



An investigation on the secondary transfer of organic gunshot residues

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ABSTRACT

Gunshot residues (GSR) are an important forensic trace in firearm-related events. Currently, routine GSR analyses focus on the detection and characterisation of the inorganic components (IGSR). The increasing prevalence of heavy metal-free ammunition challenges these current protocols and there is an increasing interest in how the organic components of GSR (OGSR) can provide complementary information. Similar to the situation with IGSR, OGSR compounds originally deposited on the shooter during the firing process may further be transferred onto another individual or surface. Hence, the aim of this study was to provide additional information regarding the risk of a secondary transfer of OGSR. Two scenarios were investigated, the first one related to the arrest process and the possibilities of a secondary transfer arising between a shooter onto a non-shooter (e.g. between a police officer and a person of interest (POI)). The second scenario concerned the transfer of OGSR onto the non-shooter after handling a firearm for few minutes without discharging it. One calibre was chosen, the .40 S&W calibre, used by several Australian State police forces. A secondary transfer was observed in all cases for the two scenarios investigated, for three compounds of interest: ethylcentralite (EC), diphenylamine (DPA), *N*-nitrosodiphenylamine (*N*-nDPA). The firearm handling scenario resulted in a larger secondary transfer to that of the arrest scenario. Overall, the amounts of OGSR detected on the non-shooter were generally lower than that detected on the shooter and controls after the arrest scenario. The results of this study provide complementary knowledge about OGSR, which can be further used to improve the current practice and the interpretation of OGSR evidence. In particular, it highlights that the secondary transfer proposition must be considered during the interpretation of forensic findings, especially when small amounts of OGSR target compounds are detected.

1. Introduction

Gunshot residues (GSR) are an essential trace in firearm-related events. In practice, GSR analyses currently focus on the detection and characterisation of the inorganic components (IGSR), which are mainly particles containing some or all of lead, barium and antimony elements known to be associated with the primer mixture. IGSR protocols are challenged by the increasing probability of encountering casework involving heavy metal-free (HMF) ammunition [1]. Furthermore, given the quantity of propellant present in firearm cartridges, it is reasonable to expect that substantial traces of OGSR may be deposited during a shooting. This may provide GSR examiners with an opportunity to obtain information that is complimentary to IGSR examination, potentially enhancing the forensic evaluation process in regards to a shooting. This necessitates further research into improving our

knowledge of OGSR generated through the combustion of propellant powder, its characterisation, its transfer and persistence. Several studies already considered the question of OGSR analysis [2–9] and this study focused on the secondary transfer, beyond such analytical aspects.

While the primary transfer of IGSR and OGSR to the shooter occurs during and after the firing process, it is also important to assess further transfer of these residues from the shooter to an uncontaminated individual. Indeed, detecting GSR on an individual does not confirm the person has discharged a firearm [10,11]. Secondary transfer of GSR traces can happen when, for instance, a non-shooter makes contact with a firearm (without discharging it) or with another individual who recently discharged a firearm.

Several studies have approached secondary transfer of IGSR. Charles and Geusens [12] investigated the secondary transfer of inorganic particles from police officers onto non-shooters. They concluded that

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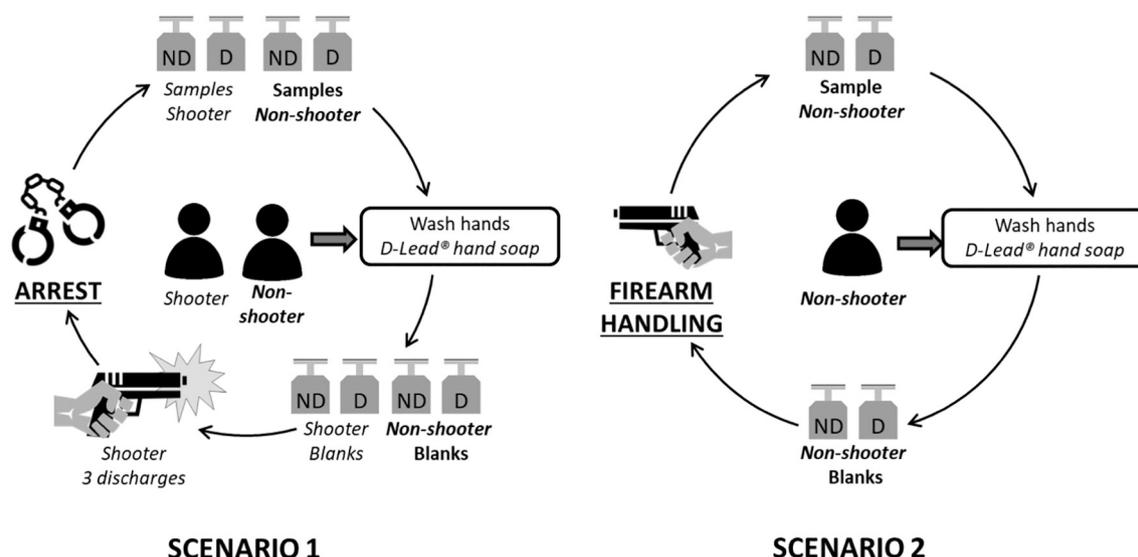


Fig. 1. The sampling procedure of the secondary transfer experiments for both scenario 1 and 2 ($n = 5$). D = dominant hand, ND = Non-dominant hand.

the risk of transfer is heavily dependent of the technique of arrest [12]; the more vigorous the arrest, the higher is the risk of transferring particles from one surface to another. They also emphasised that special unit forces officers, who have a higher degree of contact with firearms, transferred larger quantities of GSR [12]. French et al. [13] examined the secondary transfer of IGSR particles via a handshake between a shooter and non-shooter, as well as during the exchange of a firearm between a shooter and non-shooter [13]. They concluded that the average number of particles transferred via handshakes was high (> 80 characteristic particles in average) while an exchange of firearm resulted in a lower number of particles (40 characteristic particles in average) transferred [13]. The experiments were carried out immediately after the firearm was discharged, resulting in the maximum amount of GSR being present on the shooter and the firearm [13]. The study by French et al. was further extended to explore tertiary transfer by performing two consecutive handshakes [14]. A reduction in inorganic particle transfer was observed, however IGSR particles were still detectable following the tertiary transfer scenario [14]. Similar observations were made by Girvan et al. [15], where the transfer of inorganic particles during the arrest of a non-shooter by a police officer who had previously discharged a firearm was studied [15]. It was found that particles were detectable from specimens taken from the hands of the non-shooter [15].

Studies into the secondary transfer of OGSR, however, are limited. Arndt et al. [16] investigated the secondary transfer of OGSR after a handshake between the shooter and non-shooter [16]. The specimens were analysed by Ion Mobility Spectrometry (IMS) with no OGSR detected. However, several limitations, including a high limit of detection of the instrumentation, were highlighted, which could have contributed to this results [16]. Gassner et al. investigated three different scenarios just after discharge involving handshakes, transporting a firearm and arrests [17]. The secondary transfer of OGSR was observed for all scenarios when a more sensitive analytical method, UHPLC-QqQ-MS, was utilised. The firearm transportation scenario resulted in the lowest amount transferred, followed by handshaking with the arrest scenario resulting in highest amount of OGSR transferred [17]. These studies emphasised that the risk of a secondary transfer of both IGSR and OGSR is significant. Therefore, precautions are essential to avoid a transfer of GSR when a contact between police officers and non-shooters occurs and the possibility for secondary transfer of OGSR and/or IGSR must be considered when GSR case findings are evaluated.

The aim of the current study was to extend the body of knowledge regarding the secondary transfer of OGSR as only two studies tackled

such questions, and only one analysed specimens with a highly sensitive instrument. This focuses on an Australian perspective. The current ammunition used by Australian police forces is the calibre .40 S&W, which has never been studied in the context of secondary transfer. Generally, police officer(s) carry their service firearms while on duty and may potentially come in contact with non-shooter(s) during police investigations and operations. It is, therefore, essential to assess the degree of transfer that could potentially take place if physical altercations such as an arrest procedure occurs between police officer(s) and non-shooter(s). This study considered two scenarios: the first one related to an arrest scenario between the shooter (i.e. police officer) and a non-shooter (i.e. a 'clean' POI).

The aim of this first scenario was to study the potential pollution of POI by police officers during the arrest process. According to Schwendener et al. [18], "pollution" refers to the addition of some material due to a lack of precautions, while "contamination" refers to non-pertinent traces present before any investigation and cannot be avoided [18]. Pollution is therefore more suitable in the context of a secondary transfer study.

The second scenario involved a non-shooter handling a firearm without discharging it. The aim of the second scenario was to determine the amount of OGSR transferred by handling the firearm when compared to a person who discharged it. Four compounds of interest, known to be part of propellant powder and OGSR composition, were investigated: ethylcentralite (EC), methylcentralite (MC), diphenylamine (DPA) and *N*-nitrosodiphenylamine (*N*-nDPA) [19–21].

2. Material and method

2.1. Secondary transfer: shooting experiment procedure

For this study, one authorised officer in an Australian State police force facility performed the firearm discharges in an indoor shooting range. The different transfer scenarios were performed outside of the firing range to minimise the risk of polluting the hands of the non-shooter from the environment. One calibre and firearm was selected: A Glock 22® calibre .40 S&W. This was chosen as it is the service weapon and calibre of a number of Australian State police forces. The lead-free Winchester WinClean® (180Gr. Brass Enclosed Base) ammunition was chosen. The thumb-forefinger part of the palm and back of both dominant and non-dominant hand (right and left respectively) as well as the wrists were sampled separately.

The method for both scenarios is presented in Fig. 1. This involved

both participants (one shooter and one non-shooter) thoroughly washing their hands prior to the blanks being collected. The next step required the shooter to discharge three rounds of ammunition with the Glock® 22 held with two hands. To avoid pollution, the non-shooter did not enter the range at any point during the scenario. Following the firearm discharge, the shooter left the firing range and conducted the arrest scenario: the non-shooter put their hands on the top of their head before the shooter gripped the hands of the non-shooter and put them behind their back before they were handcuffed (Fig. 1, scenario 1). The handcuffing procedure simulated a typical arrest procedure. The non-shooter was also asked to resist the arrest during the handcuffing process in order to simulate a realistic scenario.

After 1–2 min, the shooter removed the handcuffs and the specimens were collected from both hands of the non-shooter, as well as the shooter. The collection of OGSR was performed with GSR stubs (Ted Pella Inc., USA). The thumb-forefinger region of the palm and back of the hand, as well as the wrist, were thoroughly sampled until the stub surface was no longer sticky [21–23]. Each hand was sampled using an individual stub with the collected specimens packaged separately by sealing with the cover and placed in their respective boxes. The specimens were stored at 4 °C until extraction. The extraction was performed within 24 h of collection to avoid degradation of the specimens [24].

For the second scenario, the non-shooter washed their hands before blanks were taken outside the firing range while the shooter discharged the firearm three times. The non-shooter then picked up, held and handled the firearm with both hands for 5–10 min, without discharging it (Fig. 1, scenario 2). Specimens from both hands were collected after the time elapsed as described previously. Both scenarios presented in Fig. 1 were repeated in quintuplicate. Controls specimens were also collected. These specimens were collected from the same shooter immediately after having discharged three rounds of the same ammunition with the same firearm. The shooter did not make contact with any other surfaces. These control specimens can be reasonably expected to contain a greater amount of OGSR because of the absence of potential losses due to further activities. Consequently, control specimens represent, on average, the maximum amount of OGSR detected from the hands of the shooter.

2.2. Analytical method: OGSR standards and UPLC-QqQ-MS conditions

Four target compound standards were used in this study: EC, MC, DPA and N-nDPA (Table 1). These standards were used for identification of compounds and analytical method validation purposes, presented in a previous publication [21]. Additionally, a 5 point calibration standard curve (0.01–1 ppm) was prepared and analysed with every run of specimens for quality control purposes.

d10-DPA has been previously utilised as a suitable internal standard (IS) [21,25]. Stock solutions of internal standard were prepared at a concentration 1000 µg/mL in methanol:acetonitrile (1:1) v/v and added to each specimen at a final concentration of 20 ppm [19]. Analysis of specimens was conducted per Maitre et al. [21]. The method was validated using the ICH Guidelines [26], and found fit for OGSR qualitative analysis purposes. The specimens were extracted using the protocol described in Taudte et al. [19,21,27].

The chromatographic separation was performed on a Waters UPLC

Table 1
Compounds of interest. IS = internal standard.

Compounds	Provider	Concentration	Solvent
EC	Novachem Pty Ltd	100 µg/mL	Methanol Acetonitrile (1:1)
MC		100 µg/mL	Methanol Acetonitrile (1:1)
DPA		1000 µg/mL	Methanol
N-nDPA		1000 µg/mL	Methanol
d10-DPA (IS)	C.D.N Isotopes Inc.	Solid	–

ACQUITY® system. An Agilent ZORBAX RRHD Eclipse XDB 80 Å C18, 3.0 × 100 mm, 1.8 µm was used coupled to a ZORBAX Eclipse XDB 80 Å C18, 3.0 × 5 mm, 1.8 µm UHPLC guard. The mobile phase used is presented in Table 2, both solvents were filtered through 0.2 µm membrane filters (Table 2). The starting conditions of the chromatographic method is 70% water and 30% methanol. The method used a gradient which includes a 4.6% increase of methanol per minute for 12 min [19,21], followed by 5 min of flushing and equilibrium before the next run. The column temperature was thermostatically maintained at 43 °C and an injection volume of 2 µL was used throughout.

The QqQ-MS conditions were as follow: the desolvation temperature was set at 250 °C and the detection was performed using Multi-Reaction-Monitoring (MRM) from 0 to 12 min with electrospray ionisation (ESI) source set at 140 °C [21].

The results were extracted and processed in order to provide a normalised response: the detected peaks were integrated using QuanLynx® (Waters software). The presence of precursors to both product ion transitions was a required condition in an abundance above the limits of detection (LOD) for considering the compounds as present. The detected compounds underwent a blank subtraction (hand blank collected before each experiment) before being normalised to the internal standard (IS). The square root of the ratio was then calculated and represent the normalised peak area [21,28,29].

3. Results and discussion

During this study, calibration curves were analysed along with the specimens and showed consistent and stable responses across each analytical analysis with the calibration curves found to be linear during each analytical run (average $R^2 > 0.99$).

All the targeted compounds were considered detected when the abundance was found above the limit of detection (LOD). The LODs were found as follow: N-nDPA: 5.64×10^{-3} ppm,

MC: 1.75×10^{-4} ppm, DPA: 2.09×10^{-3} ppm and EC: 3.82×10^{-4} ppm [21]. Blank subtractions were performed on every specimen from each of the shooter and the non-shooter to account for possible contaminations. The blank specimens arising from the non-shooter were found to be clean, i.e. with none of the OGSR compounds detected. However, it was surprising that even after hand washing, some blanks from the shooter were found to contain OGSR. Hand washing has previously been studied [16] and indicated a complete removal of OGSR traces. The difference in results might arise from the use of different soap and sanitisers. Arndt et al. have studied a different soap containing alcohol [16], which might be more efficient in the removal of OGSR. In this study the soap used was a D-lead® hand soap used to decontaminate the hands of heavy metals; such soap might not be sufficient enough to remove OGSR. In addition, different instrumentation was used, which differ in sensitivity [16]. Arndt et al. have analysed specimens by IMS which typically produces high LODs [16]. Conversely, highly sensitive UPLC-QqQ-MS instrumentation was used in this study, and it was found that hand washing was not sufficient to remove completely OGSR traces. If the hand blank was found to contain a larger amount of OGSR than that of its corresponding specimen from the experimentation, that particular specimen was considered negative for OGSR traces to avoid skewing the results.

3.1. Scenario 1: arrest process

During their duties, police officers may come into contact with a potential POI for a particular investigation. When it occurs, there is a question of potential pollution of the non-shooter by the police officer. In order to assess this, an arrest scenario was performed and investigated. The results from the UPLC-QqQ-MS analysis are reported as a normalised response (Section 2.2) [21]. Three of the four target compounds were detected (EC, DPA and N-nDPA). The manufacture of propellant powder typically involves the addition of a single centralite,

Table 2
UPLC mobile phase composition.

Mobile phase components	Solvent grade	Additive	Membrane filters
Methanol ultrapure Milli-Q® Water	Hypergrad Lichrosolv®, Merck KGaA 18.2 MΩcm, Q-POD®, Merck KGaA	+ 0.1% (v/v) formic acid	0.2 μm PTFE (47 mm, Advantec) 0.2 μm mixed cellulose ester (47 mm, Advantec)

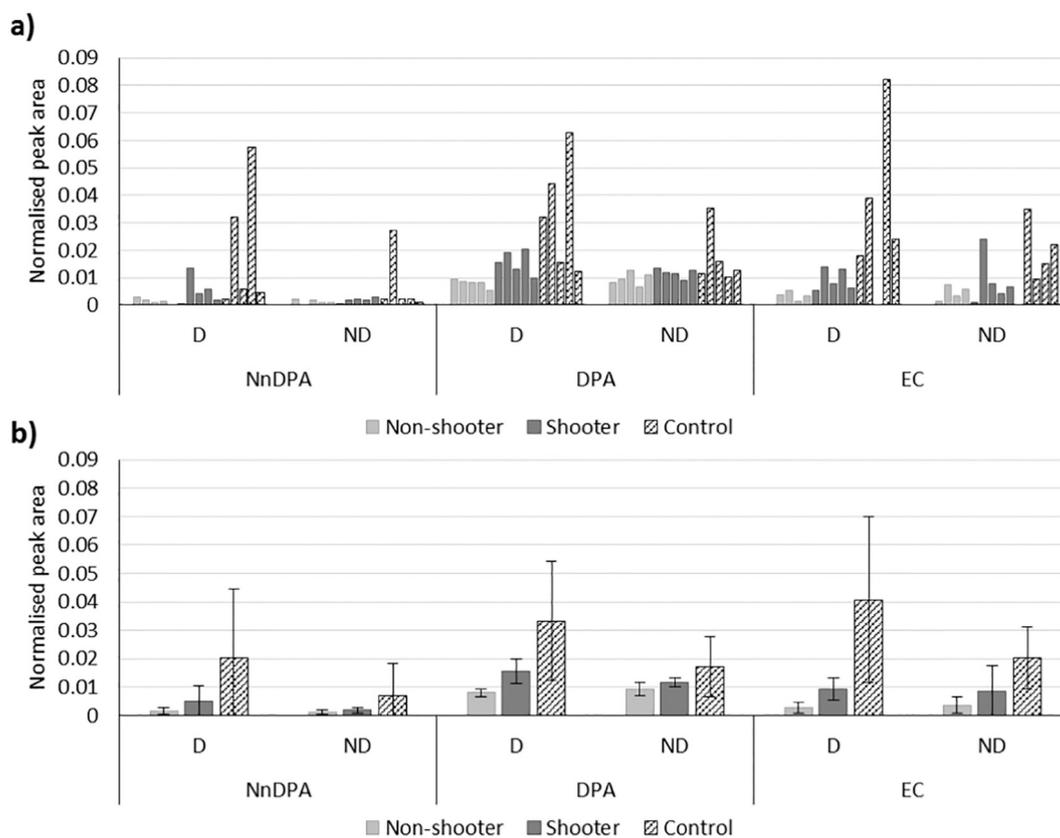


Fig. 2. Scenario 1: Arrest process. Level of OGSR detected (normalised response) from both hands of each participant. D = dominant hand, ND = Non-dominant hand. a) Each replicates separately ($n = 5$); b) Averaged amount; the error bars represent the standard deviation. The “non-shooter” represents the individual being arrested who did not have any contact with the firearm; the “Shooter” is the police officer who arrested the non-shooter after discharging the firearm. “Controls” represent the amount detected from the shooter immediately after three discharges, without having entered in contact with any other surfaces.

either EC or MC but rarely both. Fig. 2 represents the amount detected (normalised response) for the three compounds of interest collected from both hands (dominant and non-dominant) of the shooter and non-shooter in the context of the arrest scenario. Fig. 2a represents the details of the replicates, each bar-plot represents the amount detected on each hand. Fig. 2b represents the averaged amount detected from each set of replicate specimens.

It was observed that the three compounds of interest were successfully identified in each of the five replicates of the arrest scenario. The amount detected of these compounds followed the same trend across each of the dominant and non-dominant hands, with a lower amount detected on the latter when compared to the dominant hand. As expected, the largest amount was detected in the control (Fig. 2a and b), which relates to the detection of OGSR on the hands of the shooter immediately after the firearm discharges. The second largest amounts detected were from specimens arising from the hands of the shooter after they arrested the non-shooter. Finally, a secondary transfer was observed for the three compounds of interest on the non-shooter after being handcuffed by the shooter (Fig. 2). The non-shooter did not have any prior contact with any potential source of OGSR, and their blanks were found to be free of OGSR. This emphasises the fact that the OGSR found on the non-shooter arose primarily from the transfer of OGSR

from the shooter during the arrest. It must be noted that the wrists were also sampled. Hence, the handcuffs might also have contributed to the secondary transfer of OGSR onto the hands of the non-shooter in addition to the transfer from the shooter during the duration of the arrest process.

The results observed in Fig. 2b on the hands of the shooter also suggest the presence of a secondary transfer as the amount detected on the hands of the shooter is lower than the amount detected in the controls. The only difference between the control specimens and the shooter specimens was the arrest scenario. Hence, the responses detected, for each of the compounds on both hands of the shooter, were found to be lower than the controls. Such a result is likely due to the transfer of OGSR off the hands of the shooter onto the hands of the non-shooter while performing the arrest simulations. However, losses to the environment or redistribution of the OGSR onto surfaces that were not collected may also have contributed to the results. An exception is regarding DPA on the non-dominant hand (Fig. 2), where it can be observed that adding together the amounts of DPA on the shooter and non-shooter resulted in a larger amount than the control. Such a result may be the consequence of the high variability observed between specimens as illustrated by the error bars in Fig. 2b.

Different trends were observed between the non-shooter when

compared to the shooter and the controls when comparing the dominant and non-dominant hand (Fig. 2b). It was found that the detection of compounds was more consistent between both hands on the non-shooter while larger differences between hands was seen in the shooter and control specimens. Greater amounts of OGSR were detected on the dominant hand of the shooter and control when compared to the amount detected on the non-dominant hand. When semi-automatic pistols are considered, the deposition of GSR is highly dependent on the make of firearm as that dictates relative positioning of the hands to the ejection port. In this case the ejection port of the firearm was on the right. As a result, a greater amount of GSR are deposited on the right hand if that is the dominant hand [23]. The difference in the amount is consequently highly due to the activity of holding and discharging a firearm. Whereas on the non-shooter, the result related to the handcuffing scenario for which both hands of the non-shooter were usually clutched in the same manner in order to put them behind the back. This led to a lower and more consistent level of OGSR being transferred across both hands of the non-shooter than that seen in the shooter and control specimens. The large variability was also observed in Fig. 2b (as demonstrated by the error bars) for both shooter and control. This variability reflects the numerous factors influencing the primary transfer of OGSR compounds such as the firearm, the ammunition, the plume dispersion at the muzzle and the ejection port, the shooter position, skin conditions and environmental conditions. Such variations were also observed previous studies [9,17,21].

Table 3 represents the percentage when the detected responses were normalised to the control. The controls represent the largest amount of residue available as they were collected immediately after discharging the firearm (100%). A large difference was observed between the specimens (Table 3), with the amount detected on the shooter post arrest ranging between 23% (EC, D) and 68% (DPA, ND). Conversely, that of the non-shooter ranged between 9% (N-nDPA, EC, D) up to 55% (DPA, ND).

When comparing the overall averages (Table 3), it was observed that after the arrest scenario an amount corresponding to 39% of the control were still observable from the shooter, while on the arrested non-shooter, the amount detected after secondary transfer was about 23%, suggesting that a pollution of a non-shooter from shooter who proceeded to the arrest was non-negligible.

Similar trends were observed by Gassner et al. [17] with detection of OGSR on a non-shooter after an arrest scenario. The study involved the use of a different calibre (9 mm Luger) with the arrest scenario conducted differently as the non-shooter was handcuffed on the ground and then helped to get back up. They identified that a considerable amount of OGSR (41.9% for N-nDPA) was transferred during an arrest simulation. As they sampled both hands on the same GSR stub [17], the results from the present study (e.g. D and ND) were summed together for comparison purposes, and a secondary transfer of 29.6% for N-nDPA was found. Considering the large variability observed between discharges, the difference in the observed secondary transfer is likely to arise from the simulations and the way the arrests were conducted. In the study by Gassner et al. [17], the fact that the non-shooter was lying down and helped to get back up may result in a longer and more vigorous contact, which may ultimately lead to a larger amount of OGSR being transferred.

Table 3

Scenario 1: Average percent of the level of OGSR detected when normalised to Control. D = dominant hand, ND = non-dominant hand.

	N-nDPA		DPA		EC		Averages
	D	ND	D	ND	D	ND	
Control	100%	100%	100%	100%	100%	100%	100%
Shooter	25.7%	27.3%	46.9%	68.1%	22.8%	42.5%	38.9%
Non-shooter	8.6%	21.0%	24.1%	55.4%	8.4%	22.4%	23.3%

3.2. Scenario 2: firearm handling

In Scenario 2, a firearm was discharged three times and then handled by a non-shooter for approximately 10 min without further discharged. The holding activity was not restricted and the non-shooter also manipulated the firearm by opening and closing the breech. In Fig. 3a and b, it was observed that N-nDPA, DPA and EC were successfully detected on the hands of the non-shooter, confirming that secondary transfer was possible through handling of the firearm, with OGSR detected in all 5 replicates. The amount detected was consistently lower than that of the control (Fig. 3a). Fig. 3b represents the average amount (normalised response) for each participant (non-shooter and shooter) and the error bars reflect the standard deviations.

After discharge, OGSR traces might deposit onto the body of the firearm, generating an accumulation of residues on its surface. Furthermore, if the firearm is typically not cleaned frequently there may be accumulation of GSR on its outer surfaces. The source of the OGSR traces detected on the non-shooter hands is, therefore, likely to arise from the OGSR background present on the firearm during the holding contact and manipulations. Consequently, the secondary transfer of OGSR when handling a firearm might be an important or even the main contributor to the deposition of OGSR on the non-dominant hand of a shooter.

It is interesting to observe that for N-nDPA on the non-dominant hand of the non-shooter (Fig. 3), handling the firearm resulted in a similar amount of OGSR being transferred to the amount detected in control specimens. As indicated above, due to the construction of the firearm, the position of the ejection port (on the right) and the position of the hands while discharging the firearm, the non-dominant hand is often less exposed to the GSR plume when firing a gun than the dominant hand. Therefore, a large portion of the total amount of OGSR transferred onto the non-dominant hand might arise from the contact with the grip of the firearm as it usually acts as a support when holding the firearm, or by more generally handling the firearm immediately prior to discharging it.

However, as the non-shooter held the firearm without any restriction, on occasions, the firearm was passed between the two hands exposing both the dominant and non-dominant hand to OGSR. This manipulation might have likely contributed to the secondary transfer on the non-dominant hand. Furthermore, the duration of the contact was longer during the scenario (approximately 10 min) to that of the controls, where the specimens were collected immediately after discharge. A prolonged contact would also impact the level of OGSR transfer onto the surface of the hands.

When comparing the trends, Table 4 illustrates the percentage of OGSR detected on both hands of the non-shooter normalised to the control. On the dominant hand, a range of 22%–35% was detected. While on the non-dominant hand, a wider range of 33%–65% for the three compounds was detected (Table 4). On average, an amount of 40%, when compared to the control specimens, was still detected after handling the firearm. The results emphasise that handling a firearm for several minutes, without discharging it, is sufficient to successfully transfer OGSR traces in a substantial amount. The discrepancies between the dominant and non-dominant hand observed in Table 4 arose from the normalisation to the control (Fig. 3). In the control (Fig. 3), a lower amount was detected on the non-dominant hand, which resulted, after the normalisation, in larger percentages.

Gassner et al. also observed a secondary transfer of OGSR by only touching the surface of a firearm when transporting it [17]. They observed that the amount detected after touching the firearm was lower than after the arrest, while in this study the opposite trend was observed. The different conditions of the firearm manipulation and the timeframe of the experiment is likely the source of the different trends observed between the two studies. Indeed, their experiment involved transporting a firearm without any further handling, whereas in the present study, the non-shooter was allowed to manipulate the firearm,

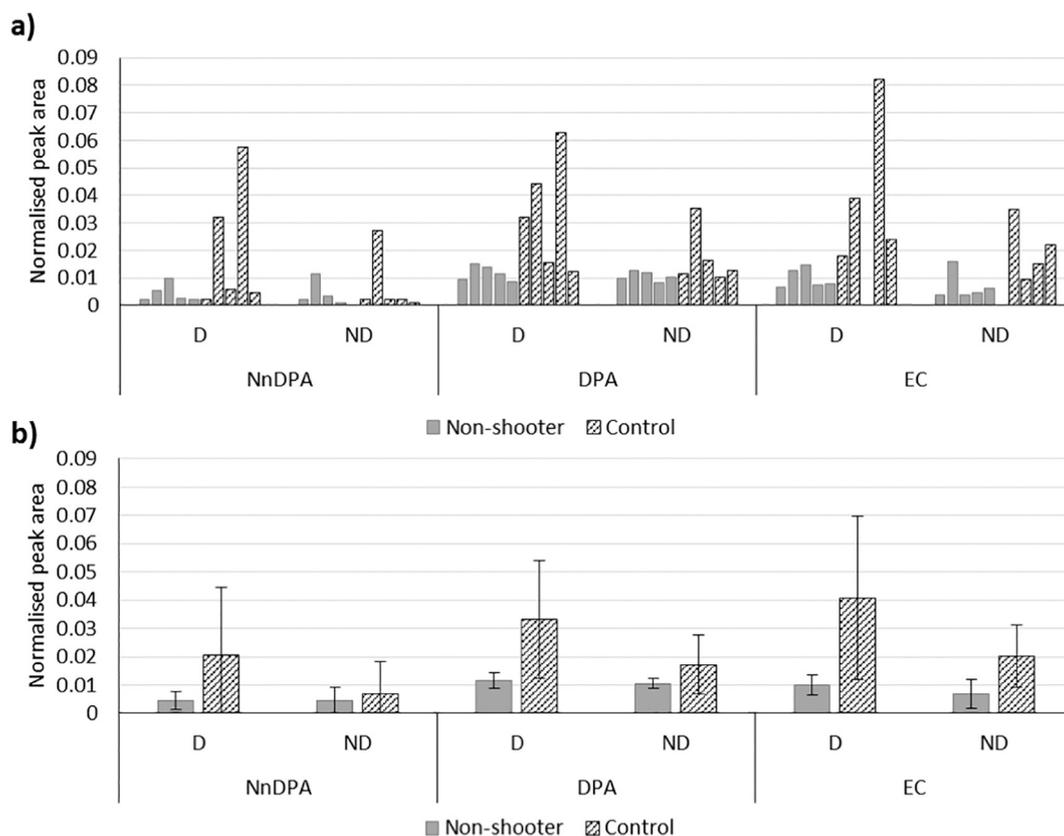


Fig. 3. Scenario 2: Firearm handling. Level of OGSR detected (normalised response) from both hands of each participant. D = dominant hand, ND = Non-dominant hand. a) Each replicates separately ($n = 5$); b) Averaged amount; the error bars represent the standard deviation. The “non-shooter” represents the individual who handled the firearm; “Controls” represent the amount detected from the shooter immediately after three discharges, without having entered in contact with any other surfaces.

Table 4
Scenario 2: average Percent of the level of OGSR detected when normalised to Control. D = dominant hand, ND = non-dominant hand.

	N-nDPA		DPA		EC		Average
	D	ND	D	ND	D	ND	
Control	100%	100%	100%	100%	100%	100%	100%
Non-shooter	21.7%	65.2%	35.1%	61.3%	24.1%	33.4%	40.2%

including opening and closing the breech. Opening and closing the breech provides access to additional sources of OGSR inside the weapon, leading to a potentially higher secondary transfer. The second substantial factor is the contact duration. In their study, contact was about 10 s [17], while here the non-shooter held the gun approximately 10 min. Such difference in the duration of the contact with the firearm may greatly influence the amount of OGSR transferred as a longer contact may result in a larger amount of OGSR transferred.

3.3. Scenario comparison: effect of different activities on OGSR traces

Fig. 4 illustrates the amount detected across the two scenarios performed in this study. It was found that a secondary transfer of OGSR occurred following both the arrest and firearm handling experiments, with similar trends observed across the three compounds detected. A substantial reduction of the response detected between the controls and the non-shooter after the arrest was observed (Fig. 4).

It was also observed that handling a firearm resulted in a higher degree of secondary transfer (Table 4, average of 40% observed) than being arrested by the shooter (Table 3, average of 23% observed). The

amount detected on the shooter after the arrest scenario is similar to the non-shooter after having handled a firearm (Fig. 4, Table 3 and Table 4, 38.9% and 40.2% respectively). Minor exceptions were seen for DPA (D) for which a larger response was observed on the shooter when compared to the non-shooter (firearm handling). The opposite results was observed with N-nDPA on the non-dominant hand, of which a difference of almost 38% was observed between the two scenarios (27% in Table 3 and 65% in Table 4). This is due to a high amount detected on the hand of the non-shooter after handling the gun and a low amount detected on the shooter after the arrest. These results emphasise the influence of the shooting process and the associated variability in the amount of OGSR recovered and detected. Finally, on average, the largest responses detected arose from specimens taken from the controls, which were taken from the shooter immediately after the firearm discharges and without any contact with any other surfaces.

In summary, the results of this study demonstrate that the risk of secondary transfer is substantial. From an investigation and interpretation perspective, it is recommended that precautions should be taken to minimise instances of secondary transfer to avoid false positive results. In an activity level context, a false positive results refer to the association of the detection of OGSR on the POI with a firearm discharge, while the actual origin of the traces was from an external and unrelated pollution (e.g. contact with a polluted officer). Because of the numerous factors involved in the formation, deposition and analysis of OGSR, as well as the unique sequence of activities that may lead to different extent of secondary transfer, it is essential to consider assessing every investigation by means of a case-by-case approach in order to ensure to interpret OGSR results in an appropriate manner in view of the circumstances surrounding the investigation [11,30].

The results suggest that standard practice should include

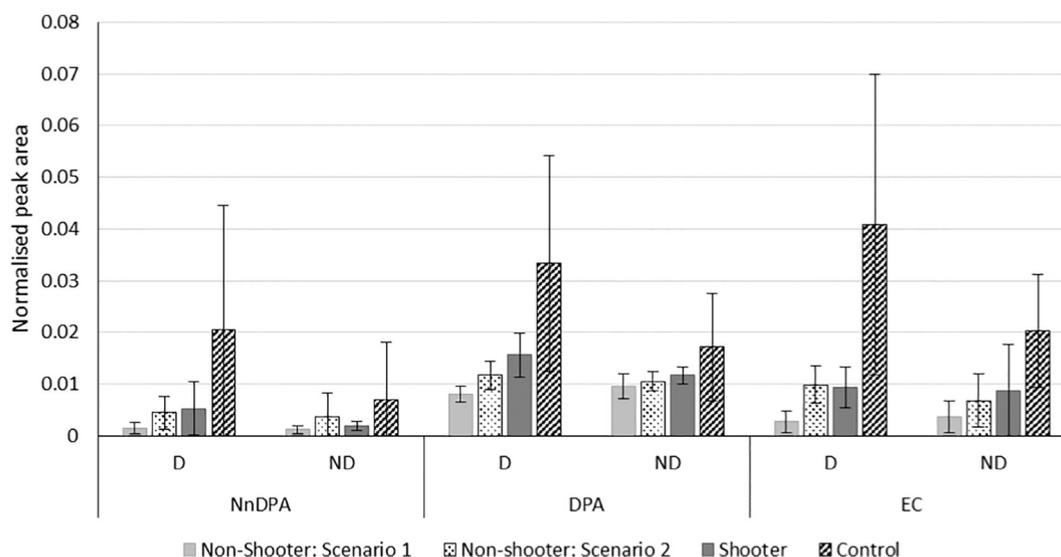


Fig. 4. Comparison between scenario 1 and 2. D = dominant hand, ND = non-dominant hand.

documentation of any contact between officers, firearms and the POI(s). Further, the results suggest that officers' firearm, handcuffs, clothes and other equipment should be cleaned and decontaminated regularly. It would be valuable for contact records to be provided to the forensic scientist in order to assist in the evaluation of the OGSR evidence. This will allow forensic scientists to approach the interpretation of the results with a better understanding of the context in which the specimens were obtained. For instance, a police officer who discharged a firearm shortly before or during an intervention should not enter into contact with POI(s) in order to restrict possible pollution and ultimately reduce the risk of false positive. If such a situation is unavoidable and occurs, sampling the police officer who arrested the POI might be valuable. Such specimens could be used as controls to assess the degree of OGSR pollution of the police officer as soon as possible.

In the global context of GSR, no direct comparison of transfer and persistence data can be performed between IGSR and OGSR because of the different formation process, composition as well as transfer and persistence mechanism. Several studies have examined the secondary transfer of IGSR [12–15]. In the context of an arrest scenario, Charles and Geusens [12] have studied two separate simulations involving a low and high level of contamination of the police officers. For the low contamination simulation, on average, 2 characteristic particles (Pb-Ba-Sb) were detected on the shooter while only 1 was detected on the non-shooter, resulting in 33% of transfer [12]. For the high simulation, however, an average of 66 particles were found on the shooter, while only 3 on the non-shooter (4% of transfer) [12]. Girvan et al. [15] have also studied the secondary transfer of IGSR through the arrest process. They found a secondary transfer of 40% for the characteristic Pb-Ba-Sb particles [15]. Such studies highlight the inherent variability of IGSR deposition and analysis, which was also observed for OGSR in this study.

Despite the differences in the formation, transfer and analysis between IGSR and OGSR. The secondary transfer studies of IGSR [12,15] have shown similar trends when compared to the results observed for OGSR presented in this study, emphasising that a secondary transfer of both IGSR and OGSR is possible in the context of an arrest. The differences are likely to arise from the different retention and transfer properties of IGSR when compared to OGSR. Consequently, such results stress the caution that should be exercised when interpreting GSR evidence, especially when a low number of particles of IGSR or a low amount of OGSR are detected. This study has emphasised the necessity and the importance of assessing the secondary transfer of OGSR. Such results might be used in order to improve the interpretation of such

traces in the context of forensic investigations. An interpretative model can be developed through the use of a probabilistic framework such as Bayesian theorem, which would enable forensic scientists to assess the likelihood ratio (LR) for OGSR outcomes in the light of the propositions of interest as well as the case circumstances. The advantages of the Bayesian approach is that it allows forensic scientists to take into account the possibility of a secondary transfer as well as the persistence when calculating the LR. Such experimental results enable forensic scientists to assess whether the outcomes obtained from the OGSR analysis is more likely to be observed in a scenario involving a firearm discharge or in a scenario where a secondary transfer may have occurred. Such experimental data provide additional and complementary information for OGSR analysis that allow to evaluate the outcomes of OGSR analysis within an activity level context.

4. Conclusion

This study evaluated and explored critical questions regarding the secondary transfer of OGSR between a shooter, who previously discharged a firearm, and a non-shooter, with no immediate prior contact with a firearm. Two scenarios were investigated, one relating to the arrest of the non-shooter by the shooter, the second one approached the handling of a firearm by the non-shooter without discharging it. A secondary transfer occurred during both scenarios, with OGSR detected in each specimen collected from the non-shooter following an arrest process. OGSR was also detected when a non-shooter handled a firearm with similar levels of OGSR detected when compared to a shooter who discharged their firearm followed by conducting an arrest scenario. On average, the amount of OGSR detected did not exceed the amount identified on the shooter who was sampled immediately after discharge.

Interestingly, the amount of OGSR detected on the hands of a shooter did not greatly exceed that detected on the hands (especially the non-dominant hands) of persons who simply handled a gun. It therefore appears logical that the amount of OGSR detected on a shooter is actually the combination of OGSR compounds deposited during firing and the compounds transferred to the hands by handling the gun. Direct transfer of GSR from the gun is usually not controlled in GSR detection experiments and the experimental results presented here indicates that it may be an important factor.

It is essential to assess such scenarios in order to develop a better understanding of OGSR behaviour. In addition, such research provides complementary information to forensic scientists in order to improve the interpretation process. When approaching the assessment of traces

such as OGSR, questions such as secondary transfer and persistence become essential. In practice, standardised protocols restricting or mitigating contact between police officers who discharged a firearm and the POI are advisable as well as recommending that firearms are cleaned on a regular basis to limit the accumulation of OGSR. Additionally, this information and the context of the arrest should be documented and provided to the forensic scientist for the evaluation of OGSR results. Such information allows the results of the analysis to be included into the global context of the case, to be combined with other findings such as IGSR particles. It also allows for consideration of the chronology of the event, the time of sampling and potential sources of pollution such as an arrest process undertaken by contaminated officers. In addition, the results presented in this study can inform an evaluative framework. The secondary transfer can be included in the interpretation process of OGSR in order to provide a more meaningful assessment of such traces. Doing so, this would allow to have a better understanding of such findings by including them in the global context of the case under investigation.

5. Ethics

UTS Human Research Ethics Committee (HREC) approved the presented research (application number 2015000480).

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References

- [1] D. Avery, R.T. Watson, Regulation of lead-based ammunition around the world. Ingestions of lead from spent ammunition: implications for wildlife and humans, (2009), pp. 161–168.
- [2] W.A. MacCrehan, K.D. Smith, W.F. Rowe, Sampling protocols for the detection of smokeless powder residues using capillary electrophoresis, *J. Forensic Sci.* 43 (1998) 119–124.
- [3] D. Laza, B. Nys, J.D. Kinder, A. Kirsch-De Mesmaeker, C. Moucheron, Development of a quantitative LC-MS/MS method for the analysis of common propellant powder stabilizers in gunshot residue, *J. Forensic Sci.* 52 (2007) 842–850.
- [4] S. Benito, Z. Abrego, A. Sánchez, N. Unceta, M.A. Goicolea, R.J. Barrio, Characterization of organic gunshot residues in lead-free ammunition using a new sample collection device for liquid chromatography–quadrupole time-of-flight mass spectrometry, *Forensic Sci. Int.* 246 (2015) 79–85.
- [5] O. Cascio, M. Trettene, F. Bortolotti, G. Milana, F. Tagliaro, Analysis of organic components of smokeless gunpowders: High-performance liquid chromatography vs. micellar electrokinetic capillary chromatography, *Electrophoresis* 25 (2004) 1543–1547.
- [6] I. Jane, P. Brookes, J. Douse, K. O'Callaghan, Detection of Gunshot Residues Via Analysis of their Organic Constituents. Proceedings of the International Symposium on the Analysis and Detection of explosives, (1983), pp. 29–31.
- [7] L.S. Leggett, P.F. Lott, Gunshot residue analysis via organic stabilizers and nitrocellulose, *Microchem. J.* 39 (1989) 76–85.
- [8] J.B.F. Lloyd, High-performance liquid chromatography of organic explosives components with electrochemical detection at a pendant mercury drop electrode, *J. Chromatogr. A* 257 (1983) 227–236.
- [9] A.-L. Gassner, C. Weyermann, LC-MS method development and comparison of sampling materials for the analysis of organic gunshot residues, *Forensic Sci. Int.* 264 (2016) 47–55.
- [10] E. Lindsay, M.J. McVicar, R.V. Gerard, E.D. Randall, J. Pearson, Passive exposure and persistence of gunshot residue (GSR) on bystanders to a shooting: comparison of shooter and bystander exposure to GSR, *J. Can. Soc. Forensic Sci.* 44 (2011) 89–96.
- [11] M. Maitre, K.P. Kirkbride, M. Horder, C. Roux, A. Beavis, Current perspectives in the interpretation of gunshot residues in forensic science: a review, *Forensic Sci. Int.* 270 (2017) 1–11.
- [12] S. Charles, N. Geusens, A study of the potential risk of gunshot residue transfer from special units of the police to arrested suspects, *Forensic Sci. Int.* 216 (2012) 78–81.
- [13] J. French, R. Morgan, J. Davy, The secondary transfer of gunshot residue: an experimental investigation carried out with SEM-EDX analysis, *X-Ray Spectrom.* 43 (2013) 56–61.
- [14] J. French, R. Morgan, An experimental investigation of the indirect transfer and deposition of gunshot residue: further studies carried out with SEM-EDX analysis, *Forensic Sci. Int.* 247 (2015) 14–17.
- [15] J. Girvan, The transfer of gunshot residue surrounding suspect apprehension [Bachelor], Canberra Institute of Technology, 2011.
- [16] J. Arndt, S. Bell, L. Crookshanks, M. Lovejoy, C. Oleska, T. Tulley, D. Wolfe, Preliminary evaluation of the persistence of organic gunshot residue, *Forensic Sci. Int.* 222 (2012) 137–145.
- [17] A.-L. Gassner, M. Manganello, D. Werner, D. Rhumorbarbe, M. Maitre, A. Beavis, C.P. Roux, C. Weyermann, Secondary transfer of organic gunshot residues: empirical data to assist the evaluation of three scenarios, *Sci. Justice* (2018), <https://doi.org/10.1016/j.scijus.2018.08.007>.
- [18] G. Schwendener, S. Moret, K. Cavanagh-Steer, C. Roux, Can contamination occur in body bags? The example of background fibres in body bags used in Australia, *Forensic Sci. Int.* 266 (2016) 517–526.
- [19] R.V. Taudte, C. Roux, D. Bishop, L. Blanes, P. Doble, A. Beavis, Development of a UHPLC method for the detection of organic gunshot residues using artificial neural networks, *Anal. Methods* 7 (2015) 7447–7454.
- [20] A.-L. Gassner, C. Ribeiro, J. Kobylinska, A. Zeichner, C. Weyermann, Organic gunshot residues: observations about sampling and transfer mechanisms, *Forensic Sci. Int.* 266 (2016) 369–378.
- [21] M. Maitre, M. Horder, K.P. Kirkbride, A.-L. Gassner, C. Weyermann, C. Roux, A. Beavis, A forensic investigation on the persistence of organic gunshot residues, *Forensic Sci. Int.* 292 (2018) 1–10.
- [22] C. Hofstetter, M. Maitre, A. Beavis, C.P. Roux, C. Weyermann, A.-L. Gassner, A study of transfer and prevalence of organic gunshot residues, *Forensic Sci. Int.* 277 (2017) 241–251.
- [23] M. Maitre, K. Kirkbride, M. Horder, C. Roux, A. Beavis, Thinking beyond the lab: organic gunshot residues in an investigative perspective, *Aust. J. Forensic Sci.* (2018) 1–7.
- [24] R.V. Taudte, C. Roux, A. Beavis, Stability of smokeless powder compounds on collection devices, *Forensic Sci. Int.* 270 (2016) 55–60.
- [25] L. Ali, K. Brown, H. Castellano, S.J. Wetzel, A study of the presence of gunshot residue in pittsburgh police stations using SEM/EDS and LC-MS/MS, *J. Forensic Sci.* 61 (2016) 928–938.
- [26] ICH, Harmonised Tripartite Guideline. Validation of Analytical Procedures: Text and Methodology Q2 (R1), International Conference on Harmonization, Geneva, Switzerland, 2005, pp. 11–12.
- [27] R.V. Taudte, C. Roux, L. Blanes, M. Horder, K.P. Kirkbride, A. Beavis, The development and comparison of collection techniques for inorganic and organic gunshot residues, *Anal. Bioanal. Chem.* 408 (2016) 2567–2576.
- [28] R.G. Brereton, Chemometrics for Pattern Recognition, John Wiley & Sons, 2009.
- [29] K. Varmuza, P. Filzmoser, Introduction to Multivariate Statistical Analysis in Chemometrics, CRC Press, 2016.
- [30] F.S. Romolo, P. Margot, Identification of gunshot residue: a critical review, *Forensic Sci. Int.* 119 (2001) 195–211.