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REVIEW

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Spectroscopic (analytical) approach to gunshot residue analysis for shooting distance estimation: a systematic review

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Abstract

Background: The determination of the shooting distance using gunshot residue (GSR) analysis is crucial in the investigation and reconstruction of firearm-related crimes. However, the conventional chemographic method for GSR analysis is destructive and has limited sensitivity and selectivity. While the spectroscopic method has potential in GSR analysis for crime investigation, there is a current lack of consistency in the spectroscopic results obtained for shooting distance estimation via GSR analysis. Addressing such limitations will enhance the forensic capabilities of law enforcement and provide an added advantage to crime laboratories during an investigation. It will also reinforce the use of such spectroscopic data in a criminal investigation.

Main text: We obtained all peer-reviewed articles relevant to shooting distance estimation from searching Scopus, Web of Science, PubMed, and Google Scholar databases. We specifically searched the databases using the keywords "shooting distance," "range of fire," "gunshot residue," "firearm discharge residue," and "firearm-related crime" and obtained 3811 records. We further filtered these records using a combination of two basic keywords "gunshot residue" and "shooting distance estimations" yielding 108 papers. Following a careful evaluation of the titles, abstracts, and full texts, 40 original peer-reviewed articles on shooting distance estimation via GSR analysis were included in the study. The forgoing included additional sources ($n = 5$) we obtained from looking through the reference lists of the forensic articles we found.

Short conclusion: This paper discusses the current scope of research concerning the chemographic and spectroscopic analysis of GSR for shooting distance estimation. It also examines the challenges of these techniques and provides recommendations for future research.

Keywords: Forensic ballistics, Forensic science, Gunshot residue, Firearm discharge residue, Shooting distance estimation, Range of fire

Background

Firearm-related crimes (FRC) such as armed robbery, homicide, suicide, and mass shootings threaten global public security and safety (Hemenway and Miller 2000). An estimated 18,000 children and adolescents die yearly in the US via injuries resulting from accidental gun firing (Cunningham et al. 2018). The foregoing is a consequence of the ease of access due to lax regulations on firearm possession (Hemenway and Miller 2000) and the influx of improvised/makeshift, including 3D printed (Chase and Laporte 2018; Walther 2015) and illegal firearms. In Ghana, an estimated 1.1 million unregistered firearms are illegally in civilian possession and have been implicated in the recent surge in FRC (Bokpe 2016).

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Analysis of gunshot residues (GSR) from discharged firearms is vital in ballistic reconstruction in instances of FRC (Chang et al. 2013a, 2013b; Saverio Romolo and Margot 2001). Notably, the shooting distance, time since firearm discharge, ammunition type, trajectory or spatial position of the discharged firearm relative to its environment and target, injury pattern reconstruction, and linking an individual to gun use (Chang et al. 2013a, 2013b; Saverio Romolo and Margot 2001) can be estimated via GSR analysis. Gunshot residue mainly comprises burnt, unburnt, and partially burnt propellant charge, smoke, a cloud of vapor, and metal particles from the ammunition, and even the firearm (Dalby et al. 2010; López-lópez and García-ruiz 2014; Muller et al. 2007; Saverio Romolo and Margot 2001; Turillazzi et al. 2013). In addition, dispersion and deposition of GSR on the targets may be impacted by the environmental conditions and/or physical activities of the shooter (Zuzanna Brozek-Mucha 2011; Jalanti et al. 1999).

Consequently, shooting distance may be estimated by a visual comparison of casework GSR pattern with that generated from test-firing using the same firearm and ammunition involved in the crime (Capannesi et al. 1993; Dalby et al. 2010; López-lópez and García-ruiz 2014; Muller et al. 2007). Visualizing the GSR pattern on the victim of a crime is conventionally based on labor-intensive and subjective chemographic techniques (presumptive color test) (López-lópez and García-ruiz 2014). Despite their continued use in casework (Martiny et al. 2008), such color tests are not only destructive but have limited sensitivity and selectivity. Furthermore, the lack of efficiency in pattern enhancement of GSR from lead-free ammunition or nontoxic ammunition (Berendes et al. 2006) precludes recognition of chemographic tests as robust.

Analytical techniques based on spectroscopy have found utility in GSR analysis with promising results (Barth et al. 2012; Zuzanna Brozek-Mucha 2014; Cecchetto et al. 2011; Leiva et al. 2019; Santos et al. 2007; Zeichner 2003; Zeichner and Glattstein 2002). Particularly, the coupling of scanning electron microscope with energy-dispersive X-ray spectroscopy (SEM-EDS) has emerged as the technique of choice (Zuzanna Brozek-Mucha 2014; French and Morgan 2015). The SEM-EDS allows for both elemental composition and morphological examination of the characteristic primer GSR, typically lead (Pb), barium (Ba), and antimony (Sb). Additionally, the technique is also spatially discriminating, non-destructive, and requires no sample preparation (Z. Brozek-Mucha 2000; Zuzanna Brozek-Mucha 2009, 2014; Dalby et al. 2010; Saverio Romolo and Margot 2001).

The merits, notwithstanding, spectroscopic analysis of GSR for shooting distance estimation (SDE) remain inconsistent. For example, while Gagliano-candela, Colucci, and Napoli claimed that atomic absorption spectroscopy (AAS) could estimate shooting distances up to 100 cm from the target, Gradaščević et al. showed that AAS was only effective up to 10 cm. This review discusses the conventional and modern techniques of GSR analysis for SDE; it also identifies challenges and gap in existing methods and provide a perspective on future direction.

Main text

Literature collection

All peer-reviewed articles relevant to SDE were obtained from searching Scopus, Web of Science, PubMed, and Google Scholar databases. We specifically searched the databases using the keywords "shooting distance," "range of fire," "gunshot residue," "firearm discharge residue," and "firearm-related crime" and obtained 3811 records (Scopus, $n=10$; Web of Science, $n=33$; PubMed, $n=28$; Google scholar, $n=3740$). We further filtered these records using a combination of two basic keywords "gunshot residue" and "shooting distance estimation" yielding 108 papers. After screening the titles and abstracts, we only included full-text articles describing original research that focused on GSR analysis for SDE ($n=40$) in this study. The foregoing included additional sources obtained from the reference list of one original article ($n=5$). We excluded supplementary published works, reviews, conference abstracts, discussion papers, editorials, errata, short communications, correspondence, and news. The search strategy is shown in Fig. 1.

Results

GSR analysis for SDE

The distance between the gun's muzzle end and the target is termed shooting distance or range of fire or muzzle-to-target distance. In the investigation and reconstruction of the FRC, determining the shooting distance is critical. The GSR exits the firearm through the muzzle end (and other openings in the firearm), travels a little distance before settling on the victim, and surfaces close to the firing site (Gallidabino et al. 2016). As a result, the relevance of the distance traveled by these residues cannot be ignored, given its critical significance in the investigation and reconstruction of shooting incidents. The methods used for GSR analysis for SDE can be categorized as chemographic or spectroscopic.

Chemographic analysis of GSR for SDE

The chemographic methods (Table 1) for GSR analysis relies on a color formation following a chemical reaction between a specific reagent and the metal elements in the GSR sample (Ananth et al. 2014; Chang et al. 2013a, 2013b; Niewoehner 2015; Saverio Romolo and Margot

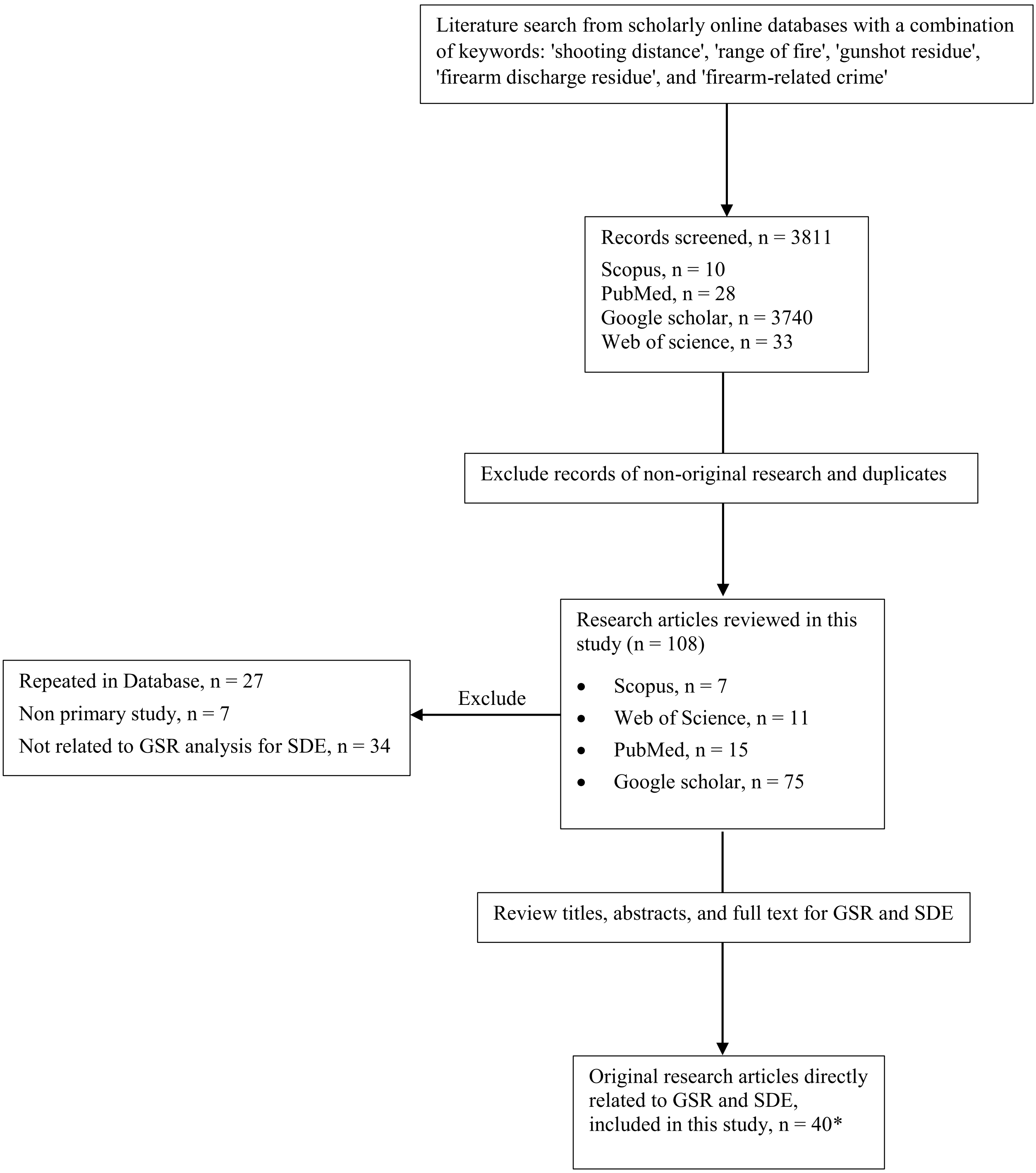

Fig. 1 Flow chart of the systematic literature search scheme employed; asterisk symbol indicates the following: includes 5 additional sources from reference lists

2001; Schwoeble and Exline 2000; Tugcu et al. 2006; Zeichner and Glattstein 2006; Niewoehner 2015; Dalby et al. 2010). Thus, the technique is suitable for detecting both inorganic (metallic) and organic elements in GSR particles.

Although the chemographic method is still used in casework for presumptive identification of a sample as

Table 1 Chemographic method for GSR analysis

Reference	Test type	Compound detected	Reagent used	Description/color reaction
Ananth, Kalthom, and Me (Ananth et al. 2014)	Dermal nitrate/paraffin test	Nitrate	Diphenylamine dissolved in strong sulfuric acid	Picked up GSR on paraffin wax + reagent gives a blue color spot indicating the presence of nitrates
Chang, Jayaprakash, and Yew (Chang et al. 2013a, b)	Spot test	Lead (Pb)	Sodium rhodizonate	GSR sample + reagent gives a pink coloration indicating the presence of lead.
Saverio Romolo and Margot (2001)	Walker test/Marshal and Tewari test	Nitrite	2-Naphthylamine sulfanilic acid and citric acid	GSR sample + reagent gives a pink coloration for the presence of nitrite
Ananth et al. (2014)	Modified Griess test	Nitrite	Griess reagent[a]	GSR sample + reagent gives a rose color for the presence of nitrite
Dalby et al. (2010)	Harrison and Gilroy's test	Lead (Pb), barium (Ba), and antimony (Sb)	• Triphenyl methylarsonium iodide alcoholic solution • Sodium rhodizonate • Dilute hydrochloric acid	GSR sample + triphenyl methylarsonium iodide alcoholic solution gives orange ring color for the presence of Sb. Two drops of sodium rhodizonate are added to the center of the orange ring, color changes to red for Pb or Ba or both. A drop of dilute hydrochloric acid is added to the red spot, color changes to blue for Pb. No color change is indicative of Ba
Ananth et al. (2014)	Lunge test[b]	Nitrocellulose (NC)	Lunge reagent	GSR sample + reagent gives a red azo color for NC
Niewoehner (2015)	Zincon reagent test[c]	Zinc (Zn) and titanium (Ti)	Zincon reagent	GSR sample + reagent gives a blue-colored complex for Zn and Ti

[a] Griess reagent: 3% sulfanilamide N-(1-naphthyl) ethylene diamide dihydrochloride in 5% sulfuric acid

[b] Detect organic compounds (NC) in GSR

[c] Applied to Pb-free ammunition, e.g., Sintox ammunition

GSR, its application in SDE has been limited and problematic. For instance, Marty, Sigrist, and Wyler determined the shooting distance (differentiated between close range and far range shot) using the rhodizonate staining technique on skin samples. The use of skin samples in this scenario increases the likelihood of generating a false positive result since the sodium rhodizonate reagents also produce positive results with keratinous structures of hair follicles (Andreola et al. 2011). The above technique was improved further via a heated press for the transference of GSR particles on clothing items for SDE (Geusens et al. 2019). Furthermore, Vinokurov, Zelkowicz, Udi, and Zeichner proposed a technique based on the influence of contamination of a victim's clothing by GSR for a range of fire estimation using the modified Griess test (MGT) method. The detection of nitrite using the MGT method follows the reaction of the GSR sample with potassium hydroxide solution plus the MGT reagent (Atwater et al. 2006). While the MGT is effective, a method that reveals the GSR pattern on an item without chemicals or reagents would be preferable. Not only would this save time on reagent preparation, but it will also alleviate contrast problems when the GSR is present on a multi-colored fabric or target (Bailey et al. 2004).

Spectroscopic analysis of GSR for SDE

The spectroscopic technique deals with the interaction between specific electromagnetic radiation and the GSR sample. This technique estimates the concentration and yields information about the elemental composition (for example, Pb, Ba, and Sb) of the GSR sample. Spectroscopic techniques employed for GSR analysis to estimate the shooting distance are presented in Table 2.

One of the earliest studies on SDE evaluated the concentration and pattern of Sb deposited around a bullet entrance hole using neutron activation analysis (NAA) at a firing distance of 16.5 cm and 26.7 cm for a first and second shot, respectively (Capannesi et al. 1993). Neutron activation analysis (NAA) is an isotope-based technique that works by bombarding stable atomic nuclei

Table 2 Spectroscopic analysis of GSR for SDE

Reference	Firearm/ammunition used	Technique	Target	Findings
Capannesi et al. (1993)	Not stated	NAA	• White cotton cloth	• Quantified Sb concentration around a bullet entrance hole at a firing distance of 16.5 cm (first shot) and 26.7 cm (second shot)
Santos et al. (2007)	6.35 mm pistol	ICP-MS	• Cotton tissue	• Quantified Pb, Ba, and Sb at a firing distance of 20 cm to 80 cm from the target at a radial distance of 1.5 cm to 5.5 cm from the bullet entrance hole
Gagliano-candela et al. (2008)	Colt 38 special	AAS	• Whatman paper on polystyrene support	• Determined the concentration of Pb on a target at a firing distance of up to 100 cm
Mou et al. (2008)	Remington model 552 rifle (0.22 caliber)	AFM and FTIR/ATR	• Polyethylene and aluminum foil sheet	• Characterized GSR particles at a firing distance of 0.5 ft, 2 ft, 10 ft, and 20 ft • AFM images revealed varying shapes of the GSR particle with size and density inversely proportional to shooting distance
Bresson and Franck (2009)	Glock 26 semi-automatic pistol	Ballistic experiment	• Concrete wall	• Estimated the shooting distance from the impacting velocity of a bullet
Cecchetto et al. (2011)	7.65 mm pistol	Micro-CT	• Human leg	• Firing distance up to 40 cm was estimated through GSR analysis in an intermediate gunshot wound
Barth et al. (2012)	Glock model 19 semi-automatic pistol with different ammunition (9 mm × 19 GECO FMJ, 9 mm × 19 Sintox FMJ, and 9 mm × 19 Sintox forensic FMJ)	m-XRF	• 100% cotton denim • A mixture of knitted 50% polyester and 50% acrylic fiber • 100% acetate satin	• Estimated the range of fire up to 80 cm from the target
Turillazzi et al. (2013)	9 × 21 mm and 7.65 mm caliber handgun	Laser confocal microscopy and ICP-AES	• Pig skin	• Obtained a greater concentration of the elemental trio (Pb, Ba, and Sb) in a close-range shot relative to mid and far-range shots for both firearms
Gradaščević et al. (2013)	Pistol Crvena zastava M70/7.65 × 17 mm; Pistol Crvena zastava M57/7.62 × 25 mm; 9 mm luger pistol/9 × 19 mm; automated rifle/7.62 × 39 mm	AAS	• Pig skin	• Estimated shooting distance in contact (5 cm) and near contact (10 cm) shot using GSR obtained from a bullet entrance hole
Hinrichs et al. (2016)	.38 special revolver/.38 cartridge	VP-SEM	• Cotton and polyester textiles	• Obtained the pattern of GSR distribution with low vacuum SEM images • Estimated firing distance using the decay length of GSR distribution pattern
Zapata et al. (2018)	Glock G17 pistol and 9 × 19 mm semi-jacketed hollow point ammunition	Multi-spectral imaging	• Cloth (white cotton fabric)	• Estimated the shooting distance of up to 220 cm
Leiva et al. (2019)	Revolver Taurus caliber with .38 ammunition	XRD	• Cotton-polyester fabric	• Quantified GSR and estimated the shooting distance up to 300 cm
Pyl et al. (2019)	9 mm pistol; Rossi revolver 0.357 magnum	LIBS	• Cotton textiles	• Produced permanent chemical images of residues for the correct classification of shooting distances

with neutrons (from a nuclear reactor) and measuring the energy and half-life of the emitted radiation (radioactive gamma rays) following the decay process (Greenberg et al. 2011). During GSR analysis, the technique exclusively measures the presence of radioactive Sb ($Sb_{121} \rightarrow Sb_{122}$) and Ba ($Ba_{132} \rightarrow Ba_{133}$). It is particularly sensitive and capable of analyzing difficult to dissolve elements (such as inorganic GSR) without sample pretreatment. However, apart from the relatively slow sample throughput, the method does not apply to the analysis of Pb and requires a powerful neutron source like a nuclear reactor. These limitations notwithstanding, NAA has been used in GSR analysis for forensic investigations (Chohra et al. 2015). The inductively coupled plasma-mass spectroscopy (ICP-MS) platform allows the examiner to obtain a very low detection limit due to its high sensitivity and facilitates rapid multi-elemental analysis, giving both qualitative and quantitative outputs (Biegstraaten and Horváth 2007). Excepting the high cost and the occurrence of spectral and non-spectral interferences during sample analysis, ICP-MS has proven invaluable for GSR analysis. Santos et al. used ICP-MS to profile the simultaneous presence of GSR elemental trio (Pb, Ba, and Sb) at a firing distance of not more than 80 cm away from the target. This finding supports the work of Barth et al. whose study estimated the shooting distance from GSR using milli-X-ray fluorescence (m-XRF). Unlike the ICP-MS, m-XRF is non-destructive, does not consume samples under examination, and efficiently detects and visualizes particles from Pb-free or nontoxic ammunition (example, sintox ammunition). Leiva et al. also estimated the shooting distance (between 5 and 300 cm) using x-ray diffraction (XRD) analysis.

Turillazzi et al. employed confocal laser microscopy and inductively coupled plasma with atomic emission spectroscopy (ICP-AES) to estimate the shooting distance and reported obtaining a greater concentration of Pb, Ba, and Sb in a close-range shot relative to mid and far range shot. The elemental trio (Pb, Ba, and Sb) establishes the identity of a sample as GSR. Further, a combination of micro-computed tomography (micro-CT) and image analysis software was used to quantify GSR present in a mid-range gunshot wound for SDE ($\leq$ 40 cm) (Cecchetto et al. 2011). Although little data was presented on the elemental trio (Pb, Ba, and Sb) unique to primer GSR, the authors used human skin samples, representing an ideal simulation of real-life crime scene scenarios. Whereas the micro-CT technique is non-destructive (Cnudde et al. 2008), it has an increased likelihood of providing false-positive results (Cecchetto et al. 2011). Additionally, a multi-spectral imaging technique was effectively used for estimating the shooting distance (from 10 to 220 cm) following GSR analysis (Zapata et al. 2018). This technique has the added advantage of automatically examining GSR patterns through quantitative approaches.

Atomic absorption spectroscopy (AAS) works by measuring the amount of electromagnetic radiation (ultraviolet-visible radiation) absorbed by free atoms in a gaseous state (Yeung et al. 2017). A study by Gagliano-candela, Colucci, and Napoli concerning SDE extrapolated the firing distance (up to 100 cm) by analyzing the concentration of only Pb on a target using AAS. This result is consistent with the work of Gradaščević et al. (2013). Mou, Lakadwar, and Rabalais employed atomic force microscopy (AFM) and Fourier transform infrared attenuated total reflectance (FTIR/ATR) for SDE. Although the firing distance was estimated at 0.5 ft (15.24 cm), 2 ft (60.96 cm), 10 ft (304.8 cm), and 20 ft (609.6 cm) away from the target, the authors argued that characterization of the specific element in the GSR sample was not successful with FTIR/ATR. The FTIR/ATR technique is very sensitive, considering its ability to quantify GSR particles at a range up to 20 ft (609.6 cm) from the target. The FTIR/ATR can also produce a profile unique to the ammunition type, making it a valuable tool for differentiating ammunitions based on the GSR particles (Mou et al. 2008).

The impact velocity of a full metal jacketed (FMJ) 9 mm Parabellum bullet was determined through a series of shooting experiments by comparing a questioned bullet to a set of test-fired bullets striking a wall (Bresson and Franck 2009). The firing distance was subsequently estimated from the impact velocity after evaluating the bullet's behavior (Bresson and Franck 2009). A variable pressure scanning electron microscope (VP-SEM) with characteristic high-resolution imaging and depth of field was introduced by Hinrichs et al. for SDE. This was achieved by establishing an exponential decay of the GSR distribution pattern on a target as a function of the radial distance of the particles from the bullet entry hole (Hinrichs et al. 2016). Pyl et al. showed that laser-induced breakdown spectroscopy (LIBS) is suitable for SDE. The LIBS performs better than the conventional chemographic technique in sensitivity, selectivity, reproducibility, accuracy, and multi-elemental detection capabilities (Pyl et al. 2019).

Limitations and future directions

Current research on GSR analysis for SDE has focused on collecting samples after test shots/firing in a controlled, indoor firing range. Whereas the findings from such studies are useful as proof of concept, it lacks practical applications in forensic casework scenarios where shootings may occur in an outdoor space exposed to natural environmental conditions. Therefore, extensive research is needed to study and understand the influence of environmental factors such as wind, temperature, rain, and humidity on the persistence and collection of GSR from a target. Further, the firearms and ammunition, range of fire, and spectroscopic technique employed varied from one study to another. Although such an approach is laudable, it has limited reproducibility for SDE. Hence, ensuring consistency and reliability in the spectroscopic results acquired for SDE will reinforce the use of such spectroscopic data in a criminal investigation. Also, extensive research and collaboration among practitioners are required to address the current lack of consensus on validated protocols, quality standards, and guidelines for interpreting spectroscopic results following GSR analysis for SDE.

Conclusions

The determination of shooting distance is a critical problem in firearm-related incidents where GSR is the only available evidence of probative value. Establishing the muzzle-to-victim distance through GSR analysis may contribute to linking a suspect to a shooting scene. However, the application of spectroscopy for SDE via GSR analysis in casework is limited due to the inconsistency in methods and lack of standardization and guidelines for interpreting results. In this paper, we have discussed the current scope of research concerning the chemographic and spectroscopic analysis of GSR for SDE, highlighted some limitations, and suggested further research directions to address them. Addressing such limitations will enhance the forensic capabilities of law enforcement and provide an added advantage to crime laboratories during an investigation.

Abbreviations

FRC: Firearm-related crime; GSR: Gunshot residue; SDE: Shooting distance estimation; AAS: Atomic absorption spectroscopy; SEM-EDS: Scanning electron microscope with energy-dispersive X-ray spectroscopy; ICP-MS: Inductively coupled plasma with mass spectroscopy; ICP-AES: Inductively coupled plasma with atomic emission spectroscopy; NAA: Neutron activation analysis; m-XRF: Milli X-ray fluorescence; XRD: X-ray diffraction; AFM: Atomic force microscopy; FTIR/ATR: Fourier transform infrared attenuated total reflectance; FMJ: Full metal jacketed; VP-SEM: Variable pressure scanning electron microscope; LIBS: Laser-induced breakdown spectroscopy; MGT: Modified Griess test; NC: Nitrocellulose; Pb: Lead; Ba: Barium; Sb: Antimony; Zn: Zinc; Ti: Titanium.

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MA and DOMB contributed to the conception of the article, research for the article, drafting of manuscript, review, and editing of the manuscript; IKB, RA, and PA contributed to the drafting of paper, review, and editing of the paper. All authors read and approved the final manuscript.

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