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Shooting Activities as the Potential Influence on the Environment at the Pokljuka Biathlon Center, Slovenia

Valentina Pezdir * and Mateja Gosar

Geological Survey of Slovenia, Dimičeva ulica 14, 1000 Ljubljana, Slovenia; mateja.gosar@geo-zs.si
* Correspondence: valentina.pezdir@geo-zs.si

Abstract

Biathlon activities can have negative effects on the environment due to the processes occurring during shooting, as the biathlon ammunition contains significant amounts of Pb, Cu and Sb. To determine these effects, we looked into the presence of lead-bearing particles in the vicinity of a shooting range in Rudno polje, Pokljuka, in Slovenia. Particles were collected from snow and later analysed using SEM/EDS, where we focused on the lead-bearing particles. These particles are composed of mainly lead or lead oxides/hydroxides with trace presence of Cu and Sb, both of which are commonly related to ammunition used in biathlon shooting and other shooting activities. To confirm that the particles originated from shooting, we compared them with particles taken from an indoor shooting range as dust and residue in ammunition casings. Lead-bearing particles show morphological changes that are caused by high temperatures and pressures during the shooting process.

Keywords: atmospheric particles; ammunition residue; SEM/EDS

1. Introduction

The biathlon is a common winter sport, combining cross country skiing and shooting. In shooting ranges, certain chemical elements, especially Pb and Sb, can have a significant presence due to the composition of the ammunition used [1]. Most biathlon rifles use lead bullets in a copper casing with traces of antimony commonly present [2,3].

As shooting can lead to elevated levels of certain elements, which can present a threat to the environment and health, many studies have focused on evaluating the influence of shooting ranges, such as the extent of element toxicity and mobility [4–7] and the environmental impact [8–10]. Many environmental impact studies that have investigated the presence of elements originating from military or other practices involving shooting have focused mainly on soils [11–13].

The main three elements present in ammunition (Pb, Sb and Cu) can be considered toxic for the environment. Lead (Pb) is a potentially toxic element, which, even in small quantities, can negatively influence the neurological development and functioning of people and other vertebrates [14–18]. In relation to shooting, lead can be transported as dust particles, which can represent health risks due to inhalation [19,20]. Lead can also present a threat to plants, as it can reduce soil enzyme activity and reduce the ability to perform photosynthesis [21–24].

The median level of Pb in Slovenian soil determined after aqua regia digestion is 34 mg/kg [25], which is higher than in European grazing land soil (18 mg/kg; GEMAS project [26]). Levels range quite significantly from 6.2 mg/kg to 850 mg/kg. The spatial distribution of Pb levels in Slovenian soil is influenced by anthropogenic factors. The spatial

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distribution shows high levels of lead and zinc in the wider area of Mežica where the most important anthropogenic factors are past mining, ore processing and smelting. It has been well described that Mežica and the surrounding area are extremely polluted with Pb, Zn, Cd and Mo [27–31]. Also, in the Litija area along the Sava River east of Ljubljana, there are elevated Pb and other PHE levels in the environment that have been ascribed to mining and smelting activities [32,33]. Typical for urban soils, the presence of Pb and other PHE levels are elevated in urban areas [34–37].

An increased presence of Cu can impact the neurological system [38], while in plants its impacts can be seen as decreased plant growth and the degradation of chlorophyll [39]. The median level of Cu in Slovenian soil determined after aqua regia digestion is 20 mg/kg [25], which is similar to the median Cu level in European soil (15 mg/kg; GEMAS project [26]). The values range between 1.4 mg/kg and 300 mg/kg. Beside geogenic factors that mainly determine the distribution of Cu in Slovenian soils, anthropogenic factors also contribute to Cu distributions in soils in urban and industrial/mining areas [25].

Although Sb in projectiles is only present in small quantities, the element is known as toxic and potentially carcinogenic to the human body [40,41]. Antimony toxicity commonly manifests as pulmonary issues and genotoxicity [42]. Increased antimony content also causes stress to plants, which can manifest as stunted growth, reduction in the uptake of essential nutrients, reduced photosynthesis, chlorosis, necrosis, and lipid peroxidation [43,44]. The median level of Sb in Slovenian soil determined after aqua regia digestion is 0.53 mg/kg [25], which is higher than the median Sb level of European soil and also of Southern European soil (0.23 mg/kg and 0.35 mg/kg, respectively; GEMAS project [26]). The values in Slovenia range between 0.06 mg/kg and 8.9 mg/kg.

This study was conducted at Rudno polje on the Pokljuka plateau in the area of a biathlon shooting range (Figure 1). The Pokljuka is a karst plateau in NE Slovenia with an elevation of approximately 1200 to 1500 m. It has a relatively flat topography and borders on valleys in the S, SW and E of the area and on a high mountain range in the NE. The Pokljuka plateau area is part of Triglav National Park. The area has experienced increased biathlon activity in winter with the addition of larger scale biathlon competitions. The biathlon shooting range has been active since at least 1992 when the first biathlon world championship in Pokljuka was held.

In Gosar et al. [25], we determined the threshold values for the elements in the soil. For the area of the Western Alps, where Pokljuka is located, the values for lead, antimony and copper based on MD2MAD (median + 2× median absolute deviation; logarithmic values) are 140 mg/kg, 1.8 mg/kg and 56 mg/kg, respectively. The median values for Pb, Sb and Cu are 42 mg/kg, 0.59 mg/kg and 19 mg/kg, respectively.

In our study, we focused on the presence of lead-bearing particles deposited in snow, presumably originating from biathlon activities. Therefore, the aim of the study was to detect the presence of free-falling residue particles, as they are released during the shooting of rifles, and to evaluate their presence at the Rudno polje shooting range. As Pokljuka is part of a protected Triglav National Park, the presence of lead particles can present an important potential environment risk.

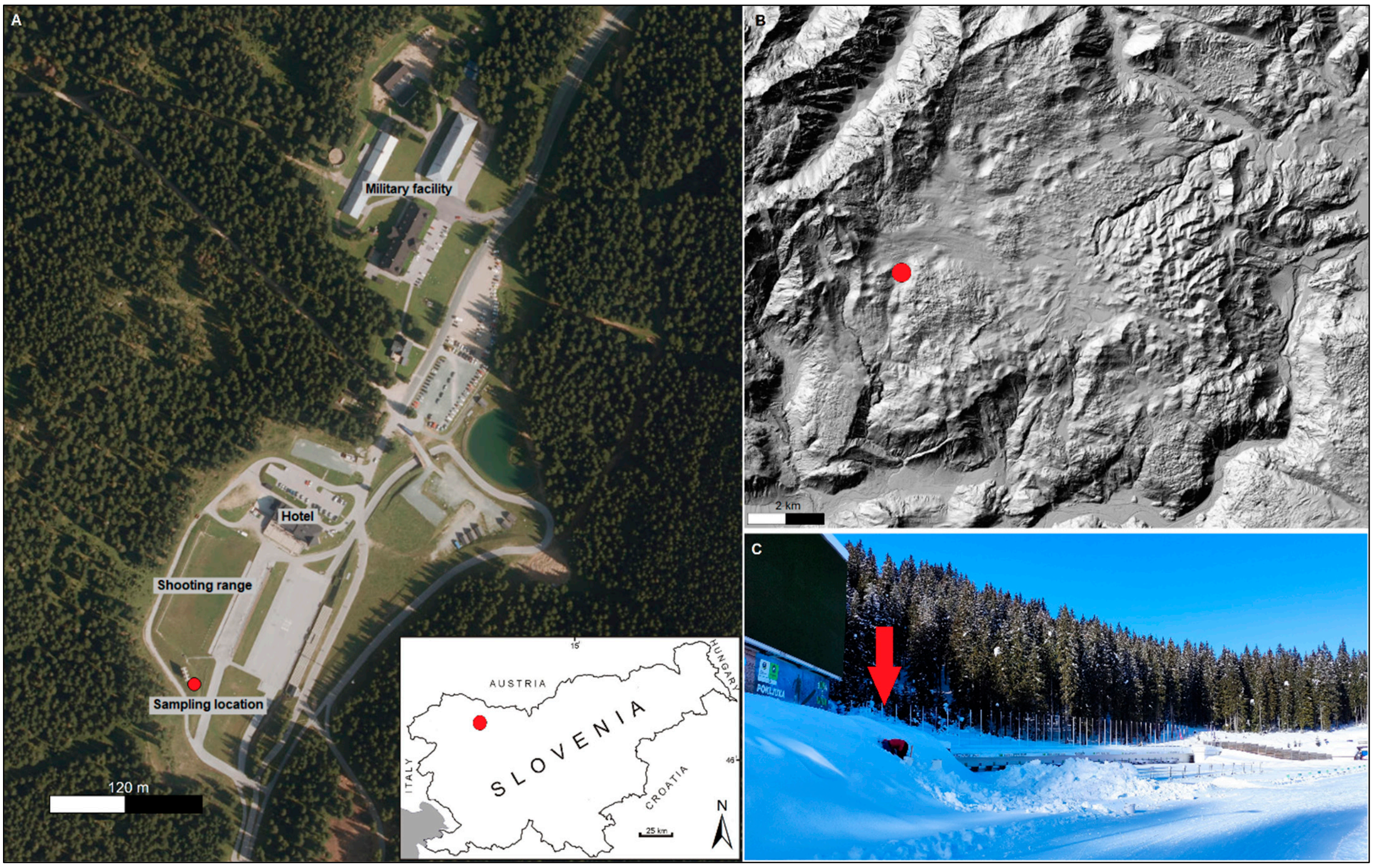

Figure 1. Location of the snow sampling. (**A**) The wider area surrounding the sampling location, with the red dot showing the sampling location next to the shooting range. (**B**) The study area of the Pokljuka plateau area with the red dot marking the location of Rudno polje (LiDAR ARSO, http://gis.arso.gov.si/evode/profile.aspx?id=atlas_voda_Lidar@Arso; accessed on 12 November 2024 [45]). (**C**) The conditions at the time of sampling in January 2023, with the red arrow marking the sampling location.

2. Materials and Methods

In 2021 and 2023, we sampled snow south of a biathlon shooting range on Rudno polje on the Pokljuka plateau in Northwestern Slovenia (Figure 1). In February 2021, we took one snow sample (M-1) from a 50 × 50 cm area to a depth of 5 cm. In January 2023, we took three snow samples from the same location 20 days after the Biathlon World Cup competition in Pokljuka. There, we sampled the older lower compact layer of snow (M-2) deposited prior to the biathlon competition, snow on the boundary between the fresh upper layer and the lower compact layer (M-3) and the whole snow profile (M-4). The latter two both contain records of the Biathlon World Cup event.

In February 2021, the average temperature was −3 °C and the total precipitation (snowfall) was 135 mm. In the 18 days prior to the first sampling, the predominant wind directions with the strongest winds were north (average speed 2.1 m/s with wind gusts of 12.3 m/s) and NNW (average speed 1.6 m/s with wind gusts of 16.7 m/s). There were also multiple events of relatively strong winds from the south and weak winds from the west.

In between the Biathlon World Cup competition and the time of sampling in January 2023, there was 149.4 mm of precipitation (snowfall) in total. As the biathlon activities on the Pokljuka plateau are ongoing the whole winter, we also sampled the fresh layer of the snow. The temperature prior to sampling ranged from −8 to 7 °C with an average temperature of −1.5 °C. The predominant wind direction was north with an average speed of 2.5 m/s. Stronger winds were observed in N and S directions (15.4 m/s and 11.1 m/s, respectively).

After the sampling, the snow was melted, and snowmelt water was filtered using isopore membrane polycarbonate filters (<0.6 μm) (Merck KGaA, Darmstadt, Germany) to obtain particulate matter. Prior to the filtering, physico-chemical characteristics of meltwater (pH and electrical conductivity) were determined. One sample was additionally analysed for its chemical composition using ICP-MS and compared to other precipitation samples taken in the years 2020–2021 from the Pokljuka plateau [46]. After the filters dried, the particulate matter was applied to carbon tape with a 0.25 cm^2 area, which was mounted on an aluminium stub. The sample was coated with carbon to increase the electrical conductivity and analysed using scanning electron microscopy coupled with energy dispersive spectroscopy (SEM/EDS) using JEOL JSM 6490LV SEM (JEOL Ltd., Tokyo, Japan) coupled with an Oxford INCA PentaFETx3 Si(Li) detector (Oxford Instruments Analytical, Ltd., High Wycombe, UK) and INCA Energy 350 processing software (Oxford Instruments Analytical, Ltd., High Wycombe, UK) installed at the Geological Survey of Slovenia. The SEM/EDS analysis was performed in a high vacuum at an accelerating voltage of 20 kV, a spot size 48–50 and a working distance of 10 mm.

For the particle analysis, we randomly determined several areas (fields of view) on the carbon tape of each sample and analysed particles within a selected area. To determine the ratios of specific mineral groups in the individual sample, we used surface EDS analysis (mapping) and visually compared the selected fields of view with the Charts for Estimating Percentage Composition of Rocks and Sediments [47]. Magnification, used for EDS elemental mapping, is ×300.

Additionally, for comparison with the particles from the snow, we obtained samples of used 22LR bullets and air rifle pellets from a shooting range in Postojna in SW Slovenia. In the case of the 22LR bullets, we collected residue within the casings of used bullets, while for air rifle pellets we collected deposited residue next to shooting targets (used pellets with dust). We applied small dust particles (<1 mm) from both ammunition samples on the carbon tape on the aluminium stub and coated it with carbon for better conductivity. Due to the nature of this collection, other dust particles were also present in the sample; however, we focused only on lead particles. We observed the ammunition residue samples using SEM/EDS.

3. Results and Discussion

3.1. Physico-Chemical Parameters and Chemical Compositions of Snowmelt Water

The measured physico-chemical parameters in the snowmelt water show that they were slightly acidic with very low electrical conductivity (Table 1). The lowest EC and pH values were measured in sample M-3, which represents the boundary between the fresh snow and the old compacted snow. Additionally, the higher values in other samples might be due to the samples being taken from more compacted layers that were exposed to the atmosphere and other influences for a longer period of time.

Table 1. Physico-chemical parameters (pH and EC) of snowmelt water.

Code	Date and Location	pH	EC (μS/cm)	Volume (l)
M-1	24th February 2021 (snow profile)	5.6	3.2	3.7
M-2	25th January 2023 (compact layer)	5.7	3.1	3.6
M-3	25th January 2023 (boundary)	5.4	2.0	4.3
M-4	25th January 2023 (snow profile)	5.7	3.0	3.6

The chemical composition of meltwater sample M-1 (Figure 2) was determined and compared with the composition of the precipitation in the vicinity of the sampling area [48]. Generally, the element concentrations in precipitation are low, as described in Pezdir and coauthors [48]. In focusing on the elements of interest (Cu, Pb and Sb), we observed that these elements had similar concentrations between all precipitation samples. In the snowmelt sample taken from the shooting range in 2021, the concentrations of Cu and Pb were similar or lower compared to the precipitation in the vicinity; however, the concentration of Sb was much higher (0.94 µg/L) in the snowmelt water from the shooting range. This could be attributed to the dissolution of Sb-bearing particles in the slightly acidic meltwater and the increased mobility of Sb in an oxidized environment [49].

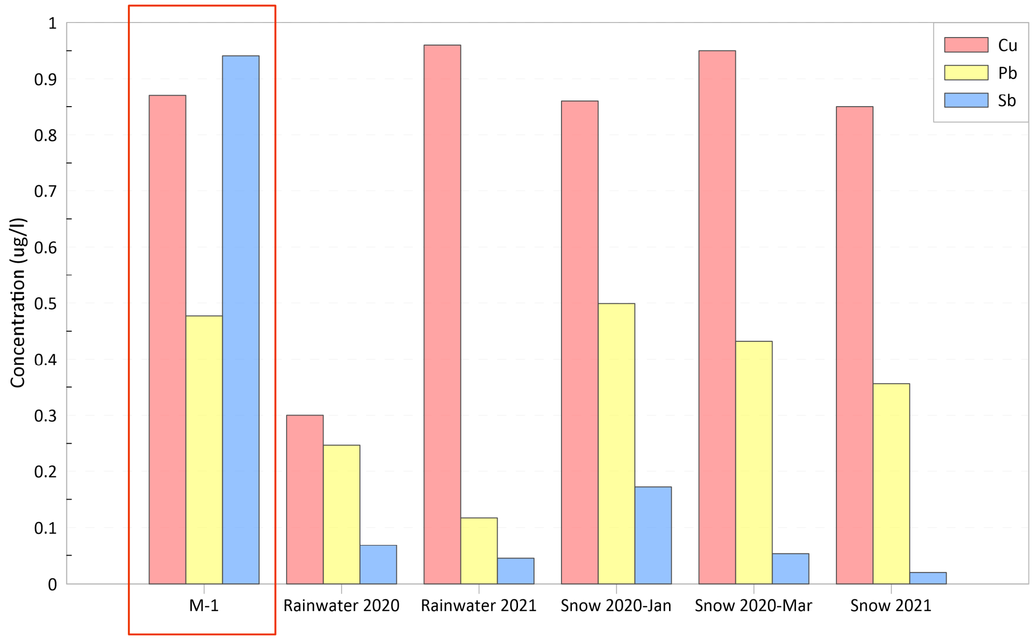

Figure 2. Median Cu, Pb and Sb concentrations in the precipitation samples [48] and comparison to the snowmelt sample from the shooting range, M-1 (red rectangle).

3.2. Characterization of Lead-Bearing Particles in the Snow

The general composition of particles in the snow samples (Table 2) was obtained with EDS elemental mapping at 300× magnification. We identified six main groups of particles. The two main groups are quartz with other silica-containing minerals and aluminosilicates, which frequently include lower concentrations of Fe and Mg. These elements are common in the pyroxene and amphibole mineral groups and could have local origins from the flysch N and SW from Rudno polje [50]. The particles have various sizes ranging from 0.5 µm to 30 µm. They have predominantly angular shapes. The next noticeable groups are represented by carbonates and organic particles. Carbonates are mainly found as smaller particles (<10 µm) with angular shapes. They also appear as crystal clusters, which indicates that they most likely crystallized from the residual snowmelt water during the drying of the filter papers. Organic particles are larger in size (>10 µm) and can be found as rounded, spherical particles or strings and coatings. Approximately 8–10% of the particles in the sample were iron oxyhydroxides. They are found in angular shapes, which can have geogenic origins, as iron-bearing minerals are naturally present on the Pokljuka plateau [51,52]. They are also present in other shapes (irregular and spherical shapes; agglomerates and shavings), which can be typical indicators of anthropogenic sources [36,37]. The last group is represented by lead oxides, which have slightly rounded, angular and irregular shapes. Their average size was approximately 13 µm. These described groups, with the exception of lead-bearing particles, were also observed in snow and rainwater from Šijec bog, which is located approximately 5 km from

Rudno polje [46]. The short distance between the locations and the lack of lead-bearing particles in Šijec bog indicate the shooting range as the potential source of the lead.

Table 2. Estimated percentage content of the main particle groups in the snowmelt samples. The content of other particles was less than 5%, but that content varied between samples and thus was not included in these calculations.

Code	Quartz and Silica Minerals (%)	Aluminosilicates (%)	Carbonates (%)	Organic Particles (%)	Iron Oxyhydroxides (%)	Lead Oxides (%)
M-1	30	25	17	13	9	6
M-2	35	25	12	14	8	6
M-3	35	24	14	12	8	7
M-4	28	23	20	12	10	7

The snow sample in February 2021 (M-1) was composed of approximately 6% lead-bearing particles (Table 2). Small amounts of other elements such as Sb, Cu, Al, Si and Ca were also detected in the particles. Particles ranged in size, from 1 µm to 30 µm, with an average particle size of 13.9 µm. Particles have various shapes with predominantly angular shapes (Figure 3a), rounded shapes (Figure 3c,d) and spheres (Figure 3b). Smaller particles can be found in clusters located on the surface of larger particles, presumed to be present due to the sample preparation process, and can together with coatings on the particles (Figure 3d) influence the detected elemental composition. Most particles either have a smooth surface, form agglomerates or are irregular in shape, which could be related to changes during the shooting process.

The presence of lead-bearing particles in the snow from January 2023 was 6%, 7% and 7% for samples M-2 (compact snow layer), M-3 (boundary) and M-4 (snow profile), respectively (Table 2). In the sample M-2, the lead-bearing particles contained smaller amounts of Cu, Sb, Al, Si and Ti. Their sizes varied from 3 µm to 29 µm with an average size of 9.9 µm. Particles can be seen in various shapes, having angular, rounded, irregular and spherical shapes (Figure 4a–c). The lead-bearing particles in sample M-3 were mainly angular in shape with an average size of 11.3 µm. Most lead-bearing particles also contained small concentrations of Sb and Cu. The sample M-4, which covered the whole snow profile, included angular and elongated lead-bearing particles (Figure 4d–f), which were generally larger compared to other samples (an average size of 15.7 µm) as individual particles often measured more than 30 µm. Most of these particles also had a small amount of Sb.

In comparing the lead-bearing particles between the snow taken in February 2021 and in January 2023, we found that the latter contained smaller amounts of Sb and Cu. In addition, the lead particles from the snow in January 2023 were rarely coated or covered by other particles (quartz, aluminosilicates or carbonates). The absence these small particles can aid in the detection of other elements.

3.3. Characterization of Lead-Bearing Particles in the Ammunition

We observed lead particles sampled as ammunition residue and dust at a shooting range in Postojna, Slovenia. The particles had various sizes, exceeding 100 µm, and shapes (irregular, spheres and agglomerates). The composition of the particles acquired from the ammunition was mainly Pb and Pb-O (Figure 5a–c) and the particles were more abundant in air rifle pellet dust, which was to be expected as the pellet dust was collected from the vicinity of the targets, while 22LR dust was collected from the bullet casings. Particles obtained from the 22LR bullet casings contained a very small amount of other elements, most commonly Cu and Sb (Figure 5e,f). Air rifle pellets also contained other dust particles, which influenced the composition of the lead particles due to the EDS measurement area. The surface morphology of the particles indicates that they were exposed to rapid changes during the shooting process. In comparison, Pb particles from Pokljuka biathlon shooting range were smaller as they were acquired at a greater distance from the shooting range. They also had morphological features (rounded edges, partially melted and deformed) showing rapid oxidation; however, their surfaces might have also been affected by post-depositional processes. The features mentioned are especially noticeable in particles from the ammunition samples (Figure 5).

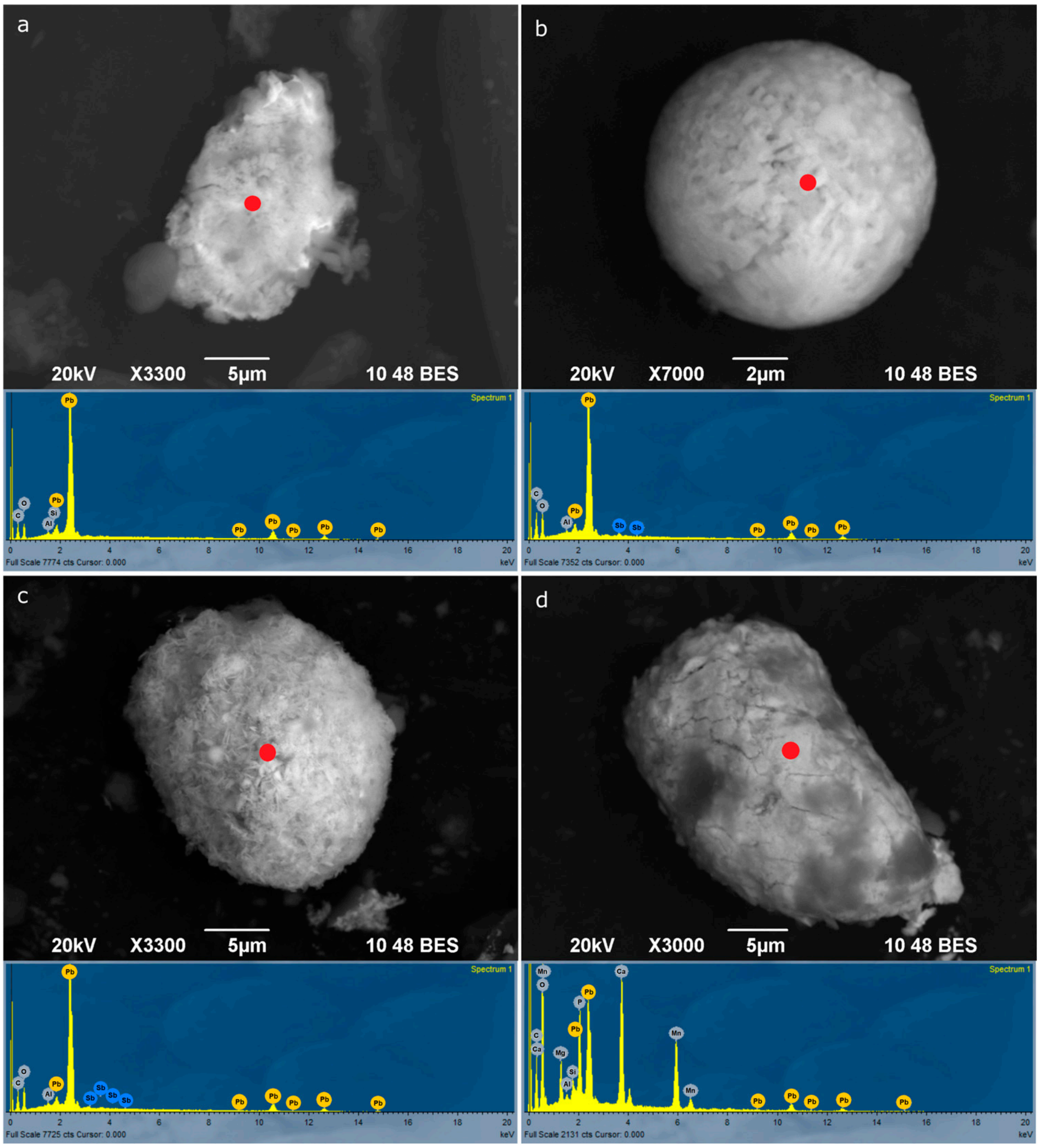

Figure 3. Particles composed of Pb-O in the snow sample from 2021. The presented images were taken in BSE modes as indicated in the lower right corner of the images. The red dots represent the location of the spectra. Particles in (**a**,**d**) show coatings on the Pb-O particle. Particles in (**b**,**c**) are spherical Pb-O particles with different textures. In EDS spectra images yellow represents Pb and blue represents Sb.

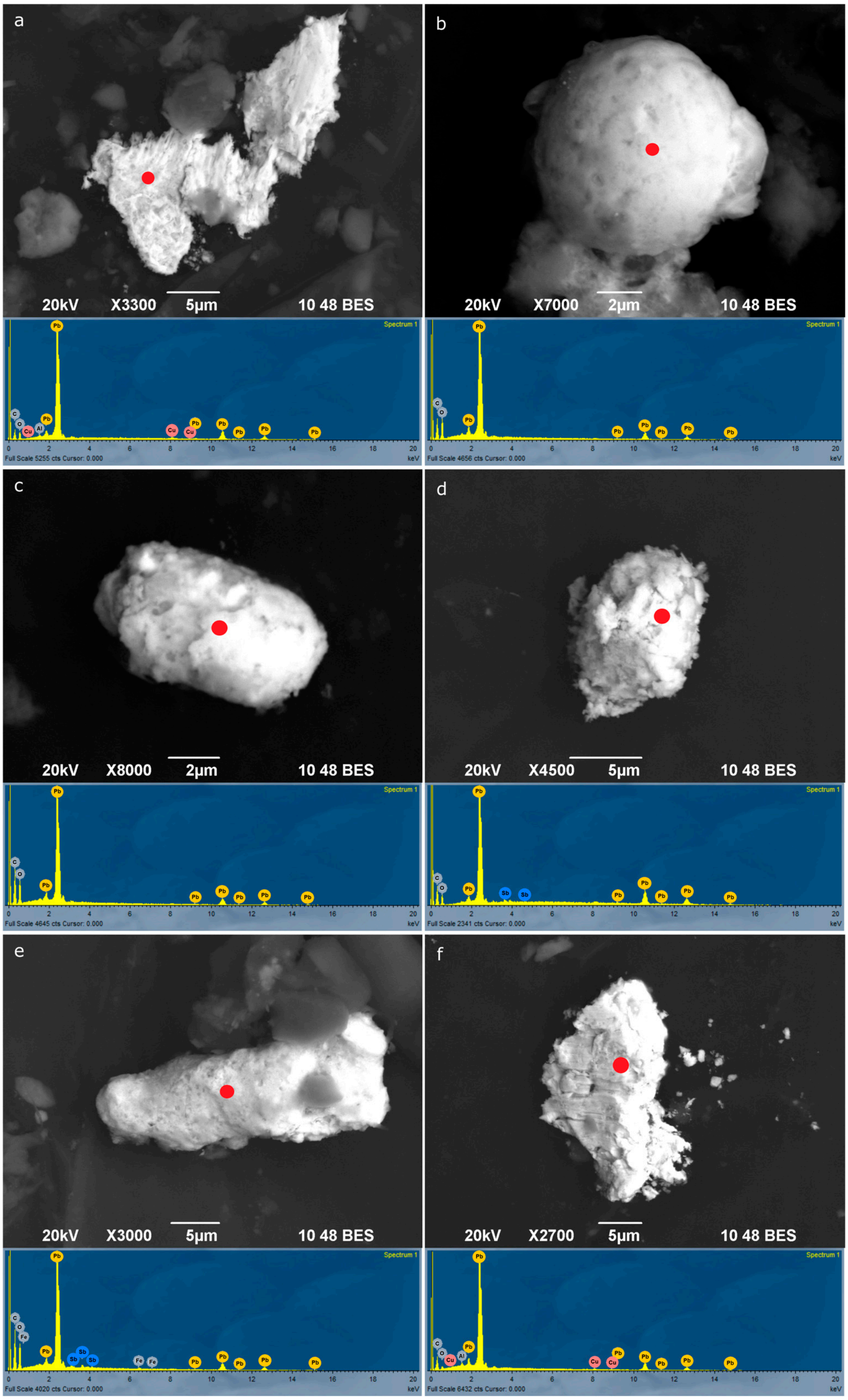

Figure 4. Particles composed of Pb-O in the snow samples from 2023. The presented images were taken in BSE mode as indicated in the lower right corner of the images. The red dots represent the location of the spectra. The particles in (**a**–**c**) were found in the snow deposited prior to the biathlon competition (M-1). The particles in (**d**–**f**) were found in the samples of the whole snow profile (M-4). In EDS spectra images yellow represents Pb, light red represents Cu and blue represents Sb.

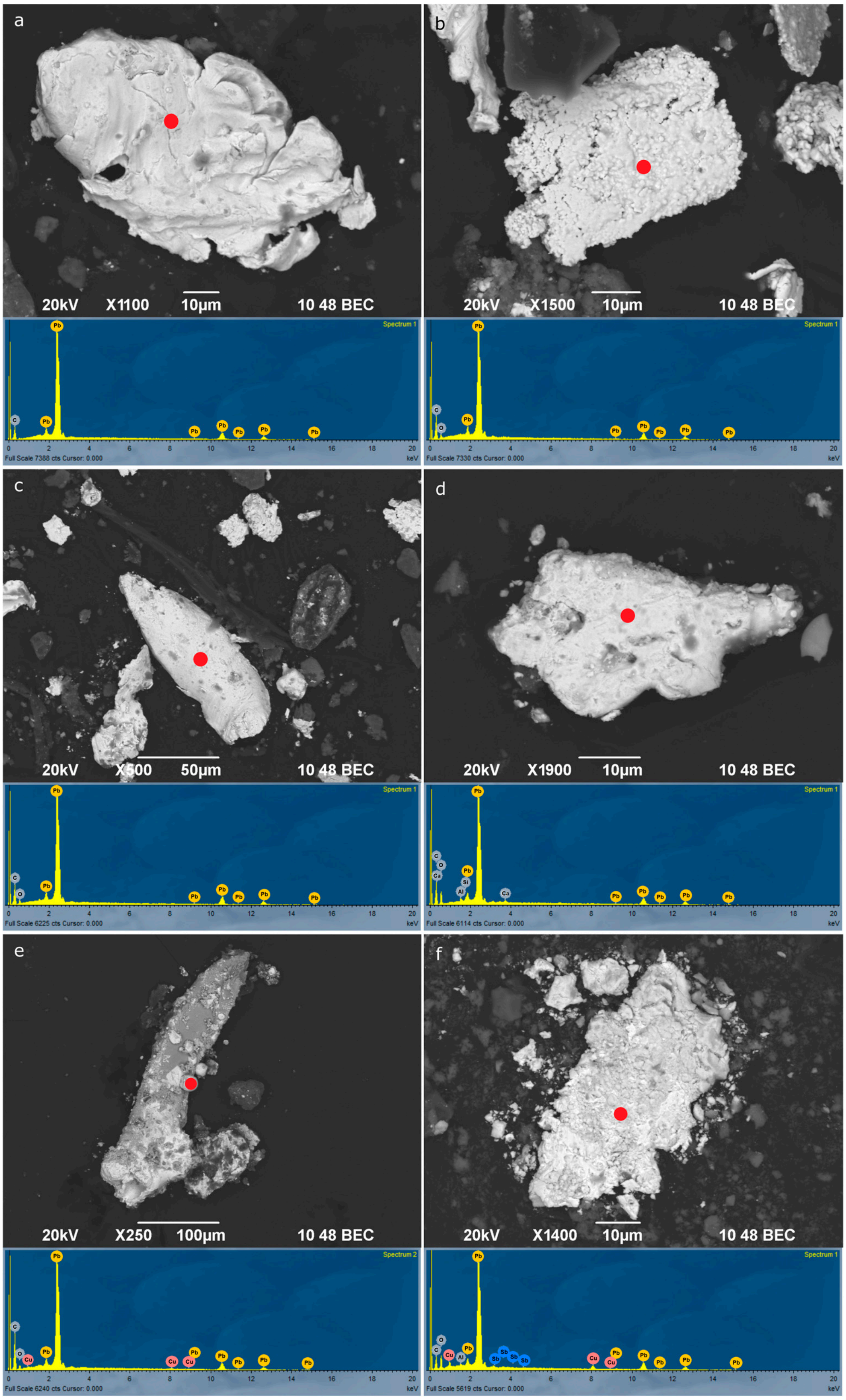

Figure 5. Particles composed of Pb-O in the ammunition samples. The presented images were taken in BSE modes as indicated in the lower right corner of the images. The red dots represent the location of the spectra. The particles in (**a**–**c**) were found in air rifle pellet dust. The particles in (**d**–**f**) were found in the 22LR bullet casings. In EDS spectra images yellow represents Pb, light red represents Cu and blue represents Sb.

4. Conclusions

Regarding the morphological characteristics (rounded edges, irregular shapes and partly melted) and chemical compositions of the solid particles found in the snow around the shooting range at Rudno polje in Pokljuka, we conclude that most of them originated from shooting. We have proven this by comparing particles from the area around the shooting range at Rudno polje with dust particles from a known indoor shooting range, although the sizes of the lead particles varied. The particles found on the Pokljuka plateau were smaller compared to the particles found in the indoor shooting range; this was likely due to the distance of the samples from the Rudno polje shooting range on the Pokljuka plateau, which was in contrast to the lead particles from the ammunition being larger as those samples were taken directly from empty ammunition casings. In both areas, the Pokljuka plateau and the indoor shooting range, the morphological features observed on the lead particles were related to rapid changes due to the shooting process.

Due to the presence of lead particles in the snow at the biathlon shooting range at Rudno polje on the Pokljuka plateau, the shooting activities can potentially have negative impacts on the environment, as the biathlon ammunition composition is primarily lead with traces of copper and antimony. Particles originating from these activities can potentially represent a risk to the environment and human health due to inhalation. The particles found on the Pokljuka plateau had various sizes, and while larger ones were mainly deposited quickly after their release close to the targets or the shooting positions, these small particles have the potential to remain in the atmosphere for longer periods of time.

To further determine the influence that biathlon shooting has on the Pokljuka plateau, we would need to determine the quantity of lead in other mediums such as soil and analyse the spatial distribution of lead and other elements in the vicinity of the shooting range in order to determine the impacted area.

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