Experience of Lead Pollution Control in Outdoor Shooting Ranges in the United States

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Abstract. In this paper, the exposure ways and risks of lead in the shooting field are discussed, the factors influencing lead migration in the soil are analyzed, and the control measures and the methods of removing and recovering lead in the soil are summarized. The results demonstrate that soil pH has a great influence on the mobility of lead, which can be reduced by applying lime to improve soil pH. However, this approach is only suitable for daily maintenance. The removal of lead completely depends on screening and soil leaching, which can remove more than 99% of the lead particles.

Keywords: lead, shooting ranges, control.

1. Introduction
Lead alloys and other metals are widely used in various applications such as ammunition preparation. The total amount of lead alloys used in the United States each year accounts for half of that in the world. Only in 2008, the United States consumed 74500 tons of lead alloys to make lead bullets. Lead, antimony, and arsenic account for 95-97%, 0.4-2.0%, and 0.2-0.8% of the total weight, respectively; the average concentration of tin, selenium, manganese, cadmium, chromium, copper, and nickel is above 30mg·kg⁻¹. According to statistics, there are 9600 outdoor shooting ranges in the United States, and the lead bullets used in the shooting will discharge millions of pounds of lead into the environment every year. This part of lead may be absorbed by plants, or enter into groundwater with rainwater runoff, resulting in a severe impact on the environment. Although using lead bullets to shoot birds and other animals is prohibited in many areas, the pollution control of lead bullets is still an urgent issue to be studied because it is the most cost-effective bullet type at present.

2. Exposure route and risk of lead

2.1. Exposure route of lead
The shooting range usually uses the slope protection composed of natural hillside or artificial soil slope. Metal fragments and particles are produced by lead bullet hitting the soil and exposed for a long time. After a series of weathering actions such as oxidation, decomposition, adsorption and precipitation, the metal migrates to the soil [1].

Cao et al. [2] discovered that found that in a shooting range operated for 20 years in Florida, there was 4.84% lead in the soil of the barrage wall and a relatively high content of lead detected at the
depth of 80cm in the soil profile of the shooting range. Lead can enter the environment of the range in one or more of the following ways: (1) lead will oxidize when exposed to air and dissolve when exposed to an acid solution or soil; (2) lead particles and dissolved lead brought by lead bullets can be moved by rainwater runoff; (3) dissolved lead can be transported to groundwater through the soil. The three main sources of human exposure to lead are lead dust, lead in soil, and lead in drinking water.

Generally, the exposure of lead to the human body in the shooting field is intake, inhalation, or contact with the lead contaminant. The first route of exposure is direct exposure to lead or lead particles that cause human intake. For example, lead particles produced by the discharge of firearms can be attached to the shooter's hands. If the shooter eats or smokes after shooting or before washing his hands, these lead particles will be absorbed into the body. The second route of exposure is to inhale lead dust because the shooter will be exposed to lead dust when shooting. Range workers may also be exposed to lead dust during routine maintenance operations, such as cleaning targets or warhead traps. The third route of exposure is exposure to pollutants containing lead, such as soil containing lead and lead bullets.

2.2. Lead exposure risk
Lead is one of the three major heavy metal pollutants. It is a toxic heavy metal element severely endangering human health. Lead exposure may cause organ damage and dysfunction in adults, and even coma or death in severe cases. Lead can affect children’s intellectual development, causing reading and attention problems. The study found that the blood lead concentration of shooters in an outdoor shooting range in South Africa was higher than that of archers in the same range. Angela Mathee et al. [3] found that the blood lead concentration of 84% of shooters and a quarter of archers exceeded the reference level of adult blood lead proposed by the American Institute for Occupational Safety and Health. This study confirmed the health problem of lead exposure can cause by lead bullet shooting.

Elevated lead levels in animals can also have harmful effects. Excessive exposure to lead is mainly ascribed to the increased mortality of cattle, sheep, and waterfowl. Livestock and wildlife may drink from contaminated streams or graze on pastures contaminated by shooting ranges [4]. Birds such as waterfowl regard lead particles as food or sand, making it easy to be eaten by mistake and resulting in acute poisoning death.

3. Influencing factors of lead migration in soil
In the process of shooting, used lead bullets are directly accumulated on the soil. When lead is exposed to air and water, it may oxidize to form several compounds. The types of compounds produced and their mobility are significantly affected by soil characteristics (such as pH and soil type). Thus, understanding the soil characteristics of shooting ranges is crucial to effectively control lead pollution.

3.1. pH value
(1) Soil pH. Under acidic (pH < 6) or alkaline (pH > 8) conditions, lead is more likely to react and migrate, suggesting that the waste lead bullets left in the soil may eventually decompose, migrate, and pollute the underlying soil. In moderately alkaline soil (pH 7-8.5), lead will precipitate from the solution and combine with the soil, inhibiting the migration of lead to deeper soil.

(2) Rainwater and surface water pH. When lead is exposed to acid solution or soil, it will be decomposed into lead oxides, carbonate compounds, and other soluble compounds due to weathering. These compounds dissolve and migrate with rainfall. The increase in the pH value of precipitation will contribute to the precipitation of dissolved lead and the decrease of lead concentration in water.

(3) Groundwater pH. During the period of no precipitation, most of the water in the river comes from the groundwater discharged into the river. Consequently, the pH of groundwater will affect the pH of surface water and further impact the solubility of lead during a rainstorm.
3.2. Annual precipitation
The larger the annual precipitation, the higher the frequency of heavy rainfall, the longer the contact
time between lead and water, the faster the weathering rate of lead, and the higher the risk of off-site
migration of lead. The bullet catcher in the outdoor shooting range will form a layer of lead slag after
use, which is very small and easy to be transported by rainwater. Besides, the smaller the particle, the
faster the degradation rate. Bullet traps need to be equipped with rainwater collection devices or laid
with a rainproof layer to minimize the release and migration of lead. The increase of precipitation will
reduce the content of calcium and other soluble minerals in the soil, as well as the precipitation of lead.

3.3. Soil properties
(1) Soil cover. The thicker the layer of fallen leaves and peat on the soil, the lower the content of lead
in water. Organic materials have a strong binding ability to lead.

(2) Soil type. The soil composed of heavy clay is relatively dense, and the free lead ions will adhere
to the clay particles. As a result, the rapid migration of lead compounds underground will be prevented.
From another perspective, the infiltration rate of rainwater in sandy soil is faster than that in clay soil.
Thus, the runoff of low permeability clay area is larger than that of high permeability sandy soil under
certain rainfall intensity, contributing to accelerating the migration of lead to a certain extent.

(3) Soil composition. The acidity of rainwater decreases with the dissolution of alkaline
components in the soil. If there are enough minerals in the soil, such as calcium, magnesium, and iron,
lead will be precipitated from the solution soon. Different soil physical and chemical properties will
affect the weathering rate and the availability of pollutants in the soil [5].

3.4. Vegetation cover and human structure
Vegetation cover will affect the mobility of lead and lead compounds. Vegetation absorbs rainwater,
reducing lead exposure to water. Besides, vegetation can also slow down surface water runoff and act
as a filter to prevent lead migration outside the site. However, lead recovery in wooded areas may be
reduced due to the inaccessibility of some large lead removal machines. Therefore, it is necessary to
understand the vegetation type, concentration, and diversity in the area.

Dikes impede the flow of water, greatly reducing the size and weight of lead particles in the water.
Since the lead particles are heavier than other suspended particles of similar size, the lead particles
will deposit when the water flow speed decreases.

4. Control measures of lead migration in soil

4.1. Adjusting soil pH

4.1.1. Lime addition. (1) Increasing soil pH. The pH value of the soil should be controlled between
6.5 and 8.5. If the pH of the soil is below 6, lime should be applied to increase the pH. Spraying lime
in the area where lead bullets accumulates can inhibit the migration of lead, increase the pH of upper
soil to the ideal level, and reduce the migration ability of lead. This is a simple, low-cost method. Lime
can be easily spread through a fertilizer spreader. The smaller limestone particles are suitable to be
directly sprinkled on the soil surface, and the larger limestone particles are suitable to be stacked in the
drainage ditch. The lead migration can be reduced by increasing the pH of rainwater runoff. M.
Levonmäki et al. [6] found that the application of lime in the soil can effectively reduce the mobility
of lead in the soil, which can be used as a daily maintenance measure of shooting range once a year,
but other methods are still needed to control the migration of lead. At the same time, lime may also
increase the solubility of As, resulting in negative effects.

(2) Breaking capillary effect. Another way to control the migration of lead is to destroy the
capillary effect at the bottom of the retaining wall. Considering that most of the soil materials used in
the retaining wall have porosity similar to that of the capillary, water is drawn up to the capillary edge
at the bottom of the retaining wall. The height of water rise depends on the soil material of the
retaining wall. Water rises more than 6 feet in clay, more than 3.3 feet in silt, more than 1.3 feet in fine sand, more than 5 inches in coarse sand, and more than 2 inches in the gravel. Due to capillarity, the lead bomb will contact with acid rain for a longer time, resulting in more dissolving leads. The loss of waste ammunition, the corrosion of the retaining wall, and the content of dissolved lead can be reduced by adding a layer of limestone or gravel at the bottom of the retaining wall to destroy the capillary phenomenon. Simultaneously, limestone can increase the pH of water, and lead dissolved in water can be precipitated.

4.1.2. Adding phosphate. This method is recommended if lead is widely distributed in the soil or likely to migrate to groundwater (such as low soil pH and shallow groundwater table). Different from lime, the main purpose of phosphate application is to combine with lead particles to form insoluble matter rather than adjusting soil pH. Moreover, the application of phosphate can effectively reduce the solubility of lead in soil because the phosphate of lead is extremely insoluble in water. This process also reduces the potential content of lead migrating outside or underground.

4.2. Control of surface water runoff
It is imperative to control soil erosion and surface water runoff to prevent the migration of lead. There are two factors affecting the content of lead in surface water: the amount of lead debris and runoff speed. Generally, the following measures can be adopted to control the water flow velocity: using plant and organic ground cover; implementing engineering control to reduce surface water runoff.

4.2.1. Surface coverage. (1) Plant cover. Vegetation can not only reduce the amount and time of rainwater contacting with lead bombs but also transfer and slow down surface runoff, contributing to preventing lead from migrating to the outside. Vegetation can impede the flow of water and reduce the amount of lead entering the soil through rain erosion. Fast-growing grass should be used to cover the lawn; plants attracting birds and other wild animals should be avoided to prevent the intake of lead by wild plants; vegetation should be used to drain the surface water out of the target area, such as planting at the bottom of retaining wall, so as to minimize the contact between water and lead bombs and reduce the migration of lead.

(2) Plastic film and organic fertilizer. Oxalic acid contained in plastic film and organic fertilizer is a kind of natural lead chelating agent, which can combine with lead in solution and reduce its mobility. These materials can cover any contaminated area or low-lying areas where runoff and lead may accumulate. Like the plant coating, the surface coating of organic materials is not waterproof. However, this method may not be appropriate if the pH value of the soil in the shooting range is low since these materials tend to be acidic especially in the process of decomposition.

4.2.2. Engineering measures. When shooting sites are located in areas with large annual rainfall, heavy rainfall will increase the risk of lead migration. In this case, strict engineering control is needed. The impact of rainfall is greater in hilly and slope areas or areas where the surface water is close to the shooting range.

(1) Filter bed. The filter bed is an outdoor engineering control facility, which is used to collect and filter the surface water in the shooting range. The collected surface water is transported to the filtration system to filter out large lead particles, improve the pH value of the water (reduce the possibility of further dissolution of lead), and discharge the water out of the area. However, lead bombs, debris, and large particles may remain in the soil. Thus, this method cannot completely prevent lead from entering the ground.

In a study, Hardison et al. [7] demonstrated that 1.5% of the total bullet mass remained on the surface of soil particles to form a layer of lead particles when the bullet passed through the retaining wall. Therefore, the filter bed should be built at the bottom of the bomb retaining wall. Thus can not only reduce the off-site migration of lead but also improve the pH of rainwater, weaken the dissolution of lead, and filter out fine lead particles from rainwater. The filter bed consists of two layers, with a
sand filter layer on the top and limestone, gravel or other neutralizing materials on the bottom. The collected water passes through the top sand filter and the neutralizing material to increase the pH value of the filtrate, and the lead particles in the rainwater are left in the sand filter layer. The filter bed is mainly used to intercept fine lead particles transported in surface water instead of filtering large particles and lead bombs. Therefore, the operation and maintenance cost of the filter bed is extremely low, limited to periodic cleaning of debris and replenishment of limestone. It is most effective to use the filter bed in the open and undulating terrain.

(2) Topography
The runoff rate can be slowed down by changing the drainage mode. In the area with a high pH value, the possibility of lead dissolution is low, which can ventilate the soil, increase the infiltration rate of precipitation, and hinder surface runoff and off-site migration. This design is actually to collect lead in the surface soil. Therefore, operation and maintenance should include lead recovery, soil pH adjustment, and phosphate application.

5. Removal and recovery of lead

5.1. Manual recovery screening
Screening bullet fragments by manual recovery is a low-tech and low-cost lead recovery method, which is usually conducted in topsoil. There are considerable lead bombs around the target and tree roots. It is necessary to manually collect the topsoil in the area, remove large fragments such as rocks, branches, and leaves, and then screen them. First, the large particles with a large size sieve are removed, and the lead pellet can pass through the sieve with this size. Then, the smaller size sieve is used to intercept the lead bullets and lead fragments, enabling the sand and other solids to pass through the sieve. The method of manual recovery of lead is suitable for small and low shooting ranges. In the process of operation, the recovery personnel should avoid contact with lead and wear appropriate protective equipment and respirator.

5.2. Mechanical separation and recovery

5.2.1. Vibration screening. Screening machine, also known as mobile vibrating screen, uses different sizes of stacked vibrating screen to screen the soil containing lead. The oversized soil blocks will be returned to the machine for re-screening to ensure that there is no lead in the fragments while the moist clay will combine to form larger soil blocks to block the screen, making it more difficult to recover lead from the moist soil. The use of mechanical devices to recover lead usually requires that there is no shrub vegetation in the area; grass, plastic film, and organic fertilizer will be removed or destroyed in the process of recovery. In the case that the vegetation cannot be completely removed, it is necessary to focus on the removal of plant debris and fallen leaves to better use the recycling machinery. The factors of lead recovery should be considered when the landscape vegetation of shooting ground is designed. For example, the larger the distance between trees is, the more conducive to lead recovery [8].

5.2.2. Vacuum screening. For hilly, rocky, and densely vegetated areas, a vacuum system can be used to collect lead bullets. This method is especially suitable for areas where lead bullets accumulate around trees, as well as dry sandy soil without a large amount of organic matter. Under the same conditions, the higher the soil moisture, the more difficult the lead recovery.

5.2.3. Soil leaching. Soil leaching is used to separate lead particles from the soil by a recovery device, and the soil is separated into gravel, sand, silt, and clay particles. Due to the high surface area and surface binding properties of clay, most of the lead pollutants adhere to clay particles. Soil leaching removes fine particles adhering to larger soil particles or transfers fine particles from larger soil particles to smaller soil particles, resulting in clean sand and gravel.
The soil is excavated from the contaminated area and mixed with a water-based detergent. Wet soil is separated by wet sieving or gravity separation technology. The advantage of this recovery technology is that soil drying is not required. Besides, soil leaching can remove up to 99% of lead particles by wet sieving and density separation.

6. Conclusion
According to the experience of lead pollution control in American outdoor shooting ground, the effective measures are determined as follows. (1) Improve soil pH. Lime can be applied to promote the combination of lead with soil and reduce the mobility of lead. (2) Reducing the contact between lead and water can be conducted by the approaches such as setting rainproof devices for bullet traps, planting plant cover, increasing filter bed, and changing topography. (3) Apply phosphate. It reacts with lead to form an insoluble substance and reduce the migration of lead. These methods are suitable for the daily maintenance measures of outdoor shooting ground, and cannot completely remove the lead in the soil. Moreover, they need to further cooperate with the recovery and screening measures. The lead in the soil can be completely removed by the methods such as artificial or mechanical separation device, vibration, vacuum, and leaching.

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