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Research Article

Investigating ambient air quality of a shooting range during official national competitions

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ABSTRACT

Shooting is among nine sports branches that formed the first modern Olympic Games in Athens in 1896. A professional shooting athlete shoots millions of bullets throughout their sports life to commune with their gun dedicatedly. The number of simultaneous shots per unit time depends on the capacity of the range. It can enormously increase when a national match is a case. Shooting can cause gunshot residue exposure, including lead, other elements, and their by-products accumulate in ambient air and reveal significant health risks. This study aims to find the levels of PM_{10} and its chemical composition during official three-day 50 m 22-cal competitions in May 2016, Mersin, Turkey. To this end, PM_{10} samples were collected on quartz-fiber filters and analyzed for elements by ICP-MS and carbonaceous material by Thermal-Optical EC/OC analyzer. The total PM_{10} mass concentration average is 28.7±7.3 µg/m^3 within the indoor threshold values of different countries. The ambient mass concentrations of PM_{10}, OC, EC, TC, Cd, and Pb were higher during once pistol matches instead of rifle matches. Although Pb values did not exceed the indoor limits for shooting ranges, it has the highest concentration among the analyzed elements. Additionally, Cr poses cancer risk potential. Except for Zn, Sr, and Cu, all the measured parameters have higher calculated emission factor (EF) values during pistol shots. To our best knowledge, this study reports the airborne mass concentrations of EC, OC, and TC from indoor shooting ranges and investigates indoor air quality for shooting sport for the first time.

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INTRODUCTION

Air pollution, which accounts for 1 in 8 deaths in 2012 according to WHO [1], is one of the most significant environmental issues in terms of human health. People are prone to high exposure risk from air pollution in indoor and outdoor environments, even at low concentrations [2]. Indoor air quality (IAQ) has been paid attention to by researchers due to the time spent in indoor environments. IAQ has been considerably influencing on well-being and productivity of people, while indoor air pollutants are increasing the risks for a variety of diseases. Jenkins et al. [3] reported that people spend 87% of their time indoors while only 6% outdoors and 7% in transit. The performed

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studies starting in the middle of the 1970s put forward that indoor concentrations of several pollutants were significantly higher than their corresponding outdoor concentrations [4].

Considerable IAQ studies on schools [5–8], elderly care centers [9, 10], homes [11–15] hospitals and nursing homes [9, 16, 17], and offices [18, 19]. On the other hand, researchers recently paid attention to the IAQ of environments used for physical exercise and sports [20–25]. Like other indoor places, construction materials, maintenance, and ventilation types form IAQ in sports centers. Moreover, higher human occupancy and the type of acts performed in the centers make them peculiar. During exercise, the respiratory ventilation per minute rises, which leads to inhalation of more air and pollutants existing in the air. Consequently, metabolic reactions to physical exercise open the human body to an elevated amount of pollutants [26]. The nasal particle-filtering system is not used during training since air is inhaled through the mouth. This process causes an increase in airflow velocity, which results in the movement of pollutants to most parts of the respiratory system and produces more risk to human health [26]. According to Ramos et al. [24], people who conduct physical exercise in polluted environments put their health at risk. Shooting is a bit tricky at this point. It is well known that a shooter's heart rate is considerably lower during shooting than in daily life. They use abdominal breathing; additionally, they inhale and exhale by their nose during aiming and triggering, not their mouth.

IAQ studies performed in sports centers revealed that occupants expose to various air pollutants, including particulate matter (PM), combustion-related emissions such as carbon monoxide (CO) and nitrogen dioxide (NO_2), carbon dioxide (CO_2). Additionally, biological pollutants such as dust mites, molds, fungus, and bacteria; volatile organic compounds (VOCs), for instance, formaldehyde and benzene; inorganic chemicals, for example, chlorinated compounds (mainly in swimming pools); heavy metals such as lead and mercury; and asbestos, which is primarily due to the building materials [27–29]. These studies focus on fitness centers [24, 30, 31] and gymnasiums and sports facilities in educational premises [32–35]. Andrade and Dominski [20] reviewed the studies performed on IAQ of places used for sports. Authors reported that the gymnasium, fitness and sports centers, and ice-skating rinks, were the most investigated places in the reviewed studies. Moreover, CO, NO_2, and PM were the IAQ parameters primarily measured in these indoor environments. On the other hand, the literature on the level and composition of emissions from firing ranges as indoor sport and recreational activity is scarce except for a few studies [36–44]. Shooting at firing ranges has become very popular among people as a recreational activity in many countries. 16,000–18,000 indoor firing ranges in the United States alone and 20 million people nationwide exercise target shooting for leisure [45]. Recreational, in other words, private sector pistol shooting ranges are about 30 thousand currently in Turkey [46]. Despite this, ranges for licensed athletes are very rare. The only world-cup standard shooting range in Turkey is located in Mersin, Erdemli. Including Erdemli shooting range, there are about 70 shooting ranges in Turkey in different cities and, these shooting ranges are operated by the Turkish Ministry of Youth and Sports [47]. A large number of these are outdoor shotgun ranges also used for recreational purposes. Only a few indoor shooting ranges are used by licensed shooting athletes, primarily for training. Operational liabilities, including ventilation, cleaning, transportation of athletes, belong to the provincial directorates. In order to reduce operating costs, unfortunately, electricity expenses such as ventilation are the first items to be reduced.

Indoor firing ranges are enclosed facilities, which have a unique operation. Improper use and design of indoor firing ranges could lead to adverse effects on human health even though military and civilian personnel prefer them for their controlled environment to outdoor counterparts. Well documented in the literature that mainly metals and gaseous compounds increase to high concentrations in the air and floor during shooting activities [38, 42, 48]. Not only major combustion gases such as carbon dioxide (CO_2) and water vapor (H_2O) but also carbon monoxide (CO), hydrogen cyanide (HCN), ammonia (NH_3), nitrous oxides (NO_x), sulfur dioxide (SO_2), and hydrogen chloride (HCl) are released during shooting [49, 50]. In addition, particulate matter (PM) consists of soot and metals, for instance, lead (Pb), copper (Cu), zinc (Zn), and iron (Fe), along with trace quantities of chromium (Cr) and molybdenum (Mo) are emitted to the indoor environment as a result of shooting activities [49]. Elevated concentrations of these airborne compounds in indoor environments cause serious health issues in occupants of indoor firing ranges.

Precisely at this point, we need to explain deeper the mechanism when a shooter pulls the trigger. A professional shooting athlete shoots millions of bullets, cartridges, or pellets throughout their shooting life, approximately starting at 13. Ammunition differs among shooting branches as lead pellets for air guns, cartridges for shotguns (rifle), and bullets for rifled guns. As in our case, in 50 m and 25 m competitions, .22-cal bullets are suitable for specialized rifles and pistols. According to the gun type, when the shooter pulls the trigger, the firing pin drops to the bullet jacket, bullet core leaves from the jacket. Due to the rifling inside the gun, the core aerodynamically travels through the shooting line till it hits the target. If the firing pin hits the jacket from the center, it is called center-fire. Otherwise, if it hits from the side part, it is called rim-fire. Compared to the center-fire bullet, less gunpowder and bullet

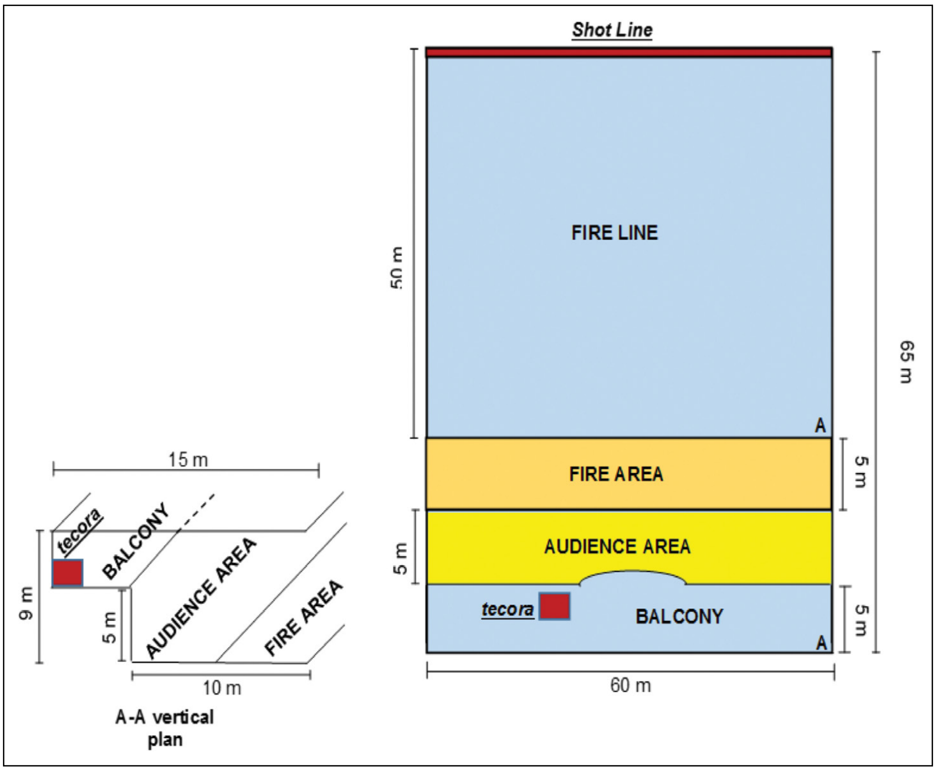

Figure 1. Schematic description of the shooting range.

materials are used in the rim-firing system. As the explosion is more efficient between jacket and core, emission rates and residues are reduced in rim-fire bullets. RWS R 50® rim-fire bullets are officially used at Turkish Shooting & Hunting Federation's competitions [51, 52].

In order to explain the health effects of shooting, Laidlaw et al. [45] reviewed thirty-six articles published in the literature and evaluated the shooters' blood lead levels (BLLs) at firing ranges. Researchers revealed that all BLL measurements exceeded the reference level of 5 µg/dL recommended by the U.S. Centers for Disease Control and Prevention/National Institute of Occupational Safety and Health (CDC/NIOSH). There is sufficient evidence that BLLs <10 µg/dL are caused to essential tremors, hypertension, cardiovascular-related mortality. Additionally, electrocardiography abnormalities and decreased kidney glomerular filtration rate among adult men and women while that <5 µg/dL leads to decreased fetal growth for an adult woman. Moreover, there is "sufficient evidence" that BLL <5 µg/dL caused several problems in children, such as declined academic achievement and intelligence quotient (IQ), reduced perinatal growth [53]. Most studies focused on the Pb because projectiles and primers contain a considerable Pb, and a huge Pb becomes airborne during shooting activities [54, 55]. International Agency for Research on Cancer (IARC) [56] classified Pb as class II (B) carcinogens, that is, possible human carcinogens. Similar to Pb, other metals emitted during shooting activities are associated with adverse health effects. Arsenic, Ni, Cd, and Cr were classified as class I carcinogenic contaminants by the IARC, while Zn, Cu, and Mn were classified as non-carcinogenic contaminants [57]. Residential exposure to a low level of Cd is related to renal toxicity, osteoporosis, and bone fractures [58]. People exposed to As by inhalation depicted an excess risk of lung cancer [59]. Moreover, upon inhalation, Zn may destroy plasmid DNA [60]. Based on epidemiological studies, it has been revealed that elemental carbon (EC) is correlated with cardiovascular and respiratory hospitalizations [61], preterm birth [62], and mortality [63]. Likewise, studies that measured organic carbon (OC) have found associations between respiratory outcomes and OC [64] and associations between cardiovascular outcomes and OC [65].

24-hr indoor PM_{10} samples were collected in three-day official competitions in this study. Collected samples were analyzed for elements and elemental and organic carbon (EC and OC, respectively). To our best knowledge, this is the first study reporting the airborne concentrations of elements, EC, and OC in a firing range in our country. This study is also unique since no indoor EC, and OC data from shooting activities have yet been produced in the literature. Another critical point is that the Mersin/Erdemli shooting range is neither commercial, recreational, nor military. So, occupants are licensed shooting athletes, their families, official coaches, referees, and audiences.

Table 1. Figures of merit of EC/OC analysis

Parameter	RSD (%) (n=3)	DL (μg/cm²) (n=7)	Sunset Lab. PES (μg/cm²) (n=5)		This study PES (μg/cm²) (n=5)	
			Avg	SD	Avg	SD
OC	4.60	0.31	16.75	0.94	13.37	1.33
EC	5.02	0.06	2.05	0.20	1.88	0.34
TC	4.65	0.34	18.80	1.14	15.26	1.67

MATERIALS AND METHODS

Sample Collection

24-hr PM_{10} samples were collected in this study during 50 m-range 22-cal rifle and pistol competitions, organized between 12 and 15 May 2016 in Erdemli (Mersin, Turkey) by the Turkish Shooting and Hunting Federation to investigate the air quality from firearms. Figure 1 depicts the schematic of the shooting range and the place where the Tecora Skypost PM_{10} sampler, which is working by the EN 12341:2014 norm, was located. According to the statutes, rules, and regulations of the International Shooting Sport Federation (ISSF), athletes cannot go beyond the 5 m distance at the shooting line, where only the referees can cross this border. Entry and exit of the athletes to the shooting area can only be performed under the supervision of referees, and all the supporters and coaches should obey this rule of ISSF during match and training times. In addition, even if referees should keep quiet during all shooting competitions except for the final match. Otherwise, supporters are invited to out of the range by the referees. Since a high-volume sampler produces an appreciable degree of noise during sampling, it was placed 10 m behind and 5 m above the fire area during the matches. Three match samples and one blank sample were collected on the pre-fired Whatman quartz fiber filter during the shooting activities. Quartz filters were pre-conditioned at 25°C and 25% relative humidity for one day before sampling, weighted and stored at -18°C until sampling. The sampler was operated only for 5 min at the sampling flow rate of 34 L/sec for the blank sample. All the samples were kept in the Petri slides in the freezer till analysis. Before analysis, the samples were pre-conditioned for one day under the same conditions stated previously, and PM load was determined by subtracting the tare of the filter. Rifle matches took place on the first and second day of the competitions, while pistol matches were last.

EC/OC Analysis

A 1.5 cm² punch was cut from the collected filters and analyzed utilizing Sunset Lab. (Oregon, USA) thermal-optical transmission EC/OC analyzer for elemental, organic, and total carbon (EC, OC, and TC=EC + OC). National Institute of Occupational Safety and Health (NIOSH) 870 protocol was followed during the analysis [66]. The details of the EC/OC analysis were provided in Öztürk and Keleş [67]. Briefly, the analyzer uses thermal, optical, and chemical principles to determine the carbon content of the filter samples. Firstly, an inert atmosphere is created by purging 100% He inside the oven, and OC formed during this step is converted to CO_2. Then, EC is transformed to CO_2 under oxidizing medium by purging a mixture of gas composed of 10% O_2 and 90% He (vol/vol). Afterward, generated CO_2 is reduced to CH_4, which is detected by a flame ionization detector (FID). The performance of FID is checked at the end of each run by injecting a fixed volume of methane (5% CH_4 plus 95% He, vol/vol) as an internal standard. As a part of the quality assurance and quality control (QA/QC) protocol, a known amount of sucrose solution was spiked over the pre-fired blank filters. Analysis was conducted under the same conditions as the filter samples. The measured sucrose concentrations have deviated only 0.10% from the standard sucrose solution based on n=14 repeated measurements. In addition, the instrument was operated without putting any sample at the beginning of each analysis day. The average OC levels determined in the instrument blanks were less than 0.02 μg/cm² while no EC was detected.

The detection limit (DL) and precision as relative standard deviation (RSD) of EC/OC measurements were also calculated in this study. DL values (three times the standard deviation of the blank filter measurements) were calculated based on the repeated blank measurements, and RSD values were estimated by analyzing the performance evaluation standard (PES) provided by the Sunset Lab. Moreover, the accuracy of the EC/OC measurements was calculated based on the data provided by Sunset Lab. for PES measurements. Table 1 summarizes figures of merit of EC/OC analysis.

As given in Table 1, the percent RSD values were almost ≤5, indicating that repeatability of the analysis is acceptable. DL values of measurements were 0.31, 0.06 and 0.34 μg/cm² for OC, EC and TC, respectively. The accuracy of the measurements was found to be 20, 8, and 18%, respectively (Table 1). The EC/OC results provided in this paper were corrected for filter blank. Since no carbonate carbon (CC) peak was detected during analysis, CC correction was not performed.

Table 2. Figures of merit of EC/OC analysis

	Step			
	I	II	III	IV
Ramp time (min)	3	2	2	1
Hold time (min)	5	10	30	10
Temperature (°C)	140	160	200	50
Pressure (bar)	30	30	35	25
Power (%)	75	85	90	0

Elemental Analysis

The rest of the filters from EC/OC analysis were digested in a mix of high purity acids (5 mL HNO_3 + 1 mL HF + 0.5 mL H_2O_2 + 1 mL de-ionized (DI) water using Berghof (speed wave-2, Germany) Microwave Digestion Oven. The steps of the digestion program are tabulated in Table 2. Acid and field filter blank were also digested along with the filter samples and treated similarly. After digestion, all the samples and blanks were diluted to 50 mL with DI water, transferred to HDPE bottles, and kept in the refrigerator till analysis. After micro-wave digestion only, samples that showed visible residues of soot carbon were filtered through 0.45 µm pore size Millipore brand mixed esters of cellulose filter (Sartorius AG).

Agilent 7700 Model Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (California, USA) was employed in this study to perform the trace element analysis of the samples. Samples were analyzed for 15 elements using ICP-MS (Be, Cd, Co, Cr, Cu, Hg, Mn, Ni, As, Ba, Pb, Se, Sr, V, and Zn). 100-ppb germanium and terbium were used as internal standards during the analysis of samples. Agilent internal standard mix for ICP-MS systems (part number 5188-6525) calibrated the instrument. Calibration standards, blanks, and samples were spiked with this internal standard to overcome instrumental and sample-related variations. Internal standard element recovery was monitored closely by following the procedure described in EPA Method 200.8, section 9.4.3 [68]. Recoveries tried to be kept between 70% and 120%. In addition, the method detection limit (MDL) of the analysis was also calculated based on the same method as described in section 9.2.4 [68]. Moreover, instrument detection limit (IDL) values were estimated following the procedure provided in the same method under section 9.2.5. The accuracy of the measurements was also monitored closely as a part of the quality assurance and quality control (QA/QC) procedure. To this end, Standard Reference Material (SRM) 2783 obtained from the National Institute of Standard and Technologies (NIST) was digested by following the procedure used to extract filter samples. The obtained results were compared with the certificate sheet of the SRM. The precision of the measurements was also estimated based on the relative standard deviation of repeated SRM analysis. The values corresponding to SRM measurements, precision, MDL, IDL, and recovery for the measured parameters are summarized in Table 3 below.

Except for the Cd, the recoveries were >70% for all of the elements. Thus, the measured concentrations were not cor-

Table 3. Figures of merit of ICPMS measurements

	SRM 2783 Certified values (mg/kg)		Obtained values (mg/kg)		Precision (%)	IDL (µg/kg)	MDL (µg/kg)	Recovery (%)	
Parameter	Avg	SD	Avg	SD				Avg	SD
As	7.0	1.6	5.8	0.3	17.1	4.2	112	79.4	2.82
Ba									
Be									
Cd	7.3	3.7	7.5	0.5	3.1	2.5	7.5	57.8	2.27
Co						6.1	163.2	75.6	2.5
Cr	80	22	54.5	4.1	24.7	32.2	125.6	79.6	5.02
Cu						20	122.5	73.3	2.24
Hg						7.5	22.6	74.9	6.05
Mn						89.8	172.2	72.8	2.51
Ni						135.5	406.5	77.2	3.46
Pb	85.9	7.2	64.6	11.7	31.9	20.8	33.3	87.0	6.83
Se									
Sr									
V						34.9	122.8	78.3	2.22
Zn						90.6	941.4	78.1	5.19

Table 4. The average concentration of parameters measured in PM10 samples during the competitions

Parameter	Unit	1st-day rifle	2nd-day rifle	3rd-day pistol	Avg	SD
PM_{10}	μg/m³	25.4	21.9	38.8	28.7	7.3
OC	μg/m³	4.02	3.72	6.47	4.74	1.23
EC	μg/m³	0.197	0.194	0.349	0.247	0.072
TC	μg/m³	4.24	3.92	6.82	4.99	1.30
As	ng/m³	0.334	0.432	0.251	0.339	0.074
Ba	ng/m³	21.9	13.8	19	18.2	3.4
Be	ng/m³	0.026	0.035	0.032	0.031	0.004
Cd	ng/m³	0.28	0.117	0.495	0.297	0.155
Cr	ng/m³	1.06	3.39	2.13	2.19	0.95
Cu	ng/m³	3.18	1.51	1.27	1.99	0.85
Mn	ng/m³	1.81	2.55	2.24	2.20	0.30
Ni	ng/m³	2.18	3.03	2.27	2.49	0.38
Pb	ng/m³	221	110	259	197	63
Se	ng/m³	1.34	2.81	1.6	1.92	0.64
Sr	ng/m³	0.266	1.349	0.548	0.721	0.459
V	ng/m³	6.35	6.7	5.86	6.30	0.34
Zn	ng/m³	4.72	4.55	1.15	3.47	1.64

rected with the recovery values. If IDL values <0 for the parameter of concern, the corresponding value was replaced with one-third of the associated MDL value. Moreover, if the measured concentration of the parameter is less than both IDL and MDL, the analyte concentration was replaced with half its corresponding MDL value to use the data in the statistical analysis. Furthermore, the metal levels reported in this study were field blank corrected.

RESULTS AND DISCUSSION

The concentration of parameters measured during 3-day competitions is summarized in Table 4, including the average (Avg) and standard deviation (SD) of measured concentrations for the whole field study. On the third day, when the pistol athletes were competing, almost two times higher PM_{10} mass concentration was measured compared to the previous two days. The PM_{10} mass concentrations for three days were measured as 25.4, 21.9, and 38.8 μg/m³, respectively. The highest concentration was measured for OC (4.02 μg/m³), while the minimum concentration was obtained for Be (0.026 ng/m³) on the first day when rifle matches took place. The maximum and minimum concentrations were obtained for the same parameters on the second and third days of the competitions, as tabulated in Table 4.

It is also worthy to note that the highest PM_{10}, OC, EC, TC, Cd, and Pb levels were measured in the samples while the pistol was being used for shooting in the third-day competitions. Grabinski et al. [48] revealed that PM mass emissions when the shooters used rifle is about an order of magnitude lower than the emissions released from pistols. This situation can be attributed to the greater barrel diameter and shorter barrel length used in pistols.

During the matches, the measured PM_{10} mass concentration was found as 25.4, 21.9, and 38.8 μg/m³, respectively, for the first, second, and third day, while the average of the whole event was 28.7±7.3 μg/m³. The measured values were below the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) and England indoor PM_{10} limit, 75 and 50 μg/m³, respectively. Hong Kong's first level value for indoor PM_{10} is 20 μg/m³ [69]. Table 4 revealed that during the matches, this limit value was exceeded. Wingfors et al. [38] reported that most of the particles released from indoor firing ranges fall to nanoparticle size regime, which is more critical in terms of human health point of view since these particles are capable of penetrating deep into the human respiratory tract by inhalation [70]. Consequently, the mass concentration and the size distribution of PM are significant for indoor shooters.

The measured PM_{10} concentrations in this study were comparable with the ones reported by Orru et al. [71]. The average PM concentration in one of the shooting ranges in which pistols were used is 28.9 μg/m³. Researchers collected size-segregated PM samples at indoor military shooting ranges and analyzed the collected samples in terms of metals, including Pb, Cu, Ni, and Zn. In contrast to PM_{10} levels, significantly higher metal concentrations were reported in this military shooting range.

Grabinski et al. [48] reported Cu and Zn concentration in a ventilated, indoor firing range, where PM samples were collected in cases both pistol and rifle used for firing. The airborne mass concentration of Cu was reported to vary from ≤1 to 16 µg/m^3, which is considerably higher than one measured in this study (average of the competition 1.99 ng/m^3). In addition, researchers found that Zn levels ranged from 0.5 to 1.8 µg/m^3, which is again much higher than the competition average value (Table 4).

Indoor EC and OC levels were published in some studies in the literature. For example, Na and Cocker [72] determined concentrations of these two parameters inside the 20 residential settings and one local high school in CA, USA. In the homes with smokers, OC levels ranged from 9.3±1.6 to 229±67.6 µg/m^3 while EC concentrations were between 1.7±0.7 and 4.0±1.5 µg/m^3 in $PM_{2.5}$ samples. Authors claimed that there are no indoor EC sources while OC considerably contributed to indoor $PM_{2.5}$ levels. In a similar study, Ho et al. [73] collected $PM_{2.5}$ samples inside the five buildings near roadsides in Hong Kong. The average inside OC and EC levels were found as 11.3±5.5 and 4.8±3.4 µg/m^3. The significant sources determining the indoor concentrations of these pollutants were attributed to the penetration of outdoor pollution. Seleventi et al. [74] performed $PM_{2.5}$ sampling inside an apartment in Athens. The researchers reported the indoor average OC and EC levels as 9.6 and 1.9 µg/m^3, respectively. The indoor source of OC was thought to be several activities performed by the residents, such as smoking, cooking, and cleaning. However, all these studies were conducted inside the residential places, and reported values cannot be compared to those measured in a shooting range. To our best knowledge, there is only one study in the literature reporting black carbon (BC) concentration for indoor air during sportive activities. Bisht et al. [75] monitored the indoor air quality for stadiums during the 19th Common Wealth Games (CWG) at Delhi (India). It is good to mention that BC determination relies on optical methods, and thermal-optical methods determine EC. Although co-located measurements showed that BC data could be 20% higher than EC levels [76], EC can be used as a surrogate measure of BC [77] In Bisht et al. [75], BC levels inside the three sports venues during CWG were reported to range about from 12 to 14 µg/m^3. Unfortunately, there is no information about the nature of sports competitions provided in the study. Researchers revealed that BC showed a positive correlation with CO, a pollutant released from incomplete combustion. Once the EC data generated in the shooting range were compared with the BC levels reported by Bisht et al. [75], it can be concluded that BC data was at least 50 times higher than EC measured in the shooting range.

Among the elements analyzed in the PM_{10} samples collected during the competitions and listed in Table 4, Cr is paying attention. Cr presents in the ambient atmosphere as Cr (III) and Cr (VI). Cr (III) is essential in trace levels for the proper functioning of living organisms. On the other hand, Cr (VI) is known as a pulmonary carcinogen by the International Agency for Research on Cancer and US Toxicology Program [78]. Indoor exposure to Cr (VI) is related to elevated lung and nasal cancer risk [79]. US Environmental Protection Agency listed Cr (VI) compounds as one of the 18 core Hazardous Air Pollutants (HAPs) [80]. It has been estimated that one in a million-cancer risk threshold for Cr (VI) is 0.083 ng/m^3. The average Cr (VI) to total Cr ratios ranged from 1 to 30% for the ambient air [81]. By taking a ratio of 15% on average, Cr (VI) values for this study can be estimated to vary from 0.159 to 0.509 ng/m^3 for the first and second day, respectively. Consequently, the emitted Cr during the competitions has cancer risk potential.

Once Table 4 is evaluated in terms of elements, it can be seen that the highest level was measured for Pb compared to other metals. The average Pb concentration for the whole study was 197±63 ng/m^3. Several guidelines regulate occupational Pb exposure in the world. For example, OSHA (Occupational Safety and Health Administration) (USA) set 30 µg of Pb per cubic meter of air as an action level for indoor environments. In addition, the time-weighted average (exposure over an eight-hour average) for Pb was set to 50 µg/m^3 in the same regulation. Furthermore, NIOSH recommended exposure limit (8-hour average) for Pb is 50 µg/m^3 while increasing Pb exposure to 100 µg/m^3 indicates the level that is Immediately Dangerous to Life and Health (IDLH) [82]. When these limit values were compared with the data generated in our study, none of the days' Pb exposure limits were exceeded. However, it should be kept in mind that the samples collected in this study for 24-hr long. Consequently, the measured Pb levels that we reported in this study were smoothed out.

The shooters are exposed to Pb from three different sources during the shooting activity. The first source is ammunition primer, composed of lead styphnate, which initiates the mercury fulminate explosion and lead azide propellant, released to the ambient air upon firing. The second one is burning propellant in the cartridge, which vaporizes the Pb due to extreme temperatures as high as 1100 °F. The last is associated with the dust and lead oxide fumes, which are emitted when the bullet hits to target [83]. Once the Pb is released to the indoor environment, the occupants inside the firing range are exposed to this metal through dermal contact [84], ingestion [85], and inhalation [84]. The adverse health impacts of elevated Pb exposure are well documented in the literature [36]. International Agency for Research on Cancer (IARC) declared inorganic lead as a probable human carcinogen (group 2A) [86]. Conversely, limited evidence has been found in human studies. In contrast, there is adequate data on the carcinogenicity of Pb in experimental animal studies [56]. Gulson et al. [87] suggested using non-lead primers to reduce the uptake of lead by recreational shooters.

Emission factors (EF) for the measured parameters were also calculated by dividing the mass concentrations of the parameters by the number of shots each day. Table 5 below summarizes the EF values for the corresponding parameters for rifle (average of first two days) and pistol (third day).

Once the normalized values were compared for rifle and pistol, it was found that Cd mass emission per bullet from pistol shooting is about four times higher than the one measured for the rifle. Similarly, about three times higher EF values were calculated for PM_{10}, TC, OC, EC, and Pb when pistol matches took place instead of the rifle. Estimated EF values for Mn, Cr, Ni, V, and Ba are about 1.5 times higher for pistol emissions than for the rifle. Zinc was the only pollutant that had a higher EF value in rifle emissions. EF values calculated for Sr and Cu are comparable.

In addition to the parameters listed herein, ammunition during shooting activities may release other stressors to the indoor environment. For example, nitrogen (NO_x) oxides are among these pollutants and irritate the eyes and respiratory system. Carbon monoxide (CO), another pollutant released into the indoor atmosphere due to firing, is known to reduce the ability of blood to carry oxygen and leads to headaches and nausea. Moreover, polycyclic aromatic hydrocarbons (PAHs), hydrogen cyanide (HCN), ammonia (NH_3), sulfur dioxide (SO_2), and hydrogen chloride (HCl) are other air pollutants released to indoor air upon firing. The literature has well reported the detrimental health impacts associated with these pollutants on humans and the environment.

Limitations

Since the competition was officially three days, PM_{10} samples, including two in rifle and one in pistol, were collected during this study, which prevents us from making a comprehensive assessment about the differences or similarities in the chemical compositions of the collected samples. In addition, 24-hr PM_{10} samples were collected during the study through the matches were performed from 09.00 am to 08.00 pm. Shooting range opening and closing times are spread over a wider range. After the scheduled matches, shooters may perform small training shots in order to adjust guns, shooting position, and other equipment revealed to prolonged shooting hours. In regular training conditions, samples may have been collected in shorter time windows. However, it should be noted that this is a well-attended national organization. Also, there is no other example in the literature on a measurement related to sports-shooting competition cases. Additionally, the ratio of Cr (VI) to Cr was used in this study based on the ambient PM data, and no value was found for the indoor environments. Consequently, one should consider these limitations while interpreting the generated data in this study.

Table 5. Calculated emission factor (EF) values for PM_{10}, carbonaceous materials, and elements for rifle and pistol shots (PM_{10}, OC, EC & TC in ng/bullet and elements in pg/bullet)

Parameter	Rifle	Pistol
PM_{10}	116	321
TC	20	56
OC	19	54
EC	0.959	2.890
Be	0.149	0.267
Cd	0.972	4.087
As	1.88	2.07
Sr	3.96	4.52
Se	10	13
Mn	11	18
Cr	11	18
Cu	11	11
Ni	13	19
Zn	23	10
V	32	48
Ba	87	157
Pb	810	2134

CONCLUSION

This study was conducted during a three-day-long official national shooting competition. To our best knowledge, we report first-time EC, OC, and TC concentrations associated with the indoor firing ranges. Another critical point is that the Mersin/Erdemli shooting range is neither commercial, recreational, nor military. So, occupants are licensed shooting athletes, their families, official coaches, referees, and audiences. It was demonstrated here that shooting activities produce a considerable amount of particulate matter, carbonaceous material, and many toxic elements associated with it. Lead was the most dominant metal component of a PM that we measured during the competitions regardless of pistol or rifle used. However, its concentration did not exceed the permissible levels for indoor firing ranges.

Another crucial point is that lead-free bullets have reduced precision and accuracy at the shot point on the target. On the other hand, in terms of athlete and environmental health, green or "lead-free" bullets should be encouraged to use during indoor shooting to reduce the risk of Pb exposure. Among the elements analyzed, Cr was one of the elements that have cancer risk potential. Additionally, there are some technical differences between a .22-cal rifle and pistol. The pistol bore length is shorter, and the bore radius is larger than the rifle. Also, the rifle has a higher bullet core release speed than the pistol can cause more pollutant accumulation close to the shooter in pistol matches. It should also be considered that pollutants can accumulate in the environment for three days and repeatedly be resuspended from the ground. Ventilation is another critical point that should not be negligible. It should be kept out of the operational cost savings. Indoor air quality in terms of PM and all these pollutants should be closely monitored at indoor firing ranges to take more proactive actions against these pollutants. Furthermore, the operator should regularly check the ventilation system of indoor air firing ranges to ensure acceptable air quality. Personal sampling provides a more accurate evaluation of human exposure during shooting activities. Consequently, personal sampling and indoor air quality monitoring should be coupled to understand better the impact of indoor air firing ranges on the occupants.

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DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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