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Review

Lead (Pb) Contamination in Soil and Plants at Military Shooting Ranges and Its Mitigation Strategies: A Comprehensive Review

Zafer Alasmary

Soil Science Department, College of Food and Agricultural Sciences, King Saud University, P.O. Box 2460, Riyadh 11451, Saudi Arabia; zalasmari@ksu.edu.sa; Tel.: +966-50540-6503

Abstract: Heavy metals, especially lead (Pb), is the major cause of pollution in the military shooting range soils. Bullets, which are primarily made of Pb, are a substantial source of this pollution. On speciation, this Pb is distributed into its different metal forms. Different physicochemical properties of the soil like pH, moisture content, cation exchange capacity (CEC), and organic matter play a very crucial role in the distribution, transformation, and bioavailability of the Pb. The concentration of Pb found in different shooting ranges is examined. Moreover, bullet weathering and the availability of contaminants in the soil are influenced by the physicochemical properties of the soil. For the management of firing range pollution, a variety of strategies have been investigated, including soil washing, phytoremediation, and chemical stabilization. This review focus on the pollution status of different shooting ranges, the impact of the physicochemical properties of soil on the distribution, speciation, and transformation of Pb, and different mitigation strategies to control Pb pollution in military shooting ranges.

Keywords: shooting ranges; Pb; soil; physicochemical properties; Pb speciation; mitigation strategies

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Academic Editors: Guining Lu, Zenghui Diao, Yaoyu Zhou and Kaibo Huang

Received: 16 December 2024
Revised: 22 January 2025
Accepted: 24 January 2025
Published: 27 January 2025

Citation: Alasmary, Z. Lead (Pb) Contamination in Soil and Plants at Military Shooting Ranges and Its Mitigation Strategies: A Comprehensive Review. *Processes* **2025**, *13*, 345. https://doi.org/10.3390/pr13020345

1. Introduction

Heavy metals such as Pb, especially from shooting ranges, regularly contaminate the soil [1–3]. The average shot has 97% of Pb, 2% of antimony, 0.5% of arsenic, and sporadically, nickel [3,4]. The use of Pb in explosives has a long history because of its unique qualities. Compared with less dense materials, its high density gives projectiles more momentum and range. In addition, due to a low melting point of 327.4 °C, it is easy to mold into bullets and shoot. Its low cost, malleability, and corrosion resistance help to reduce gun barrel abrasion [5]. According to [6] bullets, which are primarily made of Pb, are a substantial source of this pollution. Pb emerges as the primary contaminant, with concentrations reaching up to 150 g kg^{-1} in the soils from military shooting ranges [2,3,7]. Due to the annual deposit of 60,000 tons of Pb present on shooting range grounds in the US, the United States Environmental Protection Agency has designated Pb residues in such soils as hazardous materials [8]. In particular, in Sweden, shotgun ammunition uses 500–600 tons of Pb yearly. In the USA, ammunition, shots, and bullets included approximately 75,100 and 73,900 tons of Pb, respectively, in 2011 and 2012 [9]. When fired, bullets and shots build up in shooting range soils where they can undergo a variety of changes, including oxidation and hydration, under the right circumstances [10]. Pb pollution concerns are highlighted by the startling 58,300 tons of Pb that are dumped into the soil annually in the USA alone [11]. The increased Pb values reported by many countries, including Sweden, England, Denmark, and Finland range from 10,000 mg/kg to 100,000 mg/kg [1].

Moreover, according to [1], Pb concentrations in the shooting range soils range from 11 to 1000 mg kg^{-1} and can even surpass 20,000 mg kg^{-1}. The current levels greatly surpass the regulatory limit of 300 mg Pb kg^{-1} soil set by the European Council Directive (1986) and beyond the overall screening requirement of 400 mg Pb kg^{-1} soil set by the United States. The number of military and nonmilitary firing ranges in the United States is estimated to be between 16,000 and 18,000, while the annual environmental release of Pb ammunition in Europe is estimated to be in the "tens of thousands" [11,12]. Meanwhile, it has been demonstrated that in shooting range soils, the solubility of secondary minerals that form a weathering crust around the corroding bullets limits the activity of Pb^{2+} in solution [3]. Some examples of these compounds are $PbCO_3$ (cerussite), $Pb_3(CO_3)_2(OH)_2$ (hydrocerussite), PbO (massicot or litharge), and to a lesser extent, $PbSO_4$ (anglesite), which can be found in soil, depending on the condition [13]. Typically, anglesite (Pb sulfate, $PbSO_4$), cerussite (Pb carbonate, $PbCO_3$), and minium (Pb (2^+, 4^+) oxide, Pb_3O_4) are produced by oxidizing galena near the surface with other minerals [7]. Pb sulfide, often known as galena (PbS), is a frequently found main component in sulfide ore deposits. Weathering induces the gradual oxidation of galena by air oxygen, resulting in the formation of either sulfate (anglesite) or carbonate (cerussite) through the chemical reaction [8] as described below:

$$PbS + 2O_2 \rightarrow PbSO_4 \text{ (Pb sulfate: anglesite)}$$

$$PbS + CO_2 + H_2O + 2O_2 \rightarrow PbCO_3 + H_2SO_4$$

Previous studies have shown that cerussite is usually formed at a pH higher than 6, and anglesite is formed at a pH lower than 6 [14,15]. Upon the dissolution of Pb from primary minerals, it has a tendency to either mix with carbonate or sulfate ions to produce insoluble Pb carbonate or Pb sulfate, or alternatively, it can be absorbed by ferric hydroxide [10]. Furthermore, soil organic matter, clay minerals, and manganese or iron oxides can significantly bind aqueous Pb^{2+} through adsorption mechanisms [11]. The use of Pb-containing ammunition on shooting ranges significantly increases the danger of Pb pollution in nearby agricultural areas [14]. Because of its insolubility and resistance to microbial degradation, Pb has a prolonged residence time in soils, which contributes to its persistence over an extended period of time. Pb in soil has a half-life of 740 to 5900 years. Pb background levels in soils typically vary between 10 and 30 mg/kg [16]. Meanwhile, Pb concentrations in soils have been observed to reach as high as 1104 mg/kg because of shooting practices [17]. It is possible that there are 1108 spent Pb bullets per hectare in the soil at shooting ranges. Pb concentrations in shooting range soils frequently exceed legal limits and pose a serious risk to human health [18]. The intake of poisoned plant products or exposure to Pb through contaminated soils can cause serious health problems and even death [18,19].

Elevated Pb levels can disrupt ecosystem functionality by adversely affecting essential soil organisms like microbes and nematodes, leading to impaired processes such as organic matter decomposition and nutrient cycling [20]. These metalloids of Pb undergo transformation from bullet fragments into more mobile forms and accumulate in a range of organisms, including earthworms, plants, and birds [21]. The extensive soil contamination observed across different shooting ranges underscores a significant ecological risk [22,23]. To assess the environmental impact of accumulated pollutants, bioassays, particularly short-term ones like seed germination and root elongation tests, are employed to evaluate their effects [23]. The guidelines of the Organization for Economic Cooperation and Development (OECD) recommend lettuce (*Lactuca sativa* L.) as a suitable test plant for chemical testing. Previous seed germination tests using *L. sativa* have indicated that elevated metalloid contamination in shooting range soil contributes to phytotoxicity [24]. Considerable environmental and health problems are associated with the use of Pb in munitions resulting in soil pollution at shooting ranges. Therefore, it is essential to comprehend how Pb behaves in soils, how it is absorbed by plants, and how it could affect the food chain to adopt efficient regulatory measures and lessen the negative effects of Pb pollution. This review focuses on synthesizing the current knowledge of the fate and ultimate risks presented by Pb associated with military training activities. Our focus will be on understanding and identifying trends in the literature regarding Pb metal toxicity in shooting ranges. We will primarily focus on its abiotic interactions with soil and its biotic transformation by plant uptake. This review is based on Pb derived from small-arms range bullets as the most likely source of contamination from military training. Objectives of the review paper are as follows:

- To provide a comprehensive assessment of the potential risk of Pb on the vegetation growing in shooting ranges
- To examine the effect of Pb on the physiochemical properties of soil

2. Accumulation and Distribution of Pb in Shooting Range Soils

Shooting ranges have gained notoriety as significant sources of Pb pollution in the environment, ranking second only to the battery industry in Pb emissions [25]. Recent research has intensified the focus on shooting ranges as Pb pollution culprits, particularly due to the escalating use of small-arms ammunition for shooting practices [2]. Once Pb from bullets and shots enters the soil, it undergoes various chemical and physical transformations that enhance its mobility and bioavailability, potentially leading to elevated Pb concentrations in shooting range soils [8]. Studies have consistently shown that shooting ranges accumulate Pb at concentrations significantly exceeding natural levels, which typically range from 10 to 30 mg/kg [26]. Notably, Pb pollution from ammunition is a global issue, with documented extreme Pb levels in shooting range soils worldwide [25]. For instance, levels as high as 97,600 mg kg^{-1} have been recorded in the US, 29,200 mg kg^{-1} in Japan, 66,961 mg kg^{-1} in Canada, 38,386 mg/kg in Botwana, 206,600 mg/kg in New Zealand, and 300,000 mg/kg in the Netherlands [2,27]. Recent years have shown an alarming increase in the buildup of Pb in shooting range soils, with some ranges registering concentration as high as 300,000 mg/kg, and this Pb accumulation is alarming [28]. Soil properties, shooting frequency, and the age of shooting ranges play significant roles in influencing Pb concentration levels, making comprehensive assessment and remediation strategies imperative [29]. The longer that spent bullet and shoot remain in shooting range soil, the higher the potential for the oxidation process to transform them into more soluble mineral forms, further exacerbating Pb mobility and bioavailability [25]. Moreover, it has been observed that rifle shooting ranges have a tendency to accumulate more Pb than pistol shooting ranges, mostly because rifle ammunition contains more Pb [3]. Table 1 shows the concentration of Pb in various shooting ranges of the world.

Table 1. Pb contamination in the soils of shooting areas across the different countries.

Location and Year of Study	Number of Studied Shooting Ranges	Concentration of Pb Found in Soil (mg/kg)	References
Switzerland (2010)	2	621 ± 12.35	[29]
Norway (2010)	1	22,000 ± 31.47	[30]
USA (2011)	3	20,350 ± 21.62	[31]
Canada (2011)	4	24,100 ± 78.54	[32]
South Korea (2012)	1	4625 ± 19.03	[33]

Table 1. *Cont.*

Location and Year of Study	Number of Studied Shooting Ranges	Concentration of Pb Found in Soil (mg/kg)	References
Australia (2012)	4	6403 ± 00	[2]
Finland (2012)	2	30,300 ± 113.48	[34]
South Korea (2013)	1	11,800 ± 52.11	[3]
Norway (2013)	5	30,000 ± 163.90	[35]
China (2014)	1	2563 ± 41.53	[36]
Netherland (2014)	1	2158 ± 55.10	[37]
Australia (2015)	3	30,600 ± 134.88	[38]
Finland (2015)	2	23,000 ± 105.49	[18]
China (2015)	3	6019 ± 45.18	[39]
Nigeria (2016)	1	17,500 ± 59.74	[40]
Australia (2016)	4	2145 ± 21.92	[41]
Spain (2016)	1	724 ± 12.47	[42]
Pakistan	1	1331 ± 19.04	[43]
South Korea (2017)	1	3436 ± 49.13	[44]
Norway (2017)	7	2700 ± 25.97	[32]
Nigeria (2017)	1	26,933 ± 00	[45]
Spain (2017)	1	710 ± 19.16	[46]
Switzerland (2017)	2	620 ± 10.41	[47]
Botswana (2017)	1	38,300 ± 158.51	[26]
Lithuania (2018)	1	654 ± 45.12	[48]
Alaska (2019)	1	2800 ± 27.49	[49]
Poland (2019)	1	3800 ± 14.03	[50]

Additionally, Pb accumulation and dispersion in shooting range soils are significantly influenced by soil characteristics, such as pH, organic matter (OM), CEC, soil type and moisture, etc. [51]. Meanwhile, it has also been suggested that the frequency of shooting activities and the age of the shooting range are key factors affecting Pb concentration in soils [26].

3. Factor Influencing Pb Dispersal

The speciation of Pb (distribution of its different metal forms) within shooting range soils is influenced by various factors, including soil properties such as pH, CEC and chemical processes like association–dissociation, oxidation–reduction reactions, precipitation–dissolution, sorption–desorption. These mechanisms collectively determine the mobility and bioavailability of Pb in soil; for example, soil pH affects Pb bioavailability, with lower pH leading to increased dissolution and mobility of Pb. CEC plays a crucial role, with higher CEC resulting in greater binding and retention of Pb in the soil. Additionally, chemical processes like sorption–desorption and precipitation–dissolution affect how Pb interacts with soil particles and compounds. Understanding these complex interactions is essential for managing Pb contamination in shooting range soil and assessing its environmental impact [52]. Moreover, both pH and EC are crucial factors which affect Pb speciation, but generally, pH has a dominant role in this speciation due to its link with wide-ranging effects on dissolution–precipitation reactions, adsorption–desorption processes, and bioaccessibility across different digestive environments [49,52].

4. Effects of Soil Physicochemical Properties in Accumulation of Pb

The physical and chemical properties of soil play a crucial role in influencing the distribution, mobility, solubility, bioavailability, bioaccessibility, and fate of Pb in shooting range soils. Notably, higher soil moisture levels, acidic soil pH, and increased soil organic matter create conducive environments for the weathering and conversion of metallic Pb projectiles into more reactive secondary Pb compounds. These factors significantly affect the behavior of Pb in shooting range environments [8].

4.1. Effect of Soil pH

Soil pH has a significant impact on solubility, mobility, and bioavailability of Pb in soils near shooting ranges, as supported by numerous studies [5]. According to [53] the presence of acidic soils in these environments creates a favorable environment for a number of processes linked to the corrosion, weathering, and alteration of Pb bullets and shoots. According to [54] a lower pH, which dissolves the crust of secondary minerals that forms on the surface of metallic Pb, speeds up the conversion of metallic Pb shoots and bullets into secondary minerals and exposes new metallic Pb surfaces to additional weathering [55]. Additionally, the acidic nature of soil having high moisture content can cause the dissolution of Pb minerals found on the surface of bullets, enabling the leaching of Pb into subsoils. Shooting ranges have acidic soil that range in pH from 5.4 to 6.4; for instance, it was found that Pb migrated from the surface soil to subsoils at a rate of 7.5% to 46%, compared to only 6% to 18% in a shooting range of alkaline pH 9.3. Additionally, acidic sandy soils at the surface show a lower affinity for heavy metals like Pb and frequently have unsaturated sorption sites for Pb due to the limited buildup of Pb on the surface of soil [55,56]. It is significant to remember that when hydrogen ions are consumed in the presence of oxygen, the corrosion of metallic Pb bullets and shoots within acidic soils can lead to an elevation in the pH of the nearby area [11].

In conclusion, the mobility and bioavailability of Pb in shooting range soils are significantly influenced by soil pH; while alkaline circumstances seem to increase Pb stability and retention in these environments, acidic ones tend to increase Pb mobility.

4.2. Effect of Soil CEC

A key factor in determining a soil's binding capacity is its soil CEC, which measures the number of exchangeable cations that a specific soil mass can adsorb. Pb has a lower mobility and bioavailability in soils with higher CEC due to their greater propensity to bind with Pb. Shooting ranges with high CEC have the potential to collect substantial Pb concentrations, despite the fact that this relationship is not universally applicable [8]. Cao et al. [57] compared two shooting ranges in Florida. A high CEC of 42.3 cmolkg^{-1} was found to be correlated with a total Pb concentration of 48,300 mg kg^{-1} at the MPR shooting range. While the CEC of 8.52 cmolkg^{-1} with a total Pb concentration of 12,710 mg kg^{-1} was found at the TRR shooting range in Florida. Additionally, a study by [58] showed that a rifle shooting range with a CEC of 10.75 cmol/kg had a total Pb concentration of 38,386 mg/kg, whereas a comparable range with a CEC of 8.00 cmol/kg showed a total Pb concentration of 25,193 mg/kg [59]. In investigations addressing Pb contamination at various shooting ranges, CEC values were found to vary from 1.05 to 50.1 meq 100 g^{-1}. The total Pb values varied accordingly, ranging from 5040 to 60,600 mg/kg [10]. It is crucial to stress that Pb mobility, bioavailability, and density are not solely controlled by soil CEC. The weathering and transformation of Pb are also influenced by organic matter and soil pH [2]. An Australian study that found variability in Pb contamination levels and CEC across various shooting ranges serves as an illustration of this intricacy.

4.3. Effect of Soil Moisture

Pb's weathering, mobility, transformation, and fate are significantly influenced by soil moisture, a fundamental physical feature of soil. The circumstances for Pb shots and bullets to corrode and erode are caused by soil moisture. Reactive gasses like oxygen and carbon dioxide can diffuse to the surface of the Pb, and moisture helps to generate a thin watery layer on the shoots' surface. As a result of this diffusion process, the weathering compounds of Pb such as Pb oxides, carbamates, phosphates, and sulfates are produced, which eventually leads to their solubilization and dissolution [26]. Liu et al. [53] conducted a study to determine the impact of moisture present in the soil, on weathering, and the transformation rate of metallic Pb powder. The research showed that during the course of seven days, the metallic Pb in X-ray diffraction (XRD) spectra vanished and was replaced by hydrocerussite, cerussite ($PbCO_3$), and massicot (PbO). The same results were seen in the field experiment where all metallic Pb was converted into hydrocerussite, cerussite ($PbCO_3$), and massicot (PbO). This shows that abraded Pb in shooting range soils experiences a rapid change into secondary Pb compounds. However, in contrast to the Pb powder used in the above experiment, a typical shoot may take many years to go through this transformation into secondary Pb minerals. The amount of moisture in the soil has a significant effect on how Pb shots turn into more reactive substances like cerussite. The surface of Pb shots develops a coating of cerussite when soil moisture is present, and this layer combines with water in watery circumstances to form hydrocerussite, as shown by the equation below:

$$3PbCO_3(s) + H_2O(l) \rightarrow 4Pb_3(CO_3)_2(OH)_2(s) + CO_2(g) \text{ (hydrocerussite)}$$

Additionally, the cerussite and hydrocerussite layers on the Pb shots and bullets dissolve and transform into other Pb compounds when soil moisture and other favorable soil conditions, such as soil pH, are present. This exposes the Pb shot surface to further weathering and transformation. Pure water does not erode the metallic Pb shoot or bullet, but it acts as a medium for the diffusion of oxidizing gasses to metal surface and causes the creation of secondary Pb compounds [60].

4.4. Effect of Soil Texture

The distribution of soil particles, like clay, silt, and sand within the mineral composition of soil is referred to as soil texture, and it is crucial in determining how Pb changes and ends up in shooting range soils [51]. Due to their large pores, sandy soils have higher infiltration rates and less water retention than clay soils, which result in varied moisture-holding capabilities, with clay soils having the highest capacity and sandy soils having the lowest. Additionally, compared to sandy and silt soils, clay soils often have higher organic matter (OM) contents [61,62]. The type and texture of the soil significantly influence the rate of the weathering of Pb bullet and shoot soil predominantly composed of sand in berms, and bullets incubated in sandy soils exhibited lower total Pb concentrations compared to clay soils due to slower weathering rates. By forming a thin layer on the surfaces of Pb bullets and shoots, allowing reactive gasses (CO_2 and O_2) to penetrate, corrosion is sped up and Pb weathering is aided. Pb bullets are further chemically weathered by soils with lower pH and more OM, which causes metallic Pb to change into ionic Pb [63].

In a study conducted by [64] the total Pb concentration present in sand berms was significantly lower than that in the soil berms as a result of slower weathering rates, which were attributed to a lower moisture retention capacity and a lower organic matter content, both of which are essential elements in the transformatiokon of metallic Pb into ionic Pb [65]. Pb shotgun pellets undergo speciation during periods of 6 to 25 years, with Pb shots or bullets in shooting range surroundings generating a protective layer of Pb compounds [49]. Reduced particle size, on the other hand, may lead to enhanced weathering rates of Pb powder, inhibiting the creation of a protective coating and promoting total conversion into secondary Pb minerals [66].

5. Environmental Impacts of Pb

Significant environmental and health issues have been highlighted by the accumulation of Pb in shooting ranges, which is a naturally occurring heavy metal that makes about 90–99% of the core composition of small-weapons projectiles [25]. Over the past 50 years, extensive research has been performed on Pb mobility and its density in shooting range soils [32]. The concentration of 23 and 80 g/kg of Pb can be found in these soils [32]. Pb is very pH-dependent when it comes to solubility, being most soluble at high pH values of about 10.5 [20]. Despite elevated Pb concentrations in the soils of shooting ranges, surface and pore waters in the vicinity generally maintain low Pb levels. This low Pb level is maintained due to circumneutral pH conditions, the formation of less soluble Pb compounds, and abundant surfaces for Pb adsorption [65,67]. Pb exposure carries significant health hazards, especially for young infants who are more susceptible to Pb absorption. Behavioral problems, difficulties with learning, kidney failure, hearing and vision impairment, stunted growth, brain damage, and hearing loss are health problems linked to Pb exposure [25]. Pb exposure in adults can lead to memory loss, high blood pressure, joint pain, nervous system problems, kidney and intestinal disorders, and pregnancy complications [68]. The effects of Pb in small-arms ammunition on groundwater and surface water are generally regarded as insignificant, with surface water contamination primarily happening during periods of heavy precipitation [69]. Several environmental parameters, such as soil characteristics, rainwater percolation, and co-contaminants, have an impact on Pb mobility [1]. Pb mobility is influenced by the bullet's composition, with older ammunition showing slower rates of corrosion and dissolution than newer Pb alloy ammunition. Pb species are mainly found in carbonate and oxide complexes, such as hydrocerussite, cerussite, litharge, and massicot, in shooting range soil. Based on the mineralogical circumstances, Pb can also attach to sulfur and phosphorus [70].

6. Mechanisms and Effects of Pb Toxicity in Plants

The accumulation of Pb in plants and crops close to shooting range is a serious issue since it could enter the food chain. High levels of Pb have been identified in a number of plant tissues, including Vetiver grass tissue (1390–1450 ppm/kg) and plant leaves (up to 70 mg/kg). Depending on the plant type and the manner of Pb absorption, plant roots can absorb Pb concentrations ranging from 1347.1 to 3825.8 mg kg^{-1} (dry weight) [20]. The concentration of Pb in the soil has a direct impact on the pH, electrical conductivity, and mineral composition of plants [71]. According to research, plants close to shooting ranges frequently exceed the limits advised by groups like the WHO and FAO [51]. Increased soil Pb levels can have a negative impact on soil quality and agricultural production. Even minute levels of Pb can interfere with a variety of biological processes, reducing plant development and agricultural yields. Reactive oxygen species (ROS) such as hydrogen peroxide (H_2O_2) can be produced as a result of Pb exposure. ROS can cause oxidative stress, enzyme inactivation, and lipid peroxidation, all of which lower agricultural productivity [72]. Malondialdehyde (MDA), a byproduct of lipid peroxidation that causes oxidative stress in plants under high-stress conditions, can also be produced by Pb exposure [73]. After contamination, effective remediation is necessary to return soil Pb levels to normal. Critical plant activities such as cell division, chlorophyll production, transpiration, root elongation, seedling growth, and seed germination are all negatively impacted by Pb [50]. Through interactions with metabolic enzymes and the displacement of necessary ions, Pb poisoning alters the permeability of cell membranes [73]. Additionally, Pb exposure causes an overproduction of ROS, which inhibits ATP synthesis, damages DNA, and exacerbates lipid peroxidation. To combat these effects and prevent against oxidative damage, plants have evolved defensive mechanisms, such as enzyme systems and non-enzymatic antioxidant systems [74]. Through passive ion exchange mechanisms, plants make it easier for Pb to be taken up from the soil. Then this Pb moved from roots to shoots. Different plant tissues accumulate Pb differently, with roots often having higher quantities than stems and shoots. Pb is largely transported from roots to shoots via the root apoplast; Pb absorption is influenced by parameters such as root surface area and mycorrhization. Strong Pb binding in lignified root tissues restricts Pb transport to plant tissues farther from the roots [50]. In some instances, Pb deposition on leaf surfaces can cause the total Pb concentration in plant leaves to be relatively high, which can affect the plants' capacity to absorb and accumulate Pb. Differential Pb accumulation in plant organs has effects on soil remediation techniques and raises the possibility of Pb re-entry into the food chain when herbivores consume polluted plant matter [20].

Plant Tolerance Against Pb Toxicity

Although it has no nutritional value and is not necessary for plant growth, Pb is naturally found in some plant species at concentrations that range between 2.1 and 2.5 mg/kg (dry weight) [20]. However, it has been determined that soil Pb values between 100 and 500 mg/kg are harmful to plants. Similarly, Pb concentrations in plant tissues between 30 and 300 mg/kg are regarded as hazardous and can have negative impacts on plant growth, photosynthesis, dry weight, and root development [75]. Significant growth loss in pine trees under active shooting range circumstances has been noted and is attributed to damaged roots and mycorrhizal fungi. After the shelling stopped, pine trees showed signs of recovery and even grew taller than the trees at the control site. Pb accumulation in plant roots can impede root growth by preventing root tip cell division [18]. Various plant species growing in Pb-polluted soil from shooting ranges have shown suppression of root elongation. Microtubule damage from high Pb concentration in plant roots can cause blockages in prometaphase cells. Pb exposure in plants can cause chlorosis and stunted growth, as well as other harmful effects on the physiology of the plant [50]. The physiological functions of hormones, electron transport, membrane structure, and water absorption are all inhibited by pb toxicity in plants. Because abscisic acid concentrations rise with elevated Pb levels, stomata close. Pb can interfere with the activities and tertiary structure of enzymes, especially those with thiol groups (-SHs) or carboxyl groups (-COOHs). By additionally blocking carboxyl groups (-COOHs) in enzymes, Pb ions in plant tissues can further decrease enzyme function [76]. Reduced chlorophyll synthesis, interference with the uptake of vital nutrients like magnesium and iron, decreased photosynthesis rates due to chloroplast and chlorophyll degradation, and Pb-induced stomatal closure, which restricts the amount of carbon dioxide (CO_2) available, are all effects of Pb toxicity in plant leaves [76]. Pb has the ability to impair respiratory procedures and lessen the formation of adenosine 5′-triphosphate (ATP). Additionally, it may prevent cations and nitrate ions from entering plant roots, which would interfere with nutrient intake and nitrogen metabolism [25]. Pb exposure results in elevated reactive oxygen species (ROS) levels in plant tissues, which leads to oxidative stress and damages cell membranes and causes lipid peroxidation [76]. Despite the toxicity of Pb, certain plants have different strategies for tolerating high Pb accumulation. These plants have the option of using the "accumulator" or "excluder" approaches. By precipitating Pb into less harmful forms or tying it to the carboxylate groups (COOHs) of uronic acid, excluder plants keep low, non-toxic Pb concentrations in their tissues, preventing roots from absorbing it. In order to reduce Pb toxicity, accumulator plants aggressively concentrate Pb in their tissues at a wide range of soil concentrations. These resilient plants are essential for phytoremediation, which helps to regulate and control Pb pollution in shooting ranges [6]. Depending on their particular physiological adaptations, they can either accumulate or exclude Pb. For example, *Equisetum arvense* displays excluder characteristics by maintaining low Pb concentrations in plant tissues even at high soil Pb levels. Other plants, such as *Lactuca sativa*, tolerate Pb toxicity due to high organic matter content that complexes Pb and reduces its toxicity. These plants play a vital role in mitigating Pb pollution in shooting ranges through phytoremediation efforts [77].

7. Mitigation Strategies for Pb-Impacted Soil in Military Shooting Ranges

7.1. Stabilization

In order to reduce Pb's solubility, bioavailability, and mobility, stabilization is a frequently used remediation strategy for Pb-contaminated soil [78]. Due to its affordability, environmental friendliness, adaptability, simplicity, and effectiveness, this technology has attracted a lot of attention in real-world applications [79]. As stabilizing agents, a variety of substances, such as fly ash, cement, blast furnace slag, phosphate rock, red mud, and steel are frequently used [80]. The operation of cement-based stabilization through precipitation and physical and chemical fixing mechanisms has drawn particular interest for Pb-contaminated soils [81]. It is renowned for its high energy usage and carbon footprint [82]. As potential substitutes for immobilizing Pb-contaminated soil, phosphate-based materials like phosphoric acid and phosphate rock have demonstrated their potential. These have the capacity to transform Pb into stable forms like pyromorphite [$Pb_5(PO_4)_3(OH, Cl, F)$] [82]. Phosphate rock encourages the development of phosphate minerals in Pb-contaminated soil, greatly lowering Pb's water solubility, phytoavailability, and bioaccessibility [83]. Phosphoric acid can produce molecules that resemble chloropyromorphite, which can further reduce the solubility and bioavailability of Pb in highly contaminated soil when paired with KCl as a source of chloride ions. When utilizing phosphate-based chemicals to increase the solubility of phosphate and Pb-related phases, an acidic environment is frequently necessary [20].

According to research, Pb solubility in soil can be significantly decreased by simultaneously applying compounds that are both acidic and phosphate based. According to Li et al. [77] phosphate rock activated with oxalic acid (OA) demonstrated better Pb conversion from non-residual to residual fractions, hence increasing its stabilization efficiency. Additionally, according to XRD analysis, a phosphate- binder made of monopotassium phosphate, reactive magnesia, and OA-activated phosphate rock effectively reduced Pb leachability from polluted soil by forming stable Pb-related precipitates like fluoropyromorphite ($Pb_5(PO_4)_3F$) [84].

7.2. Phytoremediation

Researchers have focused on practical and long-lasting solutions in their search for environmentally friendly methods to reduce Pb pollution in shooting range soils. A highly effective and environmentally beneficial method for cleaning up severely damaged soils is phytoremediation [68]. In order to handle a variety of environmental contaminants, like trace metals, heavy metals, organic chemicals, and radioactive substances, phytoremediation strategically employs modified green plants. The purpose of this strategy is to make it easier to remove, immobilize, contain, and stabilize these contaminants inside the soil matrix [85]. The goal of phytoremediation is to improve soil by enhancing the uptake and transport of contaminants through plants through a variety of plant-driven mechanisms, including chemical, biological, and physical processes. Phytostabilization, phytoextraction, phytovolatilization, and rhizofiltration are important phytoremediation methods [68,85]. Pb is immobilized in the soil by plants through a process known as phytostabilization [2]. This is accomplished by Pb being absorbed and precipitated in the root matrix, which effectively reduces its mobility within the soil matrix. Phytoextraction is the process in which plant roots actively absorb the Pb from the soil, and then translocate it into the above-ground biomass [68]. Pb is taken up by plants and then released as secondary Pb products by transpiring in the atmosphere, and the process is known as phytovolatilization. This process is particularly prominent in plants that are actively growing because they release Pb through transpiration through their leaves after absorbing it along with water [68].

Recent research has demonstrated how phytoremediation may successfully treat shooting range soils. For instance, Dinake et al. [15] found that Pisum sativum had extraordinary phytoextraction capabilities, attaining an impressive 96.23% Pb removal efficiency from shooting range soil contaminated with more than 1331 mg/kg of Pb. *Agrostis capillaris* displayed efficient phytoremediation in a research study by Tariq et al. [42] on a Spanish shooting range by absorbing 1107 mg/kg of Pb through its roots and translocating roughly 135 mg/kg into its shoots. Grass pea plants (*Lathyrus sativus* L.) are also used for phytoremediation. Similar findings were made in the United Kingdom by Sneddon et al. [86] who discovered that *Lolium perenne*, a perennial ryegrass, absorbed 38 mg/kg of Pb into its shoots when it was growing in soil contaminated with Pb at concentrations ranging from 43.89 to 159.98 mg/kg of Pb. Compared to other soil remediation methods, including chemical and physical amendments, phytoremediation has clear advantages. By preserving soil structures and habitats for living things, it minimizes the environmental impact compared to other soil removal techniques. It is also economical, as it does not introduce foreign chemicals into the ecosystem [20]. Additionally, plants utilized in phytoremediation assist in reducing the effects of rainfall runoff and soil erosion by stabilizing the soil through their root systems. The release of chemicals into the soil by plant roots served as nutrition for bacteria in the rhizosphere emphasizes the symbiotic relationship between plants and soil microbial communities. In comparison to areas farther from plant roots, this area has a higher microbial density [87]. The United States Environmental Protection Agency (USEPA) promotes phytoremediation as a practical strategy for reducing and managing Pb pollution in shooting ranges due to its wide range of advantages [68]. Further advantages and disadvantages are mentioned in Table 2.

Table 2. A brief comparison of the Pb mitigation strategies at shooting ranges.

Mitigation Strategy	Descriptions	Advantage	Disadvantage	Reference
Chemical stabilization	This process involves the addition of stabilizing agents, such as phosphates, silicates, or carbonates, to the contaminated soil. These agents react with the Pb ions to form stable, insoluble compounds which are less mobile and less bioavailable. Generally, precipitation, adsorption, and complexation mechanisms are involved in this process.	• Cost effective • In Situ treatment • Rapid treatment • Long-term stability • Cost effective	• Soil specific • Can deteriorate soil quality • Potential for rebound	[88]
Phytoremediation	Phytoremediation involves the use of specific plants known as hyperaccumulators, which have the ability to absorb and accumulate heavy metals like Pb from the soil through their roots. The Pb is then translocated to the above-ground parts of the plant, where it can be harvested and removed. Overall, phytostabilization, phytoextraction, and rhizodegradation are the common mechanisms which are used in phytoremediation.	• Environmentally friendly • Cost effective • Long-term sustainability	• Limited to certain contaminants • Time consuming • Plant specific	[89]

Table 2. *Cont.*

Mitigation Strategy	Descriptions	Advantage	Disadvantage	Reference
Microbial bioremediation	Microorganisms, such as bacteria and fungi, are utilized to degrade, transform, or immobilize environmental pollutants, including heavy metals like Pb. Biosorption, bioaccumulation, bioprecipitation, and biovolatilization are the common mechanisms which are used in microbial bioremediation.	• Cost effective • Environmentally friendly • On-site treatment • Low environmental impact	• Limited effectiveness • Site specific • Time consuming • Limited to biodegradable contaminants	[90]
Soil washing	It is an ex situ remediation technique used to remove contaminants, such as heavy metals, from soil. Overall, soil excavation, physical separation, chemical extraction, separation, and washwater treatment are the general steps which are followed in this practice.	• Highly effective • Reduce the volume of contaminated soil • Adjustable according to soil type	• Energy consumption • Soil disruption • Residual contamination	[91]

7.3. Microbial Bioremediation

In combating Pb toxicity, bioremediation using bacterial community plays an important role. In addressing Pb toxicity, bioaugmentation stands out as a widely studied technique. Bioaugmentation involves the deliberate introduction of specific microbial communities to augment the native microbial populations at contaminated sites, thereby enhancing the degradation of contaminants or toxic substance [92]. Native microbes present at contaminated sites exist in limited quantities and are often insufficient for effective contamination reduction. Bioaugmentation addresses this limitation by introducing a substantial number of targeted microbes to facilitate comprehensive biodegradation. Microbial methods can effectively remediate soil contaminated with heavy metals. For instance, the bacterium *Rhodobacter sphaeroides* contributes to altering the speciation of Pb, converting it into inert forms with reduced bioavailability. Achal et al. explored the mechanisms behind Pb absorption by the bacterium *Leclercia adecarboxylata*, highlighting the role of extracellular polymeric substances (EPS) produced by these bacteria in binding with Pb, preventing its entry into the cell [93]. *Kocuria flava* employs calcite formation via urease, a prominent enzyme, to chelate Pb effectively, reducing its availability [94]. Bacteria, including *Kocuria flava*, are vital in Pb bioremediation due to their ability to sequester Pb, modify its oxidation state, or induce precipitation [95]. *Pseudomonas aeruginosa* has ability to reduce Pb concentrations in soil, emphasizing the potential of bacteria in Pb-contaminated soil remediation [96]. Microalgae like *Phormidium ambiguum* and *Scenedesmus quadricauda* (Chlorophyta) have been utilized for Pb removal from freshwater [97]. Fungi offer an alternative approach to mitigate Pb stress, employing distinct detoxification mechanisms compared to other organisms [98]. These mechanisms encompass intracellular processes involving polyphosphates, sulfur compounds, peptides, organic acids, and intracellular compartmentalization, as well as extracellular mechanisms like precipitation, chelation, and cell wall binding, which play a pivotal role in metal detoxification [99].

7.4. Chemical-Assisted Soil Washing

Chemical-assisted soil washing is a remediation that effectively removes particular contaminants from soil by using a variety of chemical agents, such as acids, bases, salts, chelators, and surfactants [94]. Chelators have shown significant potential in removing harmful metals from polluted soil during the washing process, among various chemical agents. Chelators can remove target pollutants from the soil surface by forming stable and soluble metal–chelator complexes [95]. Due to the durability of metal–EDTA complexes, ethylenediaminetetraacetic acid (EDTA) has traditionally been a preferred chelator, especially for remediating Pb-contaminated soils [57,98]. However, issues have been raised regarding EDTA's poor biodegradability and prolonged persistence in soils, which could result in secondary pollution and adverse effects on the ecosystem [57,96]. In consequence of these issues, biodegradable chelators are becoming more popular as EDTA substitutes for the remediation of metal-contaminated soil [88,89,97]. It is crucial, but unexplored, to compare the efficacy of biodegradable chelators to EDTA for remediating Pb-contaminated shooting range soil. Chelator-assisted soil washing successfully removes metal pollutants, but it may interfere with the binding of residual metals with the soil components, thus increasing their mobility and leaching [73,99].

Combining chemical immobilization with chelator-assisted washing offers a workable solution to these problems. Through the effective extraction of labile Pb from contaminated soil, the solubility of the remaining Pb in the washed-soil residue is subsequently decreased. This approach has been successful with biodegradable chelators like EDDS, GLDA, and HIDS, which offer a more sustainable option to persistent chelators like EDTA. Following chelator-assisted washing, residual Pb is only slightly mobile after a two-step post-treatment involving $FeCl_3$ and a CaO solution. A potential remediation solution for Pb-contaminated shooting range soil is provided by this combined approach [29].

8. Planned Future Use of Shooting Range Site

The planned future use of a shooting range site plays a critical role in shaping the Pb mitigation strategy for several reasons. This future use impacts risk assessment by setting different cleanup standards; for example, sites intended for residential or recreational purposes need more stringent decontamination compared to those for industrial use. The objectives of remediation are directly influenced by what the land will become, with residential areas requiring a deeper clean than commercial spaces [67].

The economic aspect of remediation is also tied to future use, where more intensive cleanup might be justified for high-value land. Regulatory compliance is another factor; understanding the end use helps in adhering to specific environmental laws [95]. Moreover, community acceptance can vary; for instance, methods like phytoremediation might be preferable for future parks but less so for industrial zones [33].

The sustainability of the mitigation method chosen must align with the intended use, such as using permanent vegetative caps for nature reserves but not for sites where construction is planned. Future use also dictates the need for ongoing monitoring post-remediation to ensure safety. Some remediation techniques could restrict future development options, like soil replacement, which might limit underground construction. The environmental impact of the site after cleanup is considered, with priority given to ecological restoration for conservation areas [90]. Lastly, funding for remediation is often directed toward sites with the most significant potential impact or health risks, based on their planned future use [90]. In summary, by considering the future use of a shooting range site in the planning of Pb mitigation, it can be stated that cleanup efforts are both effective and sustainable, tailored to the site's ultimate purpose, thereby optimizing resource use and minimizing environmental risks in the long term.

9. Conclusions and Future Perspectives

In summary, the presence of heavy metals, particularly Pb, in military shooting range soils represents a significant and concerning source of environmental pollution. Pb-based bullets are a primary contributor to this contamination. Through speciation analysis, it has become evident that Pb can exist in various chemical forms within these soils, and specific physicochemical soil properties, including pH, moisture content, CEC, and organic matter, heavily influence its distribution, transformation, and bioavailability. The strategies discussed include Pb recovery and recycling initiatives, soil stabilization techniques, routine soil monitoring to assess contamination levels, THE proper disposal of Pb-contaminated materials, and adopting Pb-free ammunition. It is crucial to acknowledge that addressing the pollution status of military shooting ranges is imperative, as the impact of soil physicochemical properties on Pb distribution, speciation, and transformation underscores the need for site-specific solutions. Current mitigation strategies involve containment methods like bullet traps and backstops to collect spent Pb, alongside substitution with non-Pb alternatives like steel or bismuth, though these have met resistance due to cost and performance issues. Legislative efforts and educational programs are also incrementally promoting the reduction in Pb use. Looking forward, innovative approaches are anticipated to include fully biodegradable or encapsulated ammunition to prevent Pb leaching, and advanced remediation techniques like bioremediation and nanotechnology to clean up existing contamination. The development of smart systems for the real-time monitoring of Pb levels and policy enhancements toward creating Pb-free zones in ecologically sensitive areas are also on the horizon.

However, challenges persist, including economic barriers to adopting more expensive non-Pb ammunition and cultural resistance from traditional shooting enthusiasts. Opportunities for change arise from technological advancements that could lower costs, increased public health awareness, and collaborative efforts among scientists, policymakers, and the shooting community. Future research should focus on long-term efficacy studies of new mitigation technologies and understanding the ecological impacts of alternative materials. The integration of AI and IoT for smarter environmental management, along with comprehensive predictive models for Pb dispersion, could revolutionize how shooting ranges operate, ensuring they align with both ecological and public health standards. Ultimately, the goal is to mitigate the legacy of Pb pollution, fostering environments that are safer for both human communities and wildlife.

Funding: This research received no external funding.

Data Availability Statement: Not applicable.

Acknowledgments: The author would like to express his heartfelt gratitude to the Deanship of Scientific Research at King Saud University for its generous support and encouragement of scientific research and researchers. He also greatly appreciates the editors' and anonymous reviewers' insightful comments and recommendations.

Conflicts of Interest: The author declares that he has no personal relationships or competing financial interests that could have appeared to influence the work.

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