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Review

Soil pollution at outdoor shooting ranges: Health effects, bioavailability and best management practices☆

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A R T I C L E I N F O

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A B S T R A C T

The total lead (Pb) concentrations of the surface soil, sub surface soil, vegetation and surface waters of outdoor shooting ranges are extremely high and above regulatory limits. Lead is dangerous at high concentrations and can cause a variety of serious health problems. Shooters and range workers are exposed to lead dust and can even take Pb dust home to their families while some animals around the shooting range can ingest the Pb bullets. The toxicity of Pb depends on its bioavailability which has been determined to be influenced greatly by the geochemical properties of each site. The bioavailability of Pb in shooting ranges has been found to be higher than other metal contaminated soils probably because of its very low residual Pb (<1%). Despite being an immobile element in the soil, migration of Pb within shooting ranges and offsite has been reported in literature. Best management practices to reduce mobility of Pb in shooting ranges involve an integrated Pb management program which has been described in the paper. The adoption of the non-toxic "green bullet" which has been developed to replace Pb bullets may reduce or prevent environmental pollution at shooting ranges. However, the contaminated soil resulting from decades of operation of several shooting ranges still needs to be restored to its natural state.

1. Introduction

Shooting ranges are recreational facilities which provide a place for training in the use of ammunitions. Lead (Pb) is the primary contaminant at shooting ranges because bullets and pellets are mainly composed of Pb. Pb shot contains 97% Pb while Pb bullets contains 90% metallic Pb (Scheuhammer and Norris, 1995; Cao et al., 2003a). Pb is ranked as the most common inorganic contaminant on the national priority list of sites in the United States because of its widespread environmental and health impacts (Smith et al., 2011).

Pb is a contaminant of concern in the environment because of its toxicity and adverse effect on human and ecosystem health. There are three ways Pb ammunition used in outdoor shooting ranges can have an adverse effect in the environment; first, ingestion of bullets can poison wildlife; secondly, they can contaminate ground water and poison wells and third effect is that they can also contaminate nearby surface water bodies impacting the aquatic ecosystem (VPC, 2001). The pollution of water bodies has implications for public health and exposure of wildlife to Pb can affect their survival. Venäläinen (2011) stated that the ecotoxicity of Pb depends on its bioavailability.

Bioavailability has been defined as the soil pool most available to living organisms which integrates chemical, physiological and toxicological aspects of the environment (McLaughlin, 2001; Lanno et al., 2004; Hettiarachchi and Pierzynski, 2004; Berthelot et al., 2009). The total amount of metals in the soil is not available because they are adsorbed or bound by organic and inorganic constituents in the soil such as organic matter, Fe and Al oxides and clay minerals. Bioavailability of metals can be determined by measuring the fraction of free metals present in the soil solution or by determining uptake by plants, soil biota or absorption into blood stream of humans (Hare and Tessier, 1996).

Soil Pb bioavailability depends on the solubility of Pb solid phases and other site-specific soil chemistry (Hettiarachchi and Pierzynski, 2004). Retention of metals on the solid phase of the soil is affected by pH, cation exchange capacity (CEC), organic matter and mineralogy of the soil (Takac et al., 2009). Soil pH is the most important factor affecting bioavailability because it determines solubility and soil reactions (Muhlbachova et al., 2005).

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Soil reactions that control bioavailability include sorption/desorption, complexation and redox reactions (Violante et al., 2010; Hernandez-Soriano and Jimenez-Lopez, 2012).

The soil chemistry and mineralogy of shooting range soils are very unique and different from other metal contaminated soils due to the fragmentation of lead bullets after impact. The extent of fragmentation depends on the distance travelled, the soil type, bullet velocity and force of impact (Griggs et al., 2011). Dermatas et al. (2006a) reported that Pb bullet fragments played an important role in the rate and amount of Pb released in shooting range soils. The transformation of metallic Pb fragments into secondary Pb minerals in shooting ranges may also increase the mobility of Pb thereby dispersing Pb contamination in the environment.

The mobility of Pb has been reported at shooting ranges by several scientists indicating the need for urgent remedial action and effective management practices in shooting ranges (Cao et al., 2003a; Chrastny et al., 2010). Several best management practices have been recommended by United States Environmental Protection Agency (USEPA) to reduce the migration of Pb at shooting ranges. The goal of this paper is to review 1) pollution in shooting ranges and the resulting health effects 2) the effect of soil chemistry and mineralogy on bioavailability of Pb 3) best management practices for shooting ranges.

2. Soil pollution in outdoor shooting ranges

The National Shooting Sports Foundation estimates that there are 10,000 shooting ranges in the United States (Sullivan et al., 2013). This suggests that many people are involved and indicates that Pb is being deposited at an increasing rate on the soils of these shooting ranges. It has been estimated that about 80,000 tonnes/year of Pb was used in the production of Pb ammunition in the United States in the late 1990s (USEPA, 2001; Hardison et al., 2004). About 200–6000 tonnes of Pb was deposited annually in the Netherlands, Denmark, Canada and England shooting ranges (Jørgensen and Willems, 1987). Annual Pb loading of 15 tonnes was reported for shooting ranges in Finland (Tanskanen et al., 1991), more than 400 tonnes for Switzerland (Evangelou et al., 2012) and more than 500 tonnes in Sweden (Lin et al., 1995).

The sources of Pb pollution at shooting ranges are the bullets which are aimed at targets positioned at several distances from the shooting box. These bullets are fragmented and pulverized upon impact with the ground, backstop, berms or bullet trap (O'Connor et al., 2009). Fragmentation alters the particle size distribution of shooting range soils and weathering of these fragments eventually results into lead contamination of the environment.

Though several metals and metalloids such as Pb, Cu, Sb, As, and Zn are used in the manufacture of bullets, Pb, because of its higher concentration and toxicity, is the most studied and monitored at shooting ranges (Bannon et al., 2009). Pb-bullets contains 95–97% metallic Pb by weight, 0.4–2.0% Sb, 0.2–0.8% As, and mean concentrations of Sn, Se, Mn, Cd, Cr, Cu, and Ni greater than 30 $mgkg^{-1}$ (Yin et al., 2010).

2.1. Distribution of Pb in shooting ranges

The total soil Pb of shooting ranges varies both vertically down the soil profile and horizontally within each range showing the heterogeneity of Pb contamination at these sites. Cao et al. (2003a) reported that Pb concentration increased as distance from the firing line increased. They also detected substantial amount of Pb in surface soils close to the firing line (close to shooting box) probably because discharge of bullets is accompanied with release of Pb powder causing deposition of Pb dust on the soil close to the firing line.

This was corroborated by Edwards (2002) who reported that significant amounts of fine particulate Pb was generated during shooting, close to the shooting box but absent at distances beyond 50 m. They reported that maximum concentrations occurred at distances of 28 m, 80 m and 180 m. Contrary to this report, a previous study reported that between 0 and 72 m, there were no bullets or bullet fragments in shooting range soils in Florida (Chen et al., 2002).

The berm (also known as stop butts or backstop) is the area behind the target into which the shot impacts having passed through the target. It's usually soil built like a slanting wall and contains the highest density of Pb bullets in shooting ranges. A previous study in a shooting range (operated 14yrs) found weathered bullets and fragments weighing 180 g kg^{-1} soil in the middle of the berm (mid-berm) (Chen et al., 2002). Bullets weighing 400 g kg^{-1} were reported in the mid-berm soil of a Denmark range (operated 30 yrs) while the top-berm soil contained bullets weighing 47 g kg^{-1} soil (Astrup et al., 1999).

The high density of bullets in the mid-berm soil may be responsible for the elevated total Pb concentrations (Table 1) of these soils. Total Pb soil concentrations ranged from 10,068 to 70,350 mgkg $^{-1}$ in the mid-berms of shooting ranges in Florida (Chen et al., 2002; Cao et al., 2003a, b; Fayiga et al., 2011). In another study, total Pb concentrations of berm soils of shooting ranges ranged from 1025 to 27,417 mgkg $^{-1}$ (Dermatas et al., 2006a). These total Pb concentrations in the surface soils are far above USEPA limit of 400 mgkg $^{-1}$.

Elevated Pb concentrations have been reported in berm soils in other countries apart from the USA. Mean Pb concentrations of the berm soil (5680 ± 2700 mg $Pbkg^{-1}$) was significantly higher than other locations at a shooting range in Nigeria, (Etim and Onianwa, 2012). Similarly, total soil Pb concentrations between 3400 and 5000 $mgkg^{-1}$ were reported in shooting ranges in England and Sweden (Lin et al., 1995) while maximum Pb concentration of berm soils of small arms firing ranges in Canada was 27,100 mgkg $^{-1}$ (Laporte-Saumure et al., 2011).

The highest concentrations of Pb was reported in New Zealand with total Pb ranging from 1.5 to 21% for three Canterbury clay target shooting range soils showing extensive contamination (Rooney and McLaren, 2000). Pb concentration reduced with depth in one of the shooting ranges while elevated Pb concentration was detected at the greatest depth in another shooting range. In Australia, the maximum concentration of soil Pb was found in the surface soils with up to 81,000 mgkg $^{-1}$ in stop butt while concentrations in the subsurface were up to 21,000 mgkg $^{-1}$ (Sanderson et al., 2010).

2.2. Mobility of Pb at shooting ranges

It is well known that Pb is an immobile element in the soil but several studies have reported high Pb concentrations in subsurface soils (Murray et al., 1997; Rooney and McLaren, 2000; Cao et al., 2003a; Olive, 2006; Sanderson et al., 2010); surface waters (Stansley et al., 1992; Cao et al., 2003a); groundwater (Sorvari, 2007) and plants (Cao et al., 2003a) in shooting ranges indicating the mobility of Pb.

Rooney and McLaren (2000) attributed the mobility of Pb to prevailing environmental conditions such as water-logging of a soil with elevated surface soil Pb concentrations. Cao et al. (2003a) explained that Pb mobility was due to enhanced solubilization of organic Pb complexes at alkaline soil pH. Sanderson et al. (2010) attributed Pb mobility in the shooting range soil to acidic soil pH indicating a proton induced metal release.

The migration of Pb in shooting range soils can eventually result in the contamination of nearby surface water and underground