised protocols (e.g., ASTM, ENFSI, NIST, OSAC) (ASTM International, 2020; Niewoehner et al., 2003; OSAC, 2020; SWGGSR, 2011). Three classes of inorganic particles have been defined by the ASTM:

- Characteristic particles (composition: PbBaSb, TiZnGd or GaCuSn)
- Consistent particles (composition: PbBa, PbSb, PbBaCaSi, BaCaSi, BaAl, BaSb, TiZn or Sr)
- Commonly associated with GSR particles (composition: Pb, Ba or Sb)

Characteristic particles are the most specific to IGSR as they are rarely encountered in the environment and are thus, strongly associated with the discharge of a firearm (Seyfang, Lucas, Redman, et al., 2019; Wolten et al., 1977). On the other hand, commonly associated with GSR particles have many alternative sources in the environment and are, therefore less specific. Based on the number of each particle detected on the stub (and their morphology), the results can be interpreted from a case-specific forensic perspective.

Depending on the composition of the ammunition (see section 2), other specific particles were suggested in the literature. For example, Bender et al. (2021) proposed a new classification to distinguish TiZn particles generated by lead-free ammunition from alternative sources of TiZn in the environment. Moreover, particles of glass elements (e.g., Al, O, Si, Na) combined with heavy metal elements (e.g., Pb, Ba, Sb) were also reported as rarely found in the environment (Seyfang, Lucas, Popelka-Filcoff, et al., 2019; Seyfang, Lucas, Redman, et al., 2019).

Guidelines have been proposed to adequately address this evaluative stage (ANZPAA-NFIS, 2017; ENFSI, 2010). The GSR trace (e.g., ***number and type of detected IGSR particles on the hands***) needs to be evaluated under two mutually exclusive propositions (Benzaquen et al., 2020; Biedermann et al., 2009, 2011; Cardinetti et al., 2006; Kaplan Damary et al., 2016; Maitre et al., 2022; Maitre et al., 2017; Romolo & Margot, 2001). For example, propositions can be formulated at the *source* level:

- Proposition: The trace is a GSR (i.e., it originates from a firearm discharge)
- Alternative: The trace is not a GSR (i.e., it originates from another source)

Addressing the source level requires data about the composition of GSR (as proposed in the classification discussed above) as well as knowledge about alternative sources of GSR-like components (e.g., welding, fireworks, motor oil, airbags). Some studies address a more specific source level with the aim to associate a GSR composition to a specific ammunition (Brozek-Mucha & Jankowicz, 2001; Gallidabino et al., 2019; Rijnders et al., 2010).

As the presence of GSR on a person of interest may be explained by legitimate activities or non-firearm sources, it is advised to address the activity level rather than the source level when considering "trace evidence[6]" (Buzzini et al., 2019; ENFSI, 2010; Margot, 2017; Roux et al., 2015). Thus, propositions can also be formulated at the *activity* level:

- Proposition: The person of interest has discharged a firearm.
- Alternative: The person of interest has not discharged a firearm, the GSR comes from another activity.

Addressing the activity level requires information on the time since the discharge and all factors influencing the transfer and persistence of GSR. Knowledge about the prevalence (background level) of the GSR trace also needs to be considered, as well as the recent activities and professional occupation of the person (to account for the potential risk of contamination). In addition, alternative activities can explain the presence of GSR and may need to be considered in the evaluation (depending on the context of the case) such as:

- Manipulation of the firearm by the POI (for example when discovering the scene),
- POI close to the discharge (but not having touched the firearm itself),

[6] The term « trace » is often used in English to refer to the « small size of materials» (see Buzzini et al. 2019). Thus, « trace evidence » is used to refer to the GSR trace. In this article, we generally use another meaning of the word « trace » (see Margot, 2017).

- POI arrested by a contaminated police officer.

Ideally, data corresponding to the specific conditions of the case (firearm, ammunition, …) should be used to evaluate the probability of finding the observed GSR (E) under both propositions. This allows the estimation of a likelihood ratio (Benzaquen et al., 2020; Biedermann et al., 2009, 2011; Cardinetti et al., 2006; Kaplan Damary et al., 2016; Maitre et al., 2022; Maitre et al., 2017). An example of required (raw) data is presented in Figure 7 for an OGSR molecule based on available literature (Gassner et al., 2016; Hofstetter et al., 2017; Maitre et al., 2018; Manganelli et al., 2019).

A first perusal of the simulated data indicates that there is:

1) some overlap between the distributions of the data (i.e., the initial transfer of GSR can be low in some instances, while prevalence can occasionally be relatively high without explanations);

2) more overlap between the distributions when longer times occur between the discharge and the collected specimen can be expected.

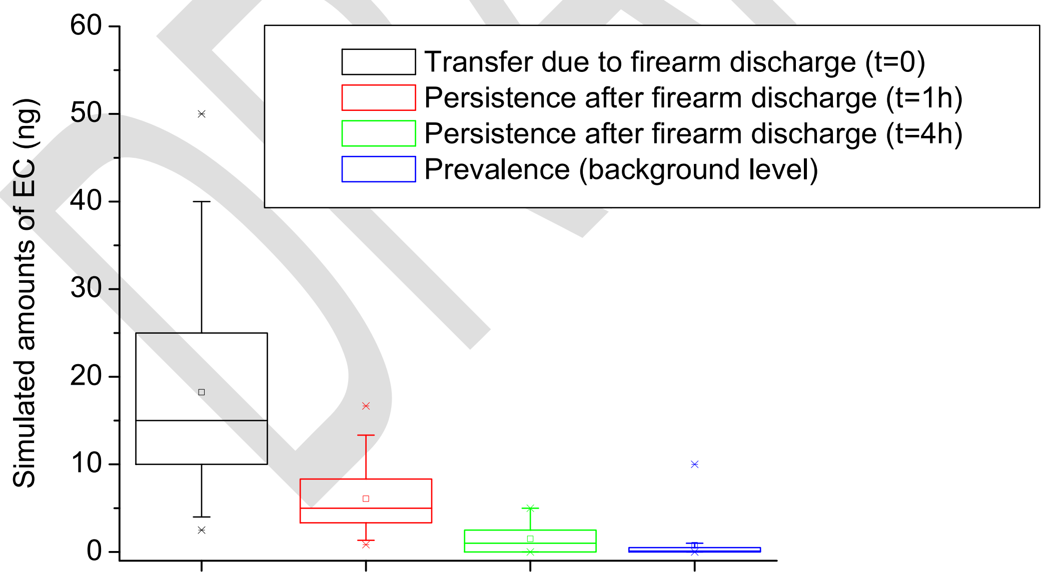


**Figure 7:** Simulated data for the transfer (t=0h), the persistence (t= 1 and 4h) and prevalence of the stabiliser, the ethylcentralite (EC), on the hands of shooters and non-shooters based on available literature on the subject (Gassner et al., 2016; Hofstetter et al., 2017; Maitre et al., 2018; Manganelli et al., 2019).

In practice, the probative value will increase if the information from several target components is combined (i.e., multivariate approaches). However, multivariate statistical models also increase in complexity, thus holistic approaches combining data from different traces (e.g., DNA, shoemarks, reconstructed trajectories of bullets, GSR) may be efficient alternatives (Crispino et al., 2021; Roux et al., 2015; Weyermann & Roux, 2021). Regarding the GSR trace, qualitative and quantitative distribution of inorganic particles and organic compounds found on different surfaces (e.g., hands, hair, clothes) and times after the discharge present important information to infer about alternative hypotheses (ANZPAA-NFIS, 2017; ENFSI, 2010).

# 6. CONCLUSION

While GSR continuously attracted interest and research over the years (Blakey et al., 2018; Charles et al., 2020; Feeney et al., 2020; Maitre et al., 2017), it mainly focused on the technical aspects rather than the trace itself (Cadola et al., 2021; Roux et al., 2021; Séguin et al., 2021; Sobreira et al., 2020; Vachon & Martinez, 2019). This may be due to several reasons, among them the fact that GSR research requires regulated material and infrastructure not easily available in all laboratories (e.g., firearms, ammunitions and shooting range). Moreover, as for many types of traces, GSR transfer, and persistence studies are much more complex and time consuming than the testing of new technological approaches on reference substances and materials. This should not divert forensic scientists from the purposes, and the need to increase forensic basic knowledge before addressing technical and organisational challenges (Roux et al., 2022).

Taking this into account, this overview identified the following main research challenges in order to improve the GSR investigative and evidential value:

- **Trace transfer**: In order to evaluate how GSR transfer (what, where and how much), it is important to ensure that recovery approaches are reproducible and allow the collection of a maximum of GSR particles. Some researchers have suggested that reproducing GSR-like material (proxy) would greatly help the field (Menking-Hoggatt et al., 2021; Sobreira et al., 2020). For example, several studies have proposed to produce tailor-made microparticles or to create synthetic GSR particles for proficiency tests and research experiments (ENFSI, 2017; Menking-Hoggatt et al., 2021; Niewoehner, 2005; Niewoehner et al., 2003). The second challenge is the infinite number of parameters that need to be tested to study the transfer of GSR, such as used material (e.g., firearm and ammunition), transfer conditions (e.g., airflow, humidity, distance to the firearm openings) and the surfaces targeted for collection (e.g., hands, face, hair, nostrils, clothes, surrounding objects). All these parameters must also be tested for subsequent transfer studies (secondary, tertiary). Thus, a more case-specific approach may be more efficient to collect (and increase) data for specific cases and conditions (Benzaquen et al., 2020). It is also important to consider the risk of pollution from the police intervention and investigation.

- **Trace persistence**: Persistence studies present the same challenges as transfer studies with the additional “time” variable. This explains why few research investigated persistence over 4 hours. The multitude of activities and contacts occurring between the discharge event and GSR collection increase the complexity of the experimental design. However, these are essential to correctly evaluate the meaning of GSR in a practical case. Research indicates that in most situations, the loss is rapid on the hands of a shooter (a few hours), but more research would be useful to orient how long after the event and where samples should be collected.

- **Trace prevalence**: Without background studies, the GSR trace cannot be correctly evaluated either. While prevalence studies are easier to carry out as no firearm, ammunition or shooting range are required, it is important to use the same collection and analysis protocols as transfer and persistence studies. For example, the results obtained for GSR collected on the hands using a stub cannot be compared with those obtained with a swab on the hair. While large studies were carried out for IGSR, more data is needed on OGSR and combined GSR prevalence.

- **Trace evaluation**: with a few exceptions, GSR interpretation still mainly focused on the type of detected particles and their specificity (ASTM International, 2020). Such guidelines are useful but remain very technical without consideration about quantities, localisation and context specific information (Romolo & Margot, 2001). While multivariate statistical approaches may contribute to the interpretation of the GSR trace, holistic interpretation approaches should also be considered for a better integration of the GSR trace with other available clues and information in caseworks (Roux et al., 2022; Roux et al., 2015; Weyermann & Roux, 2021).

- **Technical considerations**: Finally, on the more technical aspects, while SEM-EDX has been the method of choice for IGSR analysis, it does not mean that less costly methods such as LIBS or XRF, may not be an interesting alternative in practice (Almog, 2006; Berendes et al., 2006; Lopez-Lopez et al., 2017; Postek et al., 2010). Organisational habits should not hamper forensic development. The same observation can be made about the forensic potential of OGSR, as a complementary or alternative source of information to IGSR in practice (Roux et al., 2021). Similarly, increasingly complex statistical modelling (including AI approaches) will not entirely solve the root forensic issues (i.e., the case-specific nature of the trace) (Roux et al., 2022). Simplified and holistic models should also be considered to promote a better understanding of the trace by the different stakeholders of the justice system.

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