

1 **Surveys of organic gunshot residue prevalence: comparison**
2 **between civilian and police populations**

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12 **Abstract**
13

14 This study investigated the prevalence of eight OGSR compounds in a civilian and a police
15 population. Specimens were collected from the hands and sleeves of 122 civilians and 115 individuals
16 working in police services using carbon stubs. Data was acquired using liquid chromatography
17 coupled to tandem mass spectrometry.

18 Results indicated a non-negligible prevalence in the civilian sample, with 18% of the 122 civilians
19 sampled having one or more OGSR related compounds on their hands and 11.5% on their wrists or
20 sleeves. For the police population, the prevalence was logically higher than for civilians, with 36.5%
21 of the hand specimens and 33% of the wrist specimens positive for one or more compounds. A higher
22 prevalence was expected for the second population due to the possession of service weapons regularly
23 used during shooting exercises. These results demonstrate that the presence of one OGSR compound
24 is not a rare occurrence, even in a civilian population. Considering the results of this research together
25 with information on alternative sources of the targeted OGSR compounds can serve as a basis for
26 OGSR interpretation in casework.

27
28 Keywords: Forensic science, firearms, firearm discharge residue, contamination
29

30 **1. INTRODUCTION**

31 According to the *Small Arms Survey*, the estimated number of firearms all over the world, licit and
32 illicit, might exceed one billion [1]. They are divided between firearms held by civilians (84.6%), state
33 militaries (13.1%) and law enforcement agencies (2.2%). These proportions are highly country-
34 dependent due to various policies on firearms, crime rates and ongoing armed conflicts. Incidents
35 involving firearms are usually investigated by forensic science services. Gunshot residues (GSR)
36 produced during a firearm discharge can be detected and analysed in casework to evaluate if a person
37 has been involved in a shooting incident [2]. During a discharge, GSR are ejected from all firearm
38 openings and deposited onto the shooter and various surfaces in close proximity. They originate from
39 the primer and propellant, as well as other firearm and ammunition components such as metals (from
40 the bullet, cartridge case or gun barrel) and lubricant [3, 4]. GSR are classified into primer residues,
41 also called inorganic GSR (IGSR), and propellant residues that will be referred to as organic GSR
42 (OGSR) [2]. It is important to note that even though IGSR are called primer residues, their chemical
43 composition is not limited to that of the primer, but contributions from other metallic surfaces
44 involved in the discharge or to bluing products used to protect firearms from corrosion [5] have been
45 observed. The analysis of IGSR is routinely performed by scanning electron microscopy coupled to
46 energy dispersive X-ray spectroscopy (SEM-EDX) [2], whereas OGSR usually remain unexploited in
47 routine analyses.

48 GSR can be transferred primarily to the shooter, but also to their clothing, a bystander, the victim or
49 any object or surface in close proximity. The amount of OGSR transferred will depend on the
50 ammunition (composition, combustion efficiency) and firearm (model, memory effect from previous
51 ammunition, lubricant) used. The number of cartridges shot might also have an influence. Moreover,
52 GSR might be further transferred to other surfaces and individuals through processes called secondary
53 transfer and tertiary transfer. Thus, it might be possible to detect GSR on an individual not involved in
54 any firearm discharge. As indicated in the literature, such events are not negligible and should be
55 considered when interpreting GSR traces [6-10]. Therefore, it is essential to estimate the chances of an
56 individual testing positive for GSR without being involved in a firearm incident.

57 To date, the prevalence of IGSR has been investigated in various populations. Lucas *et al.* studied the
58 occurrence of IGSR in a random population of 289 Australians [11]. Their results showed an overall
59 prevalence of 0.3% for characteristic PbBaSb particles, 8% for PbSb and about 7% for single Pb, Ba
60 or Sb particles. In Poland, Brozek-Mucha studied a population of 100 non-shooters and 50 shooters
61 [9]. Only one PbBaSb particle was detected among individuals who had no contact with firearms,
62 whereas the numerous particles found among shooters showed a strong correlation with the time
63 elapsed since the last shooting session. The prevalence of IGSR was also investigated among police
64 officers. Gialamas *et al.* found 3 specimens with one PbBaSb particle in a population of 43 non-
65 shooting police officers in California [12]. In Canada, Gerard *et al.* detected at least one PbBaSb
66 particle on the hands of 60% of patrol and plainclothes officers (66 individuals) and on 24% of their

67 equipment [13]. In the same study, no IGSR particles were found on the 28 civilians working in a
68 police environment and only 2 of the 18 vehicles sampled had one characteristic GSR particle. In
69 Australia, Cook sampled 33 police officers immediately after the start-of-shift handling of their
70 firearm and 28 had PbBaSb particles on their hands, with an average of 64 such particles [14]. Another
71 study by Lindsay *et al.* targeted 13 employees of firearm manufactures [15]. PbBaSb particles were
72 found on nine of the employees, and no more than two characteristic particles were found on the hands
73 of the five individuals who had no direct contact with firearms or items that had been in the firing
74 range. For the other four employees, the number of particles ranged from nine to 424. In the United
75 States, Berk *et al.* investigated police vehicles and detention facilities in Chicago [16]. A total of 201
76 specimens were analysed and a total of 56 PbBaSb particles were found in 23 specimens. Only two
77 vehicles had one particle. The other particles were recovered from detention facilities with a maximum
78 of seven particles collected from a table surface and restraining bars. Finally, in Ireland, Hannigan *et*
79 *al.* collected 100 upper body garments submitted to a forensic laboratory that were not related to a
80 firearm offence [17]. 98% of the specimens collected from the cuffs were negative: up to two PbBaSb
81 particles were detected on two garments. The aforementioned studies showed that prevalence can
82 considerably differ depending on the targeted population, people and objects. The items/people
83 directly in contact with firearms generally presented the largest prevalence.

84 In the case of IGSR, the elemental composition, the morphology and the composition of the entire
85 particle population of the specimen are considered by the analyst [18]. However, due to the common
86 use of bulk analytical techniques such as micellar electrokinetic capillary electrophoresis (MEKC)
87 [19-22], gas chromatography (GC) [23-25] or liquid chromatography coupled to mass spectrometry
88 (LC-MS) [26-30], the sole overall specimen composition can be obtained for OGSR. A positive result
89 is based on the detection of an OGSR compound, regardless of its source. Hence, the knowledge of
90 potential alternative sources of OGSR compounds is a prerequisite for the interpretation of that trace.
91 Currently, there is only limited or obsolete information regarding alternative sources, leading to a lack
92 of precise definition of which OGSR compounds are the most characteristic [31]. However, if a
93 sufficient amount of data is collected, prevalence studies might also help in identifying alternative
94 sources. Few prevalence researches have been published concerning OGSR. Various explosive
95 background studies including the detection of nitroglycerine (NG) and dinitrotoluenes (DNT), which
96 are also major components of smokeless powders, indicate a low prevalence of these compounds in
97 public places [32-34]. Regarding OGSR prevalence, the hands of 100 volunteers were sampled in the
98 United States and no specimen tested positive using MEKC [19]. Another study based on a sample of
99 73 people including law enforcement personnel of Morgantown, West Virginia (USA), showed less
100 than 5% of positive results for the specimens analysed by ion-mobility spectrometry (IMS) [35]. The
101 authors used neural networks and the results were based on pattern matching. Thus, the proportion of
102 positive results depended on the threshold defined by the user. In that study, when a likelihood ratio of

103 10:1 was used, the frequency of positives dropped under 2%. In Switzerland, a preliminary study
104 concerning 27 people detected no positive specimen using LC-MS and also concluded that OGSR
105 occurrence was rare [36]. Furthermore, as suspects might be contaminated during an arrest,
106 transportation in a police car or detention, it is important to estimate the level of contamination of
107 police officers and premises. Hofstetter *et al.* collected specimens from 25 individuals working in a
108 forensic laboratory [36], only two positive results were observed. Ali *et al.* [37] collected seventy
109 specimens from four Pittsburgh police stations and vehicles and analysed both IGSR and OGSR. Only
110 one characteristic IGSR particle was detected, whereas ethylcentralite (EC) was the only organic
111 compound quantified in four specimens. Altogether, even though the aforementioned studies indicate a
112 rather low prevalence, the amount of prevalence data for OGSR molecules required to interpret that
113 trace evidence remains very limited. More studies using sensitive instrumental techniques and a higher
114 number of specimens need to be carried out.

115 In order to gain more knowledge about the prevalence of OGSR in Switzerland, the present work
116 investigated the detection of eight OGSR compounds in two populations to provide useful information
117 for the assessment of OGSR evidence. Specimens were collected from 122 civilians and 115
118 individuals working in a police environment. Collection was performed using carbon stubs and data
119 was acquired using LC-MS/MS with electrospray ionisation in positive mode. The results obtained
120 from both populations were compared and discussed in a forensic perspective.

121

122 **2. MATERIAL AND METHODS**

123 **2.1 Target populations**

124 Specimens were collected from two populations. The first one is a so called *civilian population*
125 composed of 122 volunteers with no or limited contact with firearms. All of them completed a
126 questionnaire evaluating their firearm exposure. Data such as occupation, gender, hunting/military
127 activity and firearm ownership (model) were collected. If volunteers indicated ownership or use of a
128 firearm, data regarding the ammunition type, the frequency of use, the date of the last shooting session
129 and the number of cartridges discharged was also collected. Finally, they were also asked about the
130 last time they washed their hands and whether they thought there was a reason they would test positive
131 for OGSR.

132 The second survey targeted a *police population*. This included police officers, police recruits as well as
133 scientific collaborators working in three regional Swiss police services. 115 individuals were sampled
134 and filled in a questionnaire similar to the one submitted to the civilian population, with an additional
135 question included regarding the use of a firearm outside the purpose of their profession.

136

137 **2.2 Specimen collection and preparation**

138 Specimens were collected using carbon stubs from Plano (Wetzlar, Germany), consisting of an
139 adhesive carbon tab 12 mm in diameter mounted on a 12.5 mm aluminium stub. This assembly was
140 inserted in a plastic vial with a screwed cap. Two stubs were used for each participant, one for their
141 hands and the other for their wrists or the cuffs of their sleeves (depending whether they wore a short
142 or long-sleeved shirt). Following recommendations from Zeichner *et al.* [38], the stubs were dabbed
143 about 100 times on the skin and 200 times on the sleeves. For the hands, sample collection was first
144 performed on the thumb-index region and then on the back and palm. To minimize contaminations,
145 the person in charge of specimen collection wore gloves and changed them after each participant's
146 sampling. It must be noted that 50 specimens (civilian population) were auto-collected, that is to say
147 that the participants were given a sampling kit to stub themselves. Prior to that, a presentation
148 explaining the goal of the study and the sampling procedure was given.

149
150 To extract the compounds present in the specimens, the carbon adhesive was removed from the stub
151 with clean tweezers and transferred to a 20 mL scintillation vial containing 1 mL MeOH. After 15
152 minutes ultrasonication at room temperature, the resulting solution was filtered through a 0.2 µm
153 Chromafil PTFE syringe filter (Macherey-Nagel, Düren, Germany) to remove carbon particles. To
154 detect potential laboratory contamination during specimen preparation, methanol blanks were prepared
155 before and after each extraction session. Likewise, a blank carbon tab was extracted to check for
156 potential contamination of the stub batch. For all these control samples, no OGSR were detected.

158 **2.3 Chemicals**

159 Acetonitrile, methanol, formic acid (FA) and water were of ULC-MS grade (Sigma-Aldrich, Buchs,
160 Switzerland). The study targeted eight OGSR compounds: diphenylamine (DPA) from Fluka (Buchs,
161 Switzerland); ethylcentralite (EC), *N*-nitrosodiphenylamine (*N*-nDPA), 4-nitrodiphenylamine (4-
162 nDPA), akardite II (AK II) and *N,N*-diphenylformamide (*N,N*-DPF) from Sigma-Aldrich (Buchs,
163 Switzerland); 2-nitrodiphenylamine (2-nDPA) from Alfa Aesar (Karlsruhe, Germany);
164 methylcentralite (MC) from MP Biomedicals (Illkirch, France). Standard solutions at 1 mg/mL were
165 prepared in MeOH and stored at 4°C.

167 **2.4 Instrumentation**

168 The specimens were analysed using an Agilent Infinity 1290 ultra-high performance liquid
169 chromatography (UHPLC) from Agilent Technologies. The instrument was equipped with a binary
170 pump enabling a maximum delivery flow rate of 5 mL/min, an autosampler, and a thermostatically
171 controlled column compartment. Separation was performed using a C18 Kinetex core-shell column
172 from Phenomenex (2.6 µm, 2.1 mm × 100 mm). A SecurityGuard ULTRA cartridge with C18
173 selectivity was used to protect the analytical column. The UHPLC system was coupled to a triple
174 quadrupole mass spectrometer (5500 QTrap) from AB Sciex. Electrospray ionization was operated in

175 positive mode. $[M+H]^+$ was defined as the precursor ion for each of the target compounds, and
 176 quantification was obtained from the SRM measurements. The source parameters were as follows: the
 177 desolvation temperature was set to 500°C, the nebulizer gas to 60 psig, the turbo gas to 50 psig and the
 178 curtain gas to 25 psig. The IonSpray voltage was adjusted to 5500 V. Data acquisition, treatment and
 179 instrument control were monitored using Analyst software. Details of the LC method, MS/MS
 180 parameters and limits of detection (LOD) can be found in Tables 1 and 2.

181

182 **Table 1:** LC parameters [30]

Column type	C18 (2.6 μ m, 2.1 mm \times 100 mm),		
Column temperature	40 °C		
Flow rate	0.25 mL/min		
Injection volume	5 μ L		
Gradient table	t / min	% A	% B
		H₂O+0.1%FA	ACN+0.1%FA
	0	65	35
	0.5	65	35
	6	20	80
	7	0	100

183

184 **Table 2:** MS parameters [30]

	SRM transitions	LOD [ng/mL]	Declustering potential [V]	Collision energy [V]
Akardite II (AK II)	227.1 \rightarrow 170.1 227.1 \rightarrow 91.9	0.01	120	27 36
Methylcentralite (MC)	241.2 \rightarrow 134.1 241.2 \rightarrow 105.9	0.01	125	24 36
<i>N,N</i>-diphenylformamide (<i>N,N</i>-DPF)	198.1 \rightarrow 92 198.1 \rightarrow 65	0.02	130	30 54
Ethylcentralite (EC)	269.2 \rightarrow 147.9 269.2 \rightarrow 120	0.01	120	20 33
2-nitrodiphenylamine (2-nDPA)	215.1 \rightarrow 197 215.1 \rightarrow 180.1	0.02	80	14 23
4-nitrodiphenylamine (4-nDPA)	215.1 \rightarrow 197.8 215.1 \rightarrow 167.1	0.02	60	18 47
Diphenylamine (DPA)	170.1 \rightarrow 93 170.1 \rightarrow 66	0.5	200	32 58
<i>N</i>-nitrosodiphenylamine (<i>N</i>-nitrosoDPA)	199.1 \rightarrow 169 199.1 \rightarrow 66	0.02	60	15 30

185

186 Semi-quantitative data were obtained from a calibration curve (11 levels, 2 replicates) measured for
 187 each sequence of experiments with levels ranging between the LOD and 10 ng/ml, except for DPA
 188 with a 100 ng/mL highest level.

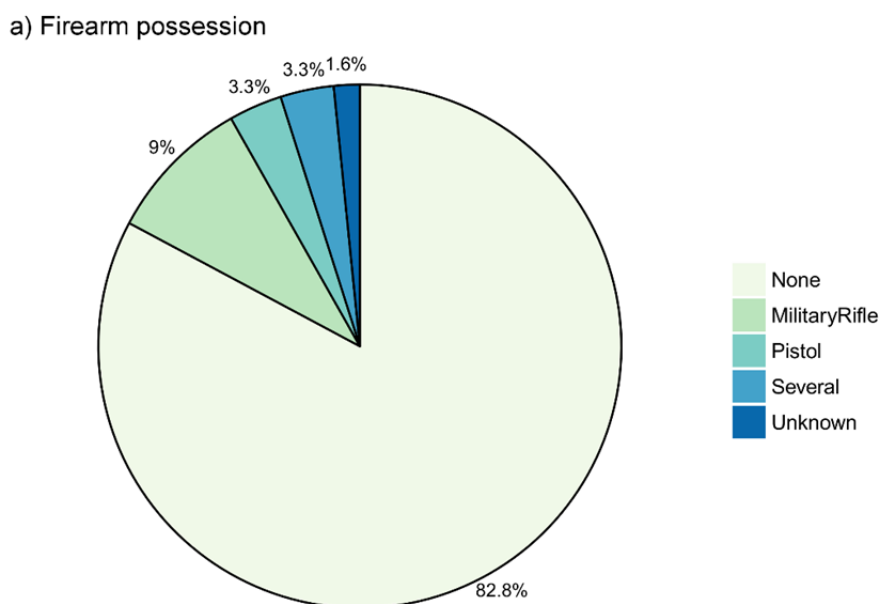
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190 **3. RESULTS AND DISCUSSION**

191 **3.1 Civilian population**

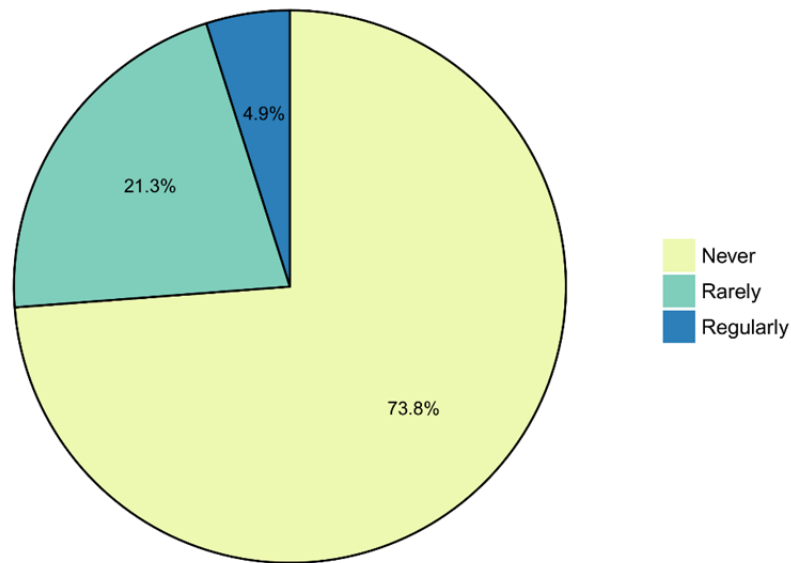
192 The civilian population consisted of 122 participants. The questionnaires were used to extract
193 information relative to the occupations of the participants and to evaluate their potential exposure to
194 GSR in order to help interpret positive results. Among the 122 individuals, the gender was equally
195 distributed with 61 men and 61 women. 68 students were sampled, representing 55.7% of the whole
196 dataset. The majority of the participants (101 people) did not possess a firearm (Fig. 1a), while 11 had
197 a military rifle, 4 owned a pistol (9 mm calibre), 4 owned more than one firearm. Two people did not
198 answer that question. The frequency of firearm use (Fig. 1b) was divided into three categories:
199 “never”, “rarely” (people who shoot at least once a year) and “regularly” (those who shoot at least
200 once a month). It was observed that less than 5% of the individuals regularly used firearms. Only one
201 individual reported discharging a firearm in the two weeks before specimen collection. It must be
202 emphasized that this participant was sampled only ten minutes after the last shooting practice, did not
203 wash his hands after shooting, and as expected, tested positive for OGSR. Of the 82.8% of the
204 individuals who did not own a firearm, 73.8% of them reported having never shot. Finally, the
205 participants were asked if they thought they might be contaminated. Seven individuals answered
206 positively. Their explanations for potential contamination were the following: cleaning, manipulating
207 or discharging a firearm (four participants), visiting premises contaminated with GSR (two
208 participants) and installing a stand in the boot of a police vehicle (one participant).

209



210

b) Firearm use frequency



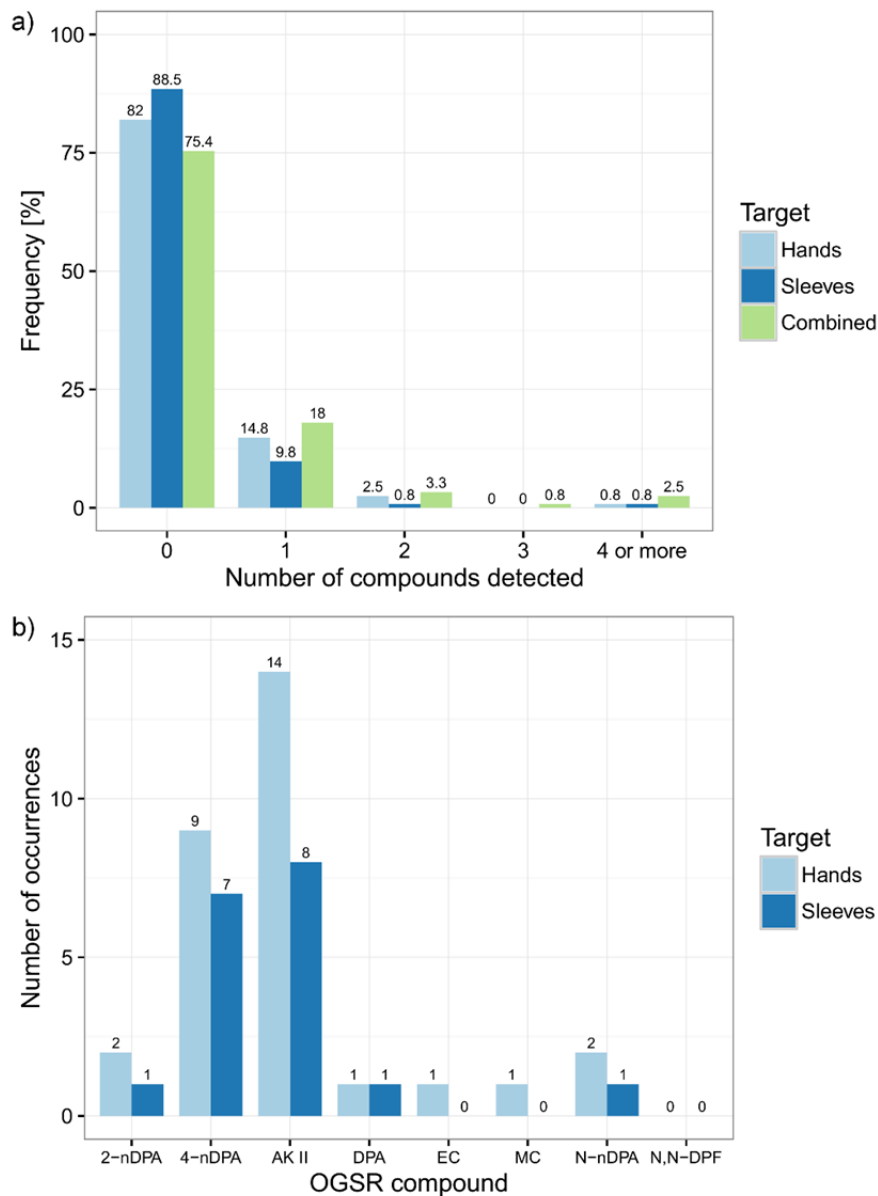
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212 **Figure 1:** a) Firearm possession and b) Frequency of firearm use in the population of 122 civilians

213

214 Among the 122 individuals sampled in this study, only a minority tested positive for OGSR (Fig. 2a).
215 For the hand specimens, 100 individuals (82%) were free from OGSR, 18 individuals (14.8%) had one
216 OGSR compound, three individuals (2.5%) had two compounds and only one individual had six
217 compounds on their hands. Comparable results were found for the sleeve specimens with slightly
218 fewer participants testing positive. In fact, 108 individuals (88.5%) showed no OGSR, while 12 people
219 (9.8%) had one compound, and two people had two and four compounds respectively. It is interesting
220 to note that the hand and sleeve specimens with the highest number of compounds were not collected
221 from the same person. When considering the hands and sleeves together, a higher percentage of people
222 were contaminated with at least one compound (Fig. 2a, green bars). Thus, it might be interesting to
223 collect specimens from other surfaces than hands as hands are generally washed much more frequently
224 than the face and hair, for example [36, 39]).

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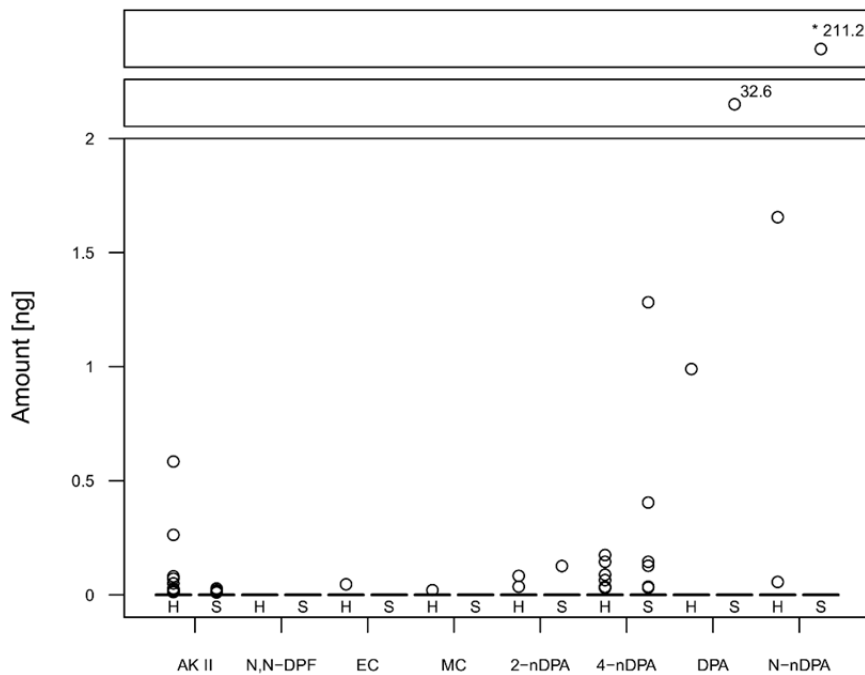
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229 **Figure 2:** Prevalence in the civilian population: a) Prevalence versus number of compounds detected. The “combined” bar
 230 considers the total number of compounds detected on both hands and sleeves irrespective of potential compound duplication.
 231 For example, if DPA was detected on the hands and the same compound was detected on the sleeves, two compounds were
 232 counted. b) Number of positive specimens for each analyte.

233

234 Regarding the number of positive results per compound (Fig. 2b), it can be seen that AK II (47% and
 235 44% of positive results for hands and sleeves respectively) and 4-nDPA (30% and 39% of positive
 236 results) were the most frequently detected compounds. Other DPA derivatives and centralites were
 237 rarely detected and *N,N*-DPF was never detected. The amounts detected (Fig. 3) were in the low ng
 238 range and very often close to the LOD. Such amounts are in the same range as those observed in
 239 persistence and secondary transfer studies [10, 40], showing that it might be very difficult to
 240 distinguish between firearm discharge and incidental contamination.



241
 242 **Figure 3:** Prevalence in the civilian population: Amount of analyte detected. The asterisk indicates an extrapolated value
 243 (outside of calibration range)
 244

245 The data from the questionnaire were investigated to look for a potential explanation for the presence
 246 of OGSR compounds. Two individuals had four or more compounds detected on their hands and
 247 sleeves respectively. The person who had six compounds on his hands (AK II, EC, DPA, N-nDPA, 4-
 248 nDPA and 2-nDPA) was the participant (student) who discharged a firearm less than ten minutes
 249 before specimen collection without washing his hands and thus expected testing positive for OGSR.
 250 Interestingly, only AK II was detected on his sleeves. The person who had four compounds on his
 251 sleeves (DPA, N-nDPA, 4-nDPA and 2-nDPA) and no compounds detected on his hands was a student
 252 who never discharged a firearm. In that case, the presence of these compounds could not be explained
 253 by the questionnaire and might be due to contamination through secondary transfer. Three compounds
 254 were only detected when the results of the hands and sleeves were added (Fig. 2a). However, three
 255 individuals tested positive for two compounds, one only on the hands (4-nDPA and N-nDPA), one
 256 with two compounds on the hands and one on the sleeves (AK II, 4-nDPA and 4-nDPA respectively)
 257 and one with the same two compounds on the hands and sleeves (AK II and 4-nDPA). These three
 258 individuals had not manipulated or discharged a firearm recently. One of them (AK II and 4-nDPA on
 259 both hands and sleeves) thought a positive result was possible because he had installed a stand in the
 260 boot of a police vehicle. That person also owned a military rifle that he had not used for more than ten
 261 months. In that case, an explanation might be the contact with a contaminated vehicle or a secondary
 262 transfer from a surface contaminated by the weapon. The other two had no explanation for that result
 263 in their questionnaire. However, all three shared the same occupation, vehicle locksmith, and worked
 264 in the same garage. Thus, working in contact with vehicles might be an alternative source for OGSR

265 compounds. Another explanation might be a tertiary transfer from a contaminated colleague as the
266 individuals worked in the same garage. Among the individuals for which only one compound was
267 detected (18 on the hands and 12 on the sleeves), three had the same compound on their hands and
268 sleeves (twice 4-nDPA and once AK II). None of them had contact with firearms. One was a
269 consultant, the other a mechanic and the last one a student. Except for five one-compound positive
270 specimens that might be explained by the possession of a firearm, the origin of the positive results is
271 unknown. It is also worth mentioning that three individuals had occupations related to cars, adding to
272 the three two-compounds positive results related to those professions. Regarding the compounds that
273 were detected alone, AK II was the most frequent (11 and 7 positives for hands and sleeves
274 respectively) followed by 4-nDPA (5 positives for both hands and sleeves).

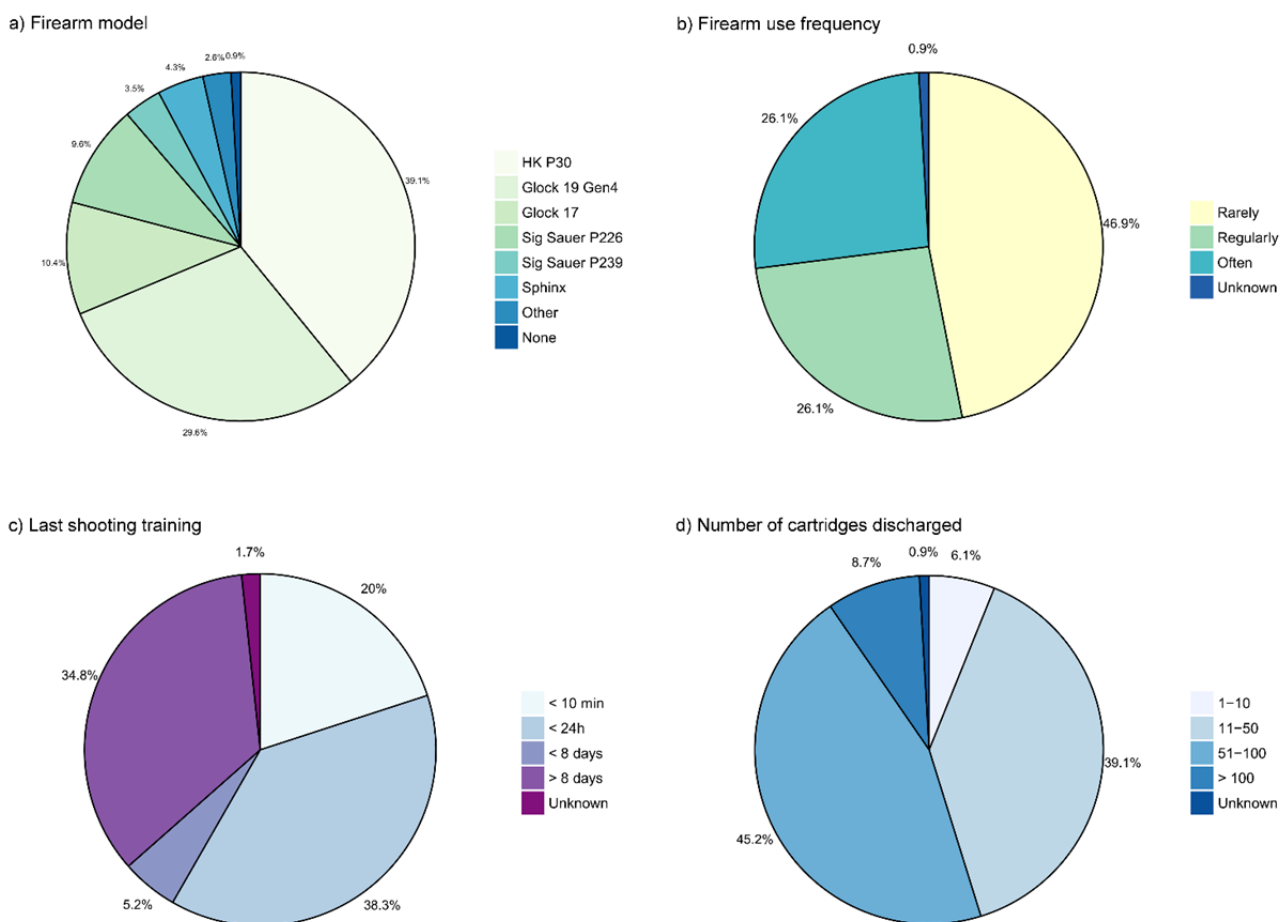
275 In summary, the present results show that finding one compound on the hands of a civilian is not a rare
276 occurrence, as the hands and sleeves of 18 and 12 individuals respectively tested positive for one
277 compound (for a total of 24.6% of the studied civilian population, hands and sleeves combined).
278 However, the presence of more than one OGSR compound remains unlikely (less than 7% of the
279 studied population with two explanations related to firearms). Results also indicate that some activities
280 in relation to vehicles might represent an alternative source for some OGSR compounds. However,
281 this should be confirmed by further research.

282

283 **3.2 Police population**

284 For the prevalence study of the police population, 115 individuals from three Swiss police services
285 were sampled. The population was composed of 97 men (84.3%) and 18 women (15.7%). All had a
286 service weapon except for one scientific collaborator. Regarding the firearm brand and model, all
287 individuals possessed a 9 mm Luger calibre service pistol from different brands (Fig. 4a). 30
288 individuals practiced shooting at least once a week (often: 26.1%), 30 at least once a month (regularly:
289 26.1%) and for 54 individuals at least once a year (rarely: 46.9%) (Fig. 4b). The date of the last
290 shooting session might also provide information as to the probability of a positive OGSR result and
291 was reported in Figure 4c. Indeed, 23 individuals (20%) had just finished a shooting session at the
292 time of specimen collection. The amounts on their hands and sleeves were therefore expected to be
293 much higher than for individuals who last practiced more than one week prior. 44 individuals had
294 practiced shooting in the last 24 hours, six in the last eight days and for 40, the last shooting training
295 was more than eight days ago. As to the number of cartridges fired during training, 52 individuals
296 (45.2%) used between 51 and 100 cartridges and 45 (39.1%) between 11 and 50 cartridges (Fig. 4d).
297 Consequently, the level of contamination from a police officer after training might be very high.
298 Lastly, the individuals were asked about shooting activities outside work. Most of the sampled police
299 officers (94 people: 81.7%) did not practice shooting as a hobby.

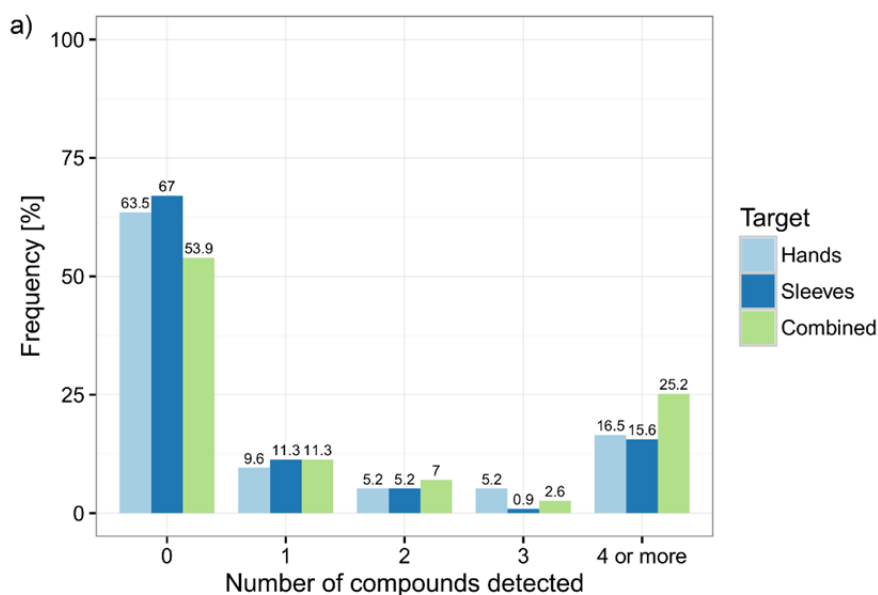
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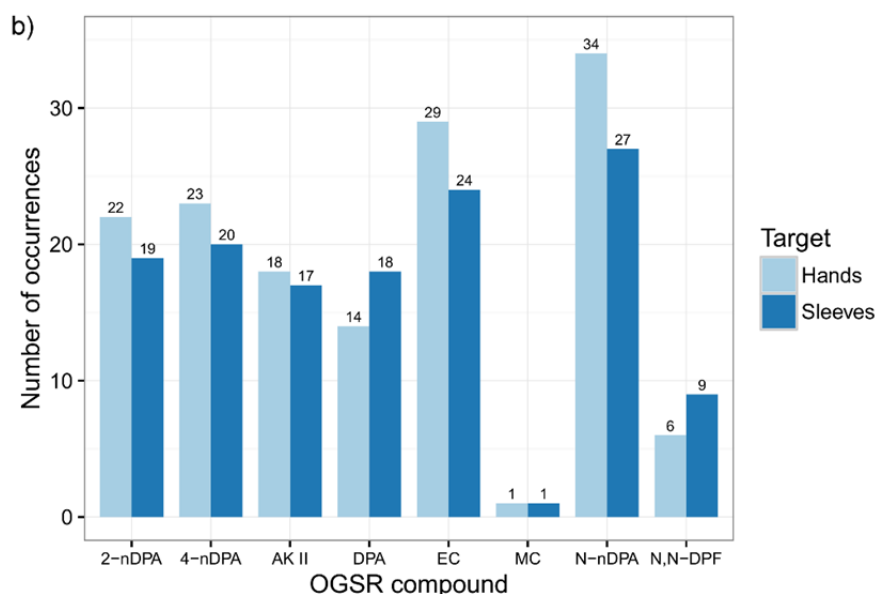
301 **Figure 4:** Distribution of the police population in terms of a) Firearm model, b) Frequency of firearm use, c) Last shooting
 302 training and d) Number of cartridges discharged (n = 115)

303
 304 In the police population, the hands of 73 (63.5%) and the sleeves of 77 (67%) individuals tested
 305 negative for OGSR (Fig.5a). One compound was detected in about 10% of the individuals for both
 306 hands and sleeves. Two and three compounds were less frequently detected (about 5%). Four or more
 307 compounds were found on the hands/sleeves of 19 and 18 individuals respectively (16.5% and 15.6%
 308 respectively). When adding the numbers of compounds on hands and sleeves, only 62 individuals
 309 (53.9%) were free from OGSR. Specimens from 25.2% (29 people) gave a total of four or more
 310 compounds. Interestingly, the police prevalence of one compound was slightly lower than that
 311 observed in the civilian population (9.6 vs 14.8% on hands).

312 Regarding the occurrence of each compound (Fig. 5b), *N*-nDPA was the most frequently encountered
 313 compound, with 34 occurrences on hands and 27 on sleeves. EC was the second most common
 314 compound, followed by DPA derivatives, AK II and DPA. Finally, *N,N*-DPF was more rarely
 315 detected, but this might also be due to its very low proportion in smokeless powders. MC was only
 316 detected once, indicating that it may not be present or only at a very low percentage in the ammunition
 317 used by the police forces.



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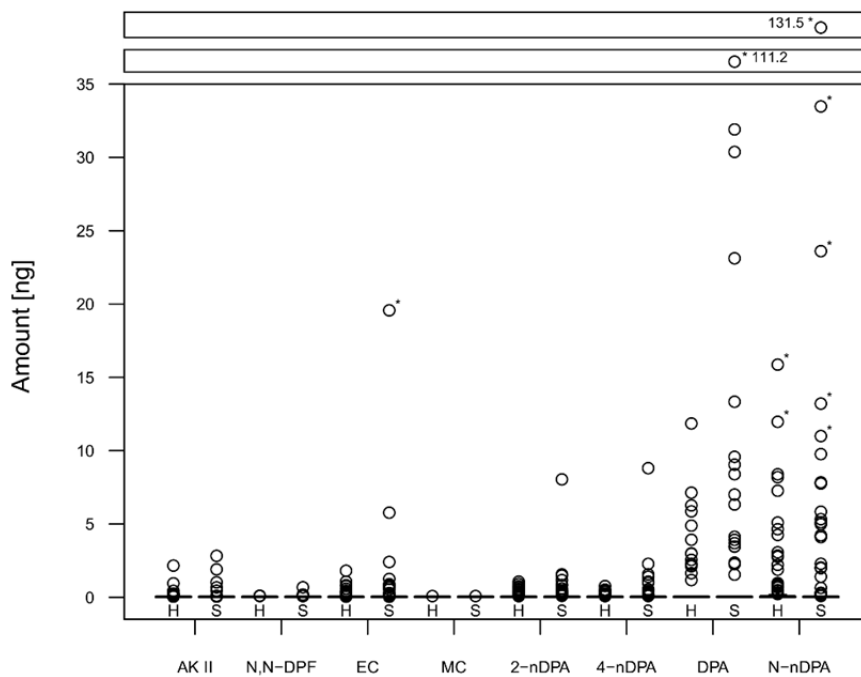


320

321 **Figure 5:** Prevalence in the police population. a) Prevalence versus number of compounds detected. The “combined” bar
322 considers the total number of compounds detected on both hands and sleeves irrespective of potential compound duplication.
323 For example, if DPA was detected on the hands and the same compound was detected on the sleeves, two compounds were
324 counted. b) Number of positive specimens per analyte

325

326 There was a substantial variation in the amounts of OGSR compounds recovered, with values ranging
327 from zero to values above the highest calibration standard (Fig. 6). The highest amounts were
328 observed for DPA and N-nDPA. The other compounds were detected at tenfold lower concentrations.
329 This might be explained by the ammunition used by police services, as the composition of smokeless
330 powders has been shown to vary between brands [19]. Thus, DPA might be a major stabilizer in the
331 smokeless powders used by the collaborators of the three police services sampled in this study.



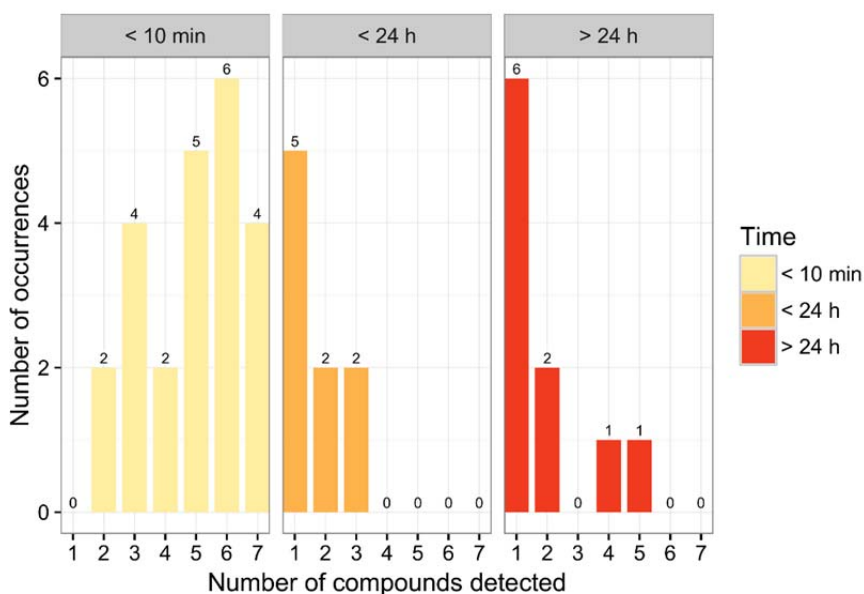
332
 333 **Figure 6:** Prevalence in the police population: Amount of analyte detected. The asterisks indicate extrapolated values
 334 (outside of calibration range)
 335

336 As expected, the highest number of compounds was detected on individuals who had just finished
 337 their shooting training (< 10 min). All the specimens from their hands tested positive for OGSR and a
 338 total number of seven compounds were detected for four of them (Fig. 7a). Such a high number was
 339 never detected for individuals for whom the time since last shooting was over ten minutes. It is
 340 interesting to emphasize that in the less than 24h category, two and three compounds were detected on
 341 the hands of individuals who had discharged a firearm less than three hours before. However, five and
 342 four compounds respectively were found on the hands of two people more than 24h after shooting. A
 343 similar trend was observed for the wrists/sleeves specimens as the highest numbers of compounds (up
 344 to eight compounds) were generally detected from the sleeves of individuals who had just finished a
 345 shooting session (Fig. 7b). Four to six compounds were detected on the sleeves of four people who
 346 fired less than 24h before. Interestingly, the two cases with six compounds and the one with four
 347 compounds were from individuals who practiced shooting less than three hours ago. After more than
 348 24 hours, a maximum of one compound was detected on the sleeves.

349 As illustrated in Fig.7a-b, the distribution of the number of compounds varied with the elapsed time.
 350 Logically, the highest numbers of compounds were detected just after firing and due to losses related
 351 to various activities, lower numbers were detected afterwards. Thus, the detection of a high number of
 352 OGSR compounds can be related to a recent discharge. Persistence studies drew attention to the rapid
 353 decrease in OGSR quantities [39, 40]. Therefore, a decrease in the number of detected compounds
 354 may be due to the early disappearance of less concentrated compounds.

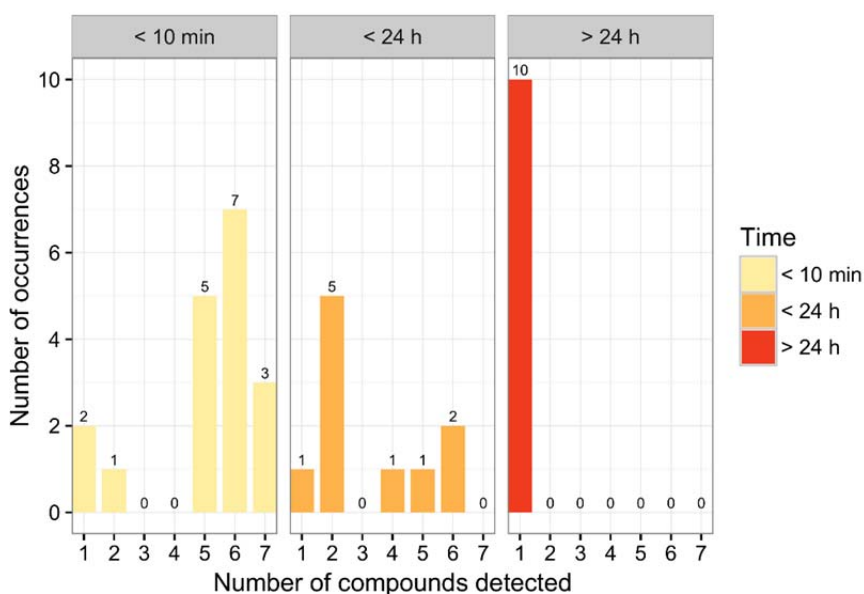
355

356 a)



357

358 b)



359

360 **Figure 7:** Number of OGSR compounds detected on a) police officers' hands, b) police officers' sleeves vs time elapsed
361 since last shooting

362

363 The detection of four or more compounds was generally related to individuals who had discharged a
364 firearm in the last few hours before specimen collection. However, in two cases, four and five
365 compounds were detected on the hand specimens of people who had not discharged a firearm recently
366 (> 24h). On referring to the questionnaire data, it was revealed that one of these two police officers
367 had shaken hands with someone who had just left shooting training (five compounds) and the other
368 had handled his service weapon not long before OGSR collection (four compounds). Weapon
369 manipulation also explained the two instances with two compounds on the hands after > 24h. These
370 results imply that it is rare to detect more than one OGSR compound beyond 24 hours without a

371 secondary transfer explanation. Two recent studies highlighted secondary transfer during a handshake
372 or firearm displacement [10, 41]. For police officers, manipulation of a firearm involves generally
373 more than ten seconds of contact, as investigated in the first aforementioned study [10]. Depending on
374 the background contamination and the intensity of the manipulation, such as opening the breech or
375 removing the charger, a high degree of secondary transfer might occur as confirmed in the present
376 study and in the second aforementioned study [41]. For the positive specimens containing one
377 compound, the results often remained unexplained, whether for hand or wrists specimens. It is also
378 worth emphasizing that wrist/sleeve specimens collected from five individuals who had just finished
379 training were negative. Thus, if the detection of OGSR compounds generally implies a firearm
380 discharge, a negative result shortly after discharge is also possible. While the aforementioned
381 observations demonstrate the contamination risks in a police environment, more research will be
382 necessary to understand the different factors at play in the transfer and persistence of OGSR. Contact
383 with contaminated colleagues, manipulation of weapons, contaminated equipment (e.g. clothing,
384 handcuffs) and premises (e.g. tables, computer, cars) are potential sources of OGSR positive
385 specimens. Moreover, while detecting four or more compounds seems highly correlated with a recent
386 firearm discharge, the presence of one compound appears to be a usual occurrence in such
387 environment.

388

389 **3.2 Discussion**

390 Among the 122 civilians sampled in this study, 22 (18%) had one or more compounds on their hands
391 and 14 (11.5%) on their wrists or sleeves. Compared to other studies, such prevalence appears to be
392 relatively high. Bell and Seitzinger obtained less than 5% positive results for a sample of 73 people
393 [35]. Two other studies by Northrop [19] and Hofstetter *et al.* [36] found no positives in population
394 samples of 100 and 27 individuals respectively. Three factors might explain this difference:

- 395 - the targeted country
- 396 - the analytical technique
- 397 - the sampled population

398 Bell/Seitzinger and Northrop collected specimens in the United States, while both the present study
399 and that from Hofstetter *et al.* were carried out in Switzerland. While Switzerland is a country where
400 many citizens own a gun due to mandatory military service, shooting sport and hunting tradition, the
401 United States is also known for a relatively high firearm possession. Indeed, the firearm density in
402 Switzerland has been estimated to 27.6 firearms per 100 inhabitants (firearm possession in the present
403 study was 22.1), whereas a value of 120.5 was given for the United States [1]. Thus, the targeted
404 country is probably not the main influence factor for OGSR prevalence.

405 Three different analytical techniques were used to study prevalence in previous studies. Bell and
406 Seitzinger used IMS, Northrop used MEKC and Hofstetter *et al.* used LC-MS for their respective
407 studies. The sensitivity can vary considerably between analytical approaches. For example, IMS is the

408 least sensitive, with reported LOD for DPA and EC of 50 and 1 ng respectively [42], while using LC-
409 MS/MS, LOD of 0.5 and 0.01 ng can be attained, respectively. Capillary electrophoresis is the most
410 sensitive technique in terms of amounts of injected analyte with a LOD of 1.8 pg and 0.9 pg for EC
411 and DPA, respectively [19]. However, the injection volume is also much lower (in the nanoliter range)
412 than those used with LC-MS/MS instrumentation and LOD should be compared in terms of
413 concentrations and not amounts loaded in the capillary or column. As the exact injection volume was
414 not indicated in Northrop study, the better sensitivity of LC-MS vs CE can only be extrapolated. Thus,
415 sensitivity issues might explain the differences observed between Bell and Seitzinger's study,
416 Northrop's and the present study.

417 Finally, the sample size and selection can also have a considerable influence on the results. Indeed, the
418 number of individuals involved in the studies remained very limited (from 27 to 122) and might not be
419 representative of the whole population due to the absence of randomization in the participant selection.
420 For example, in the present study 68 students were involved (55.7% of the whole dataset), yielding a
421 much younger population in comparison to the whole country. Consequently, larger prevalence studies
422 would allow a more accurate picture of OGSR prevalence. It might also be interesting to target
423 populations close to a person of interest in casework, as the prevalence might vary with the living area
424 (city/countryside, known firearm violence, etc).

425 This study highlighted the possibility to detect several OGSR compounds on an individual without
426 reported contact with firearms. Thus, while most positive results with three or more compounds could
427 be explained by the questionnaires, one individual had four compounds on his sleeves without
428 explanation (i.e. did not report any contact with firearms or potential secondary transfer). Given the
429 other results from the study, such a high number of OGSR-related compounds supports the hypothesis
430 of primary or secondary transfer rather than an alternative source. On the contrary, the presence of a
431 single OGSR compound is more frequent and can derive from an alternative source. For example,
432 DPA has multiple applications in the perfume industry and is used to prevent oxidation in rubber and
433 scald of some fruit crops [43]. Goudsmits *et al.* compiled the results of a number of OGSR
434 publications and listed the compounds appearing frequently in propellant formulations and having few
435 alternative sources [31]. According to their study, EC, MC, nitroglycerin and nitroguanidine might be
436 the most relevant OGSR targets. While our results confirm that EC and MC were very rarely detected
437 in a civilian population, nitroglycerin and nitroguanidine were not targeted. The first can only be
438 detected in negative mode, which was not used in this study, and the second is only found in triple
439 base smokeless powders that are exclusively used in large calibre ammunition.

440 Logically, the police population prevalence was noticeably higher, with 36.5% and 33% of the hand
441 and wrist specimens testing positive for one or more compounds respectively. A previous study
442 investigating the prevalence of OGSR for police officers found only two people positive to OGSR out
443 of 25 (7.4%) [36]. However, the population was composed of people working in a forensic laboratory
444 and not police officers holding a service weapon and practicing shooting, which might explain the

445 difference in prevalence. Contamination in the police environment is not limited to police officers, as
446 Ali *et al.* also demonstrated the presence of OGSR in police stations and vehicles [37]. Our study also
447 highlights that a vehicle locksmith who installed material in the trunk of a police vehicle had two
448 OGSR compounds on his hands and on his sleeves, indicating a potential secondary transfer from a
449 contaminated source. It would be interesting to collect more information regarding OGSR background
450 level in police premises, where a suspect might be brought during investigation. Indeed, if there is a
451 non-negligible background level, secondary transfer to people transported and/or detained in police
452 premises might occur.

453 By comparing both populations, one can observe that OGSR prevalence is about twice as high for
454 police officers (\geq one compound: 36.5% on hands and 33% on sleeves) than for civilians (\geq one
455 compound: 18% on hands and 11.5% on sleeves). This is expected, as most civilians do not have
456 regular contact with firearms. In the civilian population, the prevalence decreased as the number of
457 compounds detected together increased. Only four individuals (less than 5%) had two or more
458 compounds on the hands and/or sleeves (if we exclude the person who discharged a firearm less than
459 10 minutes before specimen collection). In the police population, the trend was dependent on the time
460 elapsed since discharge. Indeed, the highest numbers of compounds were in most cases observed for
461 individuals who discharged a firearm very recently (or explained through secondary transfer). After 24
462 hours, the distribution in the police population was more similar to the civilian one. In terms of
463 compound prevalence, the two most frequently detected compounds were AK II and 4-nDPA in the
464 civilian population, whereas *N*-nDPA and EC were the most frequently detected in the police
465 population. It is interesting to note that in the police population, most of the one compound positive
466 results (hands and sleeves considered together) concerned AK II, potentially indicating a longer
467 persistence for that particular compound. On the contrary, MC was very rarely detected in both
468 populations. As discussed above, finding traces of one OGSR compound on the hands of an individual
469 can occur without an explanation. Indeed, alternative sources were previously reported for some
470 OGSR compounds, such as DPA. In this case, how relevant would be the detection of DPA alone? As
471 highlighted by others [10, 18], the simultaneous detection of several compounds has greater value than
472 that of a single compound. This was also confirmed in the present study, as the detection of more than
473 three OGSR compounds together was generally related to a direct or indirect contact with firearms.
474 The only exception was a civilian with four compounds on his sleeves. However, while there was no
475 explanation as to the exact source of this result, its origin is more likely to be from a firearm than from
476 an environmental source. In practice, it would be important to investigate the risk of contamination
477 through secondary transfer. One should keep in mind that the number of OGSR compounds that is
478 detected simultaneously also depends on the combination firearm-ammunition, that is to say the
479 number of compounds present in the propellant and a potential memory effect. Hence, the case-by-
480 case approach proposed by Romolo and Margot recommending to evaluate evidence in light of case
481 circumstances and using whenever possible comparison of GSR with other case-specific items such as

482 the cartridge case is particularly attractive [44]. Finally, some of the civilians positive to OGSR and
483 without contact with firearms were vehicle locksmiths. It would then be advisable to look for
484 alternative sources in the automotive industry. It must be noted that vehicle parts, more particularly
485 brake pads [45-47], were also mentioned as potential alternative sources for IGSR, though a recent
486 study indicated that it is not a concern anymore [48].

487

488 **4. CONCLUSIONS**

489 The present work investigated the prevalence of eight OGSR compounds in a civilian and police
490 population to provide information for the assessment of that trace. Specimens were collected on the
491 hands and sleeves of 122 civilians and 115 individuals working in a police environment using carbon
492 stubs. One stub was used to sample both hands and the other was used to sample the cuffs of the
493 sleeves or the wrists when a short-sleeve upper garment was worn. Data was acquired using liquid
494 chromatography coupled to tandem mass spectrometry (LC-MS/MS) with electrospray ionisation in
495 positive mode.

496 Prevalence in the civilian sample was relatively high, with 18% of the 122 civilians sampled having
497 one or more compounds on their hands and 11.5% on their wrists or sleeves. More than three
498 compounds were detected simultaneously from only two individuals. Using data from the
499 questionnaire, it was possible to explain one result by a direct contact with firearms. However, no
500 explanation was found in the questionnaire for the presence of four compounds on the sleeves of the
501 second person. Results also showed that secondary transfer (through handshake or service weapon
502 handling) can explain OGSR contamination. The detection of two compounds on three vehicle
503 locksmiths might indicate alternative sources (possibly related to occupations in the automotive field).
504 Further research is required to investigate such alternative sources.

505 For the police population, the prevalence was higher, with 36.5% of the hand specimens and 33% of
506 the wrist specimens positive to one or more compounds. A higher prevalence was expected for this
507 population due to service weapon holding and regular shooting exercises. In the civilian population,
508 the prevalence decreased as the number of compound detected simultaneously increased, whereas in
509 the police population, the trend was dependent on the time elapsed since the last shooting exercise.
510 Indeed, the simultaneous detection of four or more compounds was in most cases observed for
511 individuals who discharged a firearm very recently (< 10 minutes). Due to the relatively high
512 prevalence of OGSR on police officers, it would be interesting to target their premises (police stations
513 and vehicles for example) to evaluate the background contamination. This is critical, as an arrested
514 person might be contaminated by secondary transfer from a contaminated vehicle, a police officer or
515 even a desk at a police station. In any case, it would serve as a quality assurance tool for laboratory
516 contamination monitoring if OGSR is implemented in routine work in parallel to IGSR analysis. From
517 the present results, it appears that there is an important prevalence for single compounds. The

518 simultaneous detection of two compounds shows a lower prevalence and the detection of three or
519 more compounds supports the hypothesis of a firearm origin rather than an environmental source.
520 However, results should be interpreted on a case-by-case basis, as the number of compounds that can
521 be detected depends on the composition of the propellant.

522 Further prevalence studies on a larger scale should be carried out to avoid bias in participant selection
523 and provide a more representative picture of the population. The present study was performed in ESI
524 positive mode and detecting additional compounds in the negative mode, such as dinitrotoluenes and
525 nitroglycerine, would be valuable. Further research for OGSR environmental sources is also required
526 and should be combined with prevalence data to establish the most relevant compounds and evaluate
527 what combinations of compounds are the most characteristic for firearm discharge. Such information
528 would be particularly useful for OGSR interpretation in casework.

529

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531

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539

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