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"Gheorghe Asachi" Technical University of Iasi, Romania

CHANGES IN PHYSICOCHEMICAL PROPERTIES AND CONTAMINATION WITH LEAD IN OUTDOOR SHOOTING RANGE SOILS

Gintarė Sujetovienė*, Jūratė Česynaitė

Department of Environmental Sciences, Vytautas Magnus University, Vileikos 8, 44408 Kaunas, Lithuania

Abstract

The negative environmental impact of the shooting ranges occurs through shot and bullets containing metals that are harmful to the environment. The objective of this study was to investigate the changes in physicochemical properties and contamination with heavy metals in soils of two Lithuanian outdoor shooting ranges. Soil samples were collected from the outdoor rifle shooting ranges which differed in shooting activity. The higher concentrations of Pb were found in the shooting range (8272 mg kg^{-1}) with higher activity compared with soil from the less active shooting range (6758 mg Pb kg^{-1}). Heavy metal concentrations in both shooting ranges increased with the distance from the firing line. The shooting range soil pH ranged from 6.2 to 7.4. Soil density of the shooting ranges varied between 1.03 g cm^{-3} to 1.54 g cm^{-3}. Organic matter content and porosity of the soil significantly decreased with increasing distance from the firing line along with an increase in soil density.

Keywords: lead, shooting range, soil contamination

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1. Introduction

Soil contamination is identified as a priority for the collection of policy relevant soil data to support soil management practices (Panagos et al., 2013). Pollution by organic and inorganic contaminants is practically irreversible and can seriously affect the ability of soil to perform its vital functions. Many studies of the heavily contaminated sites are indicated potential health impacts from high levels of soil contamination (Fayiga and Saha, 2016; Laidlaw et al., 2017; Li et al., 2017).

Contamination of shooting range soils is an increasing environmental concern (Conesa et al., 2010; Curran-Cournane et al., 2015; Hardison et al., 2004; Spuller et al., 2007; Urrutia-Goyes et al., 2017a; Urrutia-Goyes et al., 2017b). Accumulation of inorganic compounds is found in soils of the studied shooting ranges because bullets and jackets are made of alloys with different amounts of heavy metals (Conesa et al., 2010; Spuller et al., 2007). Aside from Pb, ammunition contains specific amounts of other contaminants, such as Sb, As, Cd, Cu, Ni, and Zn. These metals make up a small percentage of bullet mass Sb (1.9%), Cu (4.5%), Ni, and Zn (0.5%) (Conesa et al., 2010; Sanderson et al., 2012; Sanderson et al., 2014). Many recent studies have focused on contamination of firing range soils by Pb because it is the main component of a bullet (Conesa et al., 2010; Sanderson et al., 2012; Sorvari et al., 2006). Shooting range soils can contain high concentrations 70 350 mg kg^{-1} (Fayiga et al., 2011) or even 397 840 mg kg^{-1} of Pb (Dermatas et al., 2006a), which can be harmful to human health (Hardison et al., 2004; Murray et al., 1997).

Once the lead bullets and debris reach the soil, several factors determine the extent of the actual hazard it might pose. Usually dangerous

* Author to whom all correspondence should be addressed: e-mail: gintare.sujetoviene@vdu.lt; Phone: +370 37 327911; Fax: +370 37 203 858

concentrations of heavy metals are distributed unevenly throughout the area of a shooting range. It is reported that higher concentrations of heavy metals is appeared in the backstop berm (bullet traps) of shooting ranges (Okkenhaug et al., 2016). Despite spatial variability, Pb concentration depends on soil characteristics. Soil acidity is of particular importance because it controls the behaviour of metals. Heavy metal cations are highly mobile in acid soils and this makes them more available for plants or entering the water supply. Increased acidity of the shooting range soils results in dissolution and transformation of secondary Pb minerals (Cao et al., 2008; Sanderson et al., 2012). In contrast to acidification, under high soil pH the increase in the content of dissolved organic carbon due to restricted microbially decomposition resulted in the dissociation of H^+ from organic matter functional groups (Levonmäki and Hartikainen, 2007). Decomposition of organic matter is usually retarded in heavily polluted soil because Pb is observed to diminish respiration (Renella et al., 2004). A high pH was also observed to promote the formation and dissolution of organo-Pb complexes with higher organic matter content, which increase the solubility of Pb (Sauvé et al., 1998) since other studies have not recorded such a relationship (Levonmäki and Hartikainen, 2007). The increased lability of Pb at higher pH in a contaminated soil suggests a potential increase in bioavailability.

Along with the contamination of shooting range soils, physical properties of soil could also be affected. Shooting activities have a great effect on bulk density and porosity (Hardison et al., 2004). Falling bullets compact soil leads to lower content of organic matter, platy structure of the soil, decrease in the pore space and increase in the soil density (Stirzaker et al., 1996). They influence many important soil processes that impact plant growth and environmental quality. Therefore, soil density and porosity are the two most important parameters in assessing negative anthropogenic change in soil.

The possible environmental risk associated with contamination at shooting ranges is reported in various countries during the past few decades. High concentrations of contaminants in shooting range soils are far above the regulatory standards indicating potential environmental and health risks (Fayiga and Saha, 2016). In Lithuania, there are no data on the precise number of shooting ranges and the effects of the shooting activity on soil properties have not been studied. Since, the effects of the shooting activity on soil properties and contamination have not been studied in Lithuania yet, we focused on the changes in properties and contamination with heavy metals in two Lithuanian outdoor shooting range soils.

2. Material and methods

2.1. Study sites description

Field soil samples were collected from two outdoor shooting ranges located in central Lithuania. The first shooting range is located in the centre of Alytus city (54°23'48" N, 24°02'41" E, Site A) while the second shooting range was located in an industrial district of Kaunas (54°55'23" N, 23°59'41" E, Site B). Both ranges were opened, with no roofing and with two target lines at different distances (25 and 50 m). Shooting ranges were designed only for training of shooting sports athletes. The shooting ranges were used mostly to practice and improve their skills by shooting from different distances. The types of both ranges were small-bore rifle ranges and small-bore (up to 6.5 mm) weapons (rifles and pistols) and special ammunition (22.lr calibre, the diameter of the bullet ranged from 5.70 to 5.73 mm) were used in these ranges. Shotgun or other type of higher calibre guns cannot be used in these ranges due to safety regulations. The studied shooting ranges were structurally similar. The shooting ranges differed in their activity. Annually, a total of 80 athletes were trained in Site A while up to 40 athletes were training at Site B. The main elements of a shooting range were a firing line (i.e., shooting area) and target lines (i.e., area where the targets are located at the accurate distance).

2.2. Sample collection

Soil samples were collected from each shooting range at four different sites – 0, 24, 26, 50 m from the firing line (Fig. 1). This sampling design was selected according to the target lines. The first target line was located at 25 meters distance from the shooting range, the second was at 50 meters distance. While taking samples, it was kept in mind that the concentration of heavy metals might be very different between 0-24 meters compared with 26-50 meters distant because more bullet fragmentation occurs when the bullet hits the target.

At each distance, a square plot was delineated (0.2 × 0.2 m). Each soil sample consisted of five subsamples collected from the corners and centre of a square plot. Subsamples were mixed and homogenised to represent certain studied plot. Grassland soil from an unpolluted area, 13 kilometers away from the shooting range (54°25'43" N 24°14'08" E, Alytus, Lithuania) was collected and used as a reference.

2.3. Physicochemical analysis

The soil samples were air dried, homogenized and passed through 2 mm sieve. Samples were stored in plastic bags prior to laboratory analysis. The texture of the studied soils was loamy sand. The pH was measured potentiometrically in aqueous suspension with a soil-water ratio of 1:5 pH-meter (inoLab 720, WTW). Total soil organic matter content was estimated by loss on ignition method measured after burning an aliquot of approximately 5 g dried soil at 550°C in a furnace for 4 h (Ben-Dor and Banin, 1989).

Soil bulk density, particle density and porosity were determined according to the Hao et al. (2008).

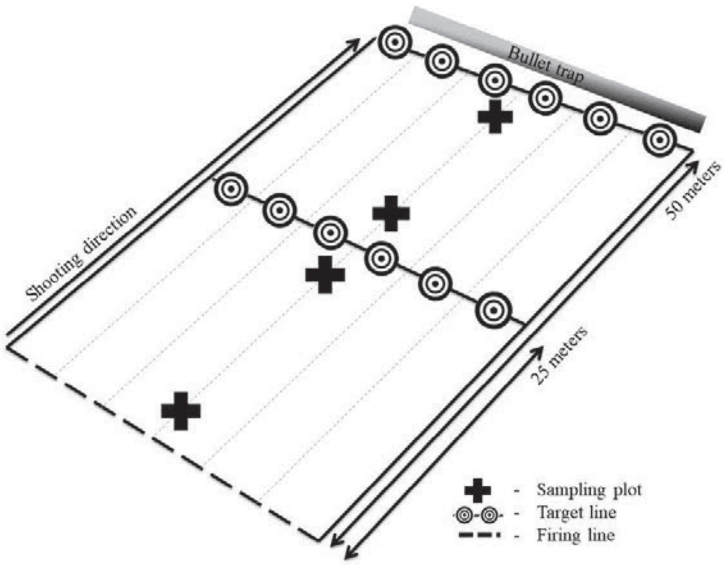

Fig. 1. Schematic representation of shooting ranges and sample plots.

Soil bulk density was determined by the core method pouring air-dried soil samples in a measured cylinder. Fresh weight of undisturbed samples was recorded in field and bulk density was calculated after samples were dried. Particle density is the volumetric mass of the solid soil. Porosity was calculated using values determined for bulk density and particle density. The percentage of porosity was calculated finding the ratio of bulk density to particle density and multiplying by 100. To analyze the contamination of the shooting range soils, we determined the concentration of Pb. Heavy metals concentration in soil sample was determined by taking about 0.5 g oven-dried soil followed by digesting it in 8 ml of HCl, 5 ml of HNO_3, 5 ml of HBr, 3 ml of HF in a Teflon vessel using a microwave digestion system (Milestone Ethos One). Concentrations of heavy metals were determined by atomic absorption spectrometer (Shimadzu AA–6800, Japan) with a graphite furnace atomizer (GFAAS). The elemental standard solutions were prepared by diluting a stock solution of 10000 mg L^{-1} Pb. The recovery was tested at four spike levels. A soil sample was divided into several aliquots. In one of the aliquots, initial concentrations of metals were measured. Other aliquots were spiked with four different concentrations of respective metal standard solutions. The final concentrations of the metals in the spiked samples were determined, and the respective recoveries were calculated. Accuracy studies were performed by analyzing certified reference material (ISE sample 918). The mean recovery of Pb was 92–109%.

2.4. Data analysis

Data were checked for normality and homogeneity of variance (Kolmogorov–Smirnov test). Log transformation had to be applied to obtain normal distribution and homogeneous variances of the data. A nonparametric Mann-Whitney U test was used to assess the statistical differences between means ($p<0.05$). Spearman's correlation analysis was conducted to test the significance of the relationships between different soil parameters ($p < 0.05$). Analysis of variance (ANOVA) was used to illustrate the impact of distance from the firing line to target line on the heavy metals concentration in soil. The Statistica 7 package was used for the statistical analysis.

3. Results and discussion

The physicochemical characteristics of the firing range soils are presented in Fig. 2. The pH of shooting range soils was slightly alkaline. Values varied from 6.45 to 7.12 and were significantly higher compared with reference soil (Mann-Whitney U test, $p<0.01$; Fig. 2). There was no significant difference between different shooting range soil's pH values ($p>0.05$). The highest organic matter content was at the firing line of shooting ranges and tended to decrease with increasing distance from the firing line towards the target line ($F = 15.88$, $p<0.001$). The organic matter content in the soil of the less active shooting range was significantly lower in comparison with the more active range ($p<0.05$).

The bulk density of the shooting range soils ranged from 1.03 g cm^{-3} to 1.54 g cm^{-3} and it was significantly higher in comparison with the reference ($p<0.05$). The highest density was observed at the second target line (site A – 1.23 g cm^{-3}, site B – 1.54 g cm^{-3}, Fig. 2). The porosity of the studied soils was similar in both shooting ranges and significantly lower (30%) compared with the reference samples (Fig. 2). The highest porosity (57%) was at the firing lines and the lowest, at the second target line. Porosity and density of soil did not change significantly with distance from the firing line towards the target line ($p > 0.05$).

The physicochemical characteristics of shooting range soils were changed under the influence of the activity.

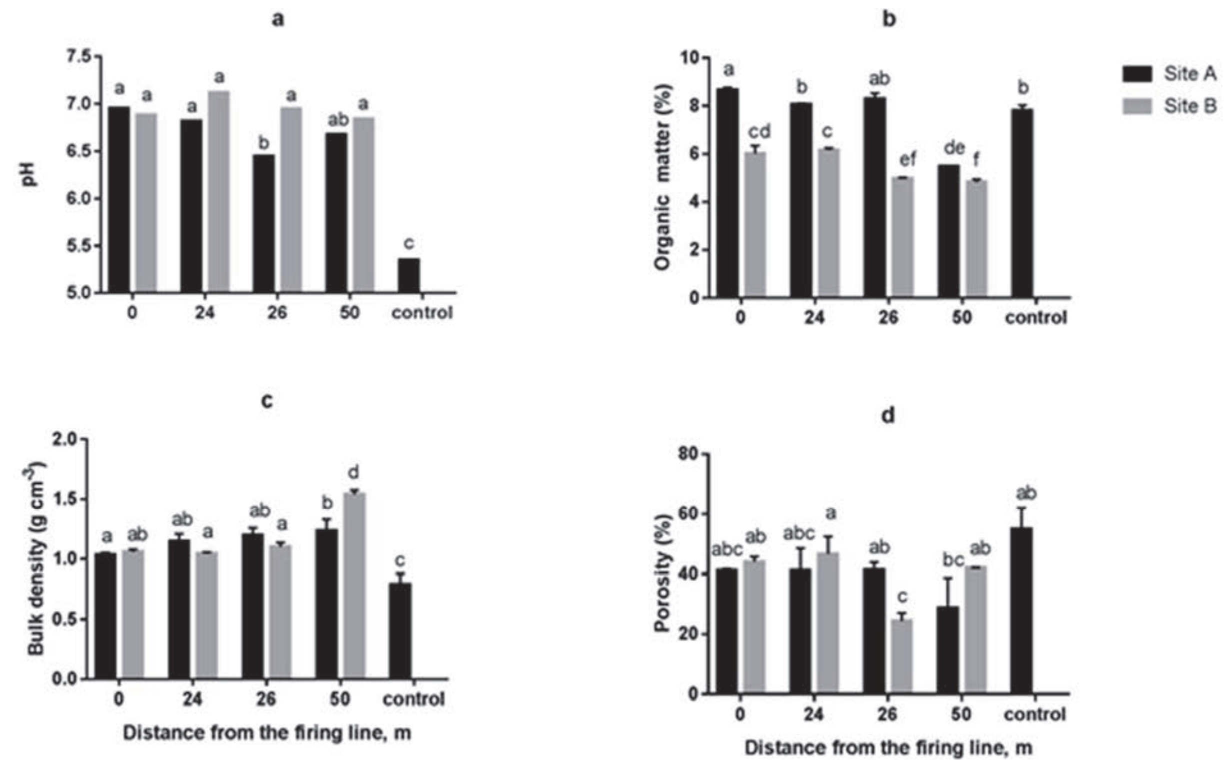

Fig. 2. Physicochemical characteristics of soil collected in Lithuanian shooting ranges at different distances from the firing line (mean ± SE): a – soil acidity, b – organic matter content, c – soil density, d – soil porosity. Site A represent more active shooting practice than Site B. Different letters in the graph represents significant differences between the study sites (Mann-Whitney U test; $p < 0.05$)

The increase in soil density along with a decrease in soil porosity was observed with the most significant changes at the farthest distances from the firing lines toward the second target line (50 m). If we compare shooting ranges according to the shooting activity, the soil exposed to more intense shooting activity (Site A) was spatially more affected than in the less intense range (Site B), but the latter had the higher values of soil density at the second fire line. This indicated the highest soil compaction as a result of the impact of applied pressure and accumulation of bullets. Soil compaction is considered a physical form of soil degradation that alters soil structure, limits water and air infiltration, and reduces root penetration in the soil (Nawaz et al., 2013). The constant addition of the fired bullets to the soil caused soil particles to compress together, and bullets were exposed to air and water on the surface of the soil. The values of surface soil density in our shooting ranges were higher compared to the results of other studies (Knechtenhofer et al., 2003: 0.47-0.59 g cm^{-3}). The observed comparatively higher density of surface soil suggests that accumulation of the bullets on the surface caused physical soil degradation over time. Compacted soils usually have a reduced rate of water penetration through the compacted layer. Changes in the porosity of soil could affect the dispersion and mobility of inorganic colloids and dissolved organic matter because diffusion of dissolved substances takes place in the pores. Studies showed that the mobility of heavy metals was determined to be higher in soils with greater porosity (Avnimelech et al., 2001; Sherene, 2010). Higher soil density and lower porosity were observed in parallel with the lowest organic matter content in soil at the second target line. This was in accordance with results of other studies where berm soil had a lower content of organic matter (Okkenhaug et al., 2016). Murray et al. (1997) observed that the soils had low to medium organic matter content at an outdoor skeet and trap shooting range and increased in the area where shooters stood and an organic matter content decrease in the primary downfall area for pellets. The decrease in the amount of organic matter was parallel with the observed reduced cover of vegetation. Since vegetation of shooting ranges will lead to uptake and immobilization of heavy metals by plant root exudations in the rhizosphere, the unvegetated areas could results in an increase in the leaching of metals. The Pb phytostabilization capacity of spontaneous vegetation (*Agrostis capillaris* L.) was indicated by the presence of secondary mineral phases of metallic Pb in the adjacent soil (Rodríguez-Seijo et al., 2016). Fayiga and Saha (2016) reported that the presence of grasses reduced metals leaching through the accumulation process. Root exudates in the rhizosphere could result in changes in soil pH and increased weathering of metals. Therefore, the observed decrease in the content of organic matter and porosity could impede the transformation and increase in accumulation of metals. Ma et al. (2007) showed that the presence of organic matter accelerated weathering because of the formation of organic acids from organic matter decomposition and microbial activity in the soil. Soil pH was generally a principal factor governing concentrations of soluble and plant available metals (Brallier et al., 1996). The pH of shooting range soil was higher compared with reference soil. Because soil acidity stimulates solubility and migration of heavy metals (especially mobile forms), the increase in soil pH could result in a decreased availability of heavy metals (Berbecia et al., 2011; Jörgensen and Willems, 1987). Okkenhaug et al. (2016) noted that the dissolved concentration of Pb and Sb was dependent on pH value; the increased mobilization of Pb at lower pH was observed. Therefore, soil pH is one of the most critical factors that influence metal distributions in surface soils.

Lead concentration increased with increasing distance from the firing line towards the target line and the highest content was found in the soil of the second target line (Fig. 3). The concentration of Pb in shooting range soils was significantly higher than the reference samples (8.1 mg kg^{-1}; $p < 0.05$).

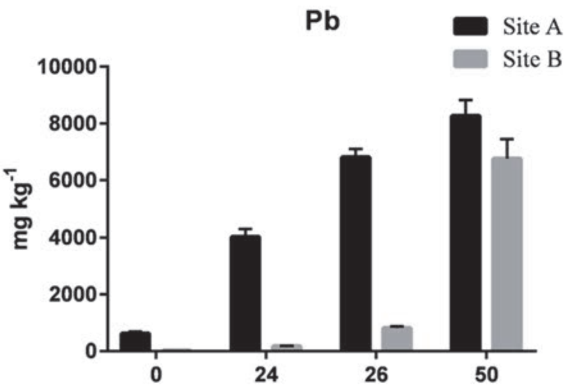

Fig. 3. The concentration of lead (mg kg^{-1}) in soil of different shooting ranges

Analysis showed that the distance between the firing and target line had a significant effect on the higher concentration of Pb in the soil samples ($F = 8.45$, $p < 0.05$). The mean concentration of Pb was related to shooting activity: higher in the more active shooting range soil (site A) than in the less active (4933 and 1941 mg kg^{-1}, respectively, Fig. 3). Correlation analysis showed that Pb concentration was negatively correlated with soil acidity ($r = 0.83$, $p< 0.01$) and positively with soil density ($r = 0.81$, $p<0.05$).

The contamination of shooting range soils with lead originating from shots and bullets had received much attention because elevated amounts of this metal could cause adverse health effects. Extensive amounts of Pb were being deposited in the environment as a result of activities that are taking place at shooting ranges. The detected levels of Pb in studied shooting range soils varied from 20 to more than 8200 mg kg^{-1}. About 60% higher concentrations of Pb was found in the more active range compared to the less active one and confirmed the influence of shooting activity level on Pb concentrations in soil. The values of Pb content were in the same concentration range as previously reported in studied shooting range soils (e.g., 1-10403 mg kg^{-1}: Sanderson et al., 2012; 21-5680 mg kg^{-1}: Etim and Onianwa, 2012; 52–3400 mg kg^{-1}: Lin et al., 1995). However, other studies had revealed even higher values in more intense and/or long-term used ranges – maximal Pb concentrations up to 70 350 mg kg^{-1} (Fayiga et al., 2011); 48 400-49 228 mg kg^{-1} (Cao et al., 2003b; Dermatas et al., 2006b) or even up to 397 840 mg kg^{-1} (Dermatas et al., 2006a). These differences in the level of contamination were related not only to activeness of shooting but also to the age of the range.

The elevated Pb concentrations in surface soils of the shooting ranges resulted from the accumulation of a high density of bullets (Chen and Daroub, 2002; Fayiga and Saha, 2016; Murray et al., 1997). It was suggested that small calibre bullets (e.g. .22 lr) are plain lead bullets basically pressed into a bullet shape from pure lead, this type of ammunition is only coated with thin layer of copper, wax or oil. Plain lead ammunition is softer than larger calibre ammunition, which is coated with harder material (e.g. alloy of 90%Pb, 5% Sn, 5% Sb). Therefore, greater bullet fragmentation could appear in ranges where small-bore guns are used. A portion of the lead bullet disintegrates into fine fragments while passing through the gun due to misalignments of the gun barrel (Cao et al., 2003a; Tripathi and Llewellyn, 1990). The lead particles, along with dust and fumes originating from the lead primer and the bullet fragments were ejected from the gun barrel. Therefore, the concentration of Pb exceeded the maximum Lithuanian guidance value concentration that ensure safety (100 mg kg^{-1}) in Site A at the firing line by almost eightfold (HN 60:2004, 2004). The distance from the firing line was correlated with the concentration of lead; it increased with increasing the distance from the firing line towards the target line. This is in accordance with the results of substantial amounts of Pb close to the target lines because of the fragmentation of bullets along with the release of Pb powder causing deposition of Pb dust on the soil surface (Cao et al., 2003b; Etim and Onianwa, 2012; Fayiga and Saha, 2016; Sanderson et al., 2012).

Results of our study confirm that soil physicochemical characteristics along with climatic variables are very important in determining the breakdown of bullets and transformation of secondary minerals (Cao et al., 2003a; Cao et al., 2003b; Ma et al., 2007). Knowing the relationships among variables, we could project the rate of conversion to more labile forms and transfer of metal to the subsoil. This evidence is helpful in risk assessment and had an implication for the proper management of these sites to minimize the adverse effects of Pb on the environment.

4. Conclusions

This study highlighted changes in soil physicochemical characteristics (pH, organic matter, soil density, porosity) in shooting range soils. The organic matter content and porosity of the soil decreased with increasing distance from the start of the firing line while soil density increased with the distance. Surface soil was enriched in Pb derived from bullets. The concentrations of Pb in the soil at the studied ranges were found to be significantly higher than the background concentration and were far above the regulatory standards indicating potential environmental and health risks. Soil was more contaminated with lead in the more active shooting range than in the less active shooting range. Pb concentrations in both shooting ranges tended to increase with the distance from the start of the firing line. Further studies are needed to combine physicochemical and biological assays to evaluate heavy metal toxicity and bioavailability of the shooting range soils.

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