

Review

Recent Trends in Sustainable Remediation of Pb-Contaminated Shooting Range Soils: Rethinking Waste Management within a Circular Economy

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Abstract: Soil metal contamination in recreational shooting ranges represents a widespread environmental problem. Lead (Pb) is the primary component of traditional ammunition, followed by metalloids such as antimony (Sb) and arsenic (As). Lead-based bullets and pellets deposited on the soil surface are subject to steady weathering; hence, metal(loid)s are released and accumulated in the underlying soil, with potential adverse consequences for ecosystem function and human health. Amongst the currently available environmentally-safe technologies for the remediation of metal-contaminated soils, chemical immobilization is recognized as the most practical and cost-effective one. This technology often uses inorganic and organic amendments to reduce metal mobility, bioavailability and toxicity (environmental benefits). Likewise, amendments may also promote and speed up the re-establishment of vegetation on metal-affected soils, thus facilitating the conversion of abandoned shooting ranges into public green spaces (social benefit). In line with this, the circular economy paradigm calls for a more sustainable waste management, for instance, by recycling and reusing by-products and wastes in an attempt to reduce the demand for raw materials (economic benefit). The objective of this manuscript is to present a state-of-the-art review of the different industrial and agro-food by-products and wastes used for the remediation of metal-contaminated shooting range soils.



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1. Introduction

The environmental impact of metal contamination from legacy shooting activities is of growing concern in modern society. Many studies have evidenced that outdoor shooting ranges used for military training or sport can constitute a significant source of metal contamination [1–4], with unintended consequences for ecosystem and human health [5–7]. In shooting ranges, soil metal contamination occurs primarily through the use of lead-core bullets and pellets. Regular ammunition used in shooting ranges typically contains lead (Pb; >90% in weight), metalloids such as antimony (Sb; 2–7%) and arsenic (As; 0.5–2%) as well as varying amounts of nickel (Ni), zinc (Zn) and copper (Cu) [2,8,9]. Although so-called environmentally friendly (“green”) ammunition, made of tungsten and nylon, has been recently introduced, large amounts of Pb-containing bullets are still used due to their high ballistic coefficient [10].

Following shooting activity, Pb bullet fragments and pellets are spread over the shooting range and its vicinity [11]. In the United States, 60,000 tonnes of Pb are deposited annually onto shooting range soils [9]. In Finland, Sweden and Switzerland, it is estimated

that the total annual Pb load in site-specific shooting ranges is about 500 tonnes. Very often, after the abandonment of a shooting range, pellets are left on the soil, and then, as a result of continuous weathering and corrosion of ammunition residues, total Pb concentrations in surface soils of shooting ranges frequently rise above 1000 mg kg⁻¹ (and often well over 10,000 mg kg⁻¹) [11–16]. These values are far higher than the regulatory limit of 300 mg Pb kg⁻¹ soil as prescribed in the European Council Directive (1986) [17] and also exceed the total screening criterion of 400 mg Pb kg⁻¹ soil as established by the U.S. Environmental Protection Agency (US EPA, 1996) [18], which provide reference points for establishing remediation targets. On the other hand, quantitative risk assessment (QRA) is an essential tool for the estimation of environmental and human risks associated with soil contamination, which helps to make appropriate decisions for soil remediation.

The physical elimination of bullet fragments and pellets from soil includes separation techniques or soil excavation and landfill disposal [19]. These traditional practices are very costly and environmentally destructive [20–22], and therefore are not feasible on a large scale [23,24]. Heavily contaminated soils (>2000 mg Pb kg⁻¹) from inactive or abandoned shooting ranges are usually landfilled or processed by chemical washing, whereas for moderately contaminated soils (<2000 mg Pb kg⁻¹), cost-effective remediation strategies are often sought [24]. Metal soil remediation strategies can be aimed at (i) stabilizing/immobilizing the metal (strategy: immobilization) or (ii) reducing its concentration (strategy: mobilization and then removal) [19]. The immobilization strategy, which can include chemical immobilization and/or phytostabilization [25–27], seeks to reduce metal bioavailability. Many remediation initiatives dealing with diffuse contamination by metals with reduced mobility, such as Pb, are based on this strategy. Mobilization, on the other hand, involves the addition of a chemical agent to the contaminated soil in order to increase metal bioavailability and mobility for its subsequent elimination/extraction from the soil. Mobilization-based techniques include soil washing and assisted phytoextraction [27,28]. This latter strategy is commonly implemented when dealing with discrete, localized metal contamination.

Phytoremediation (via phytostabilization or phytoextraction) is an affordable and environmentally friendly technology for soil remediation and is applicable when metals are located in the rhizosphere [29,30]. However, the in situ phytoremediation of shooting range soils remains poorly documented [31]. Wilde et al. (2005) [30] found that the combined use of vetiver grass (*Chrysopogon zizanioides*) and a fertilizer reduced plant uptake of soil Pb, thus decreasing the associated environmental risks. Rodríguez-Seijo et al. (2016) [10] reported the remarkable Pb-phytostabilization capacity of *Agrostis capillaris* plants when grown in shooting range soil. Tariq and Ashraf (2016) [32] showed that pea (*Pisum sativum*) and sunflower (*Helianthus annuus*) plants efficiently removed Pb from a shooting range soil contaminated with Cd, Cu, Co, Ni, Cr and Pb. Nonetheless, phytoremediation (mainly phytoextraction) generates metal-contaminated plant biomass, which requires further treatment for safe disposal, recycling or reuse. Interestingly, the combination of metal phytoextraction with ecocatalysis, an innovative biomass processing technology [33,34], allows the use of the metal-laden plant biomass, resulting from phytoextraction, as an effective industrial ecocatalyst, thus enhancing the financial viability of such phytotechnology [35].

The intended alteration of metal (Pb) bioavailability by the addition of chemical amendments represents an effective means of mitigating environmental risks without the high costs usually associated with unsustainable traditional remediation strategies [28]. Contaminated shooting range soils have been mainly treated with a variety of inorganic amendments (Table 1), which can be either synthetic or mined materials, such as phosphates [36–42], lime-based compounds [43,44], and metal oxides [24,37,45], in order to induce specific reactions within the soil components to render Pb inert without substantially affecting soil properties [28]. The chemical reactions involved in Pb immobilization by the abovementioned amendments are summarized in Table 2.

Table 1. Inorganic materials used as amendments for Pb immobilization in shooting range soils.

Type of Soil Amendment	Material	Reference	Type of Study (Duration)	Treatment Rates	Range of Soil [Total Pb] * (mg kg ⁻¹)
Phosphate amendments	Tricalcium phosphate (Ca ₃ (PO ₄) ₂), hydroxyapatite, rock phosphate and potassium dihydrogen phosphate (KH ₂ PO ₄)	Park et al. (2011) [46]	Lab (14 d)	200 mg P kg ⁻¹ soil	346
	Soft rock phosphate and phosphoric acid	Sanderson et al. (2015) [38] Sanderson et al. (2014) [47]	Lab (12 mth)	1:1 proportion P/Pb molar ratio of 5:1	399–10,403 2330–12,167
	Commercial phosphate-based amendment	Sanderson et al. (2015) [38] Sanderson et al. (2014) [47]	Lab (12 mth)	0.5, 1 and 2% (w/w) P/Pb molar ratio of 5:1	399–10,403 2330–12,167
	Phosphoric acid	Sanderson et al. (2015) [37]	Lab (1 mth)	1% (w/w)	446–8409
	Monocalcium phosphate (Ca(H ₂ PO ₄) ₂ ·H ₂ O)	Cao et al. (2008) [43] Sanderson et al. (2016) [48]	Lab (28 d) Lab (7 d)	2.5, 5.0, 7.5, and 10% (w/w) 2% (w/w)	2500–35,868 177–2545
	Calcium phosphate (Ca ₃ (PO ₄) ₂) nanoparticles	Arenas-Lagos et al. (2016) [39]	Lab (10 d)	5% (w/w)	223–6309
	Diammonium phosphate, phosphate rocks	Seshadri et al. (2017) [40]	Lab (30 d)	600 mg P kg ⁻¹ DW soil	9876
	Phosphate rock and phosphoric acid	Fayiga and Saha (2017) [41]	Lab (18 h)	P/Pb molar ratio of 4:1	739–8014
	Tricalcium phosphate (Ca ₃ (PO ₄) ₂) nanoparticles	Rodríguez-Seijo et al. (2020) [42]	Lab (10 d)	5% (w/w)	507–4452
Lime-based amendments	Calcium oxide (CaO) * Commonly known as quicklime	Cao et al. (2008) [43]	Lab (28 d)	2.5, 5.0, 7.5, and 10% (w/w)	2500–35,868
	Calcium hydroxide (Ca(OH) ₂) * * Commonly known as slaked lime	Conesa et al. (2012) [44]	Lab (14 d)	5.5 g Ca(OH) ₂ kg ⁻¹ DW soil	620
	Calcium carbonate (CaCO ₃) * * Commonly known as lime	Sanderson et al. (2015) [38]	Lab (12 mth)	0.5, 1, and 2% (w/w)	399–10,403
		Sanderson et al. (2014) [47]		5% (w/w)	2330–12,167
	Levonmäki and Hartikainen (2007) [49]	Lab (21 d)	5 t ha ⁻¹	22,600	

Table 1. Cont.

Type of Soil Amendment	Material	Reference	Type of Study (Duration)	Treatment Rates	Range of Soil [Total Pb] * (mg kg ⁻¹)
Metal oxide-based amendments	Natural iron oxide (FeO), gibbsite (GINP), and silver nanomaterial (AgNP)	Rajapaksha et al. (2015) [24]	Lab (12 mth)	0.1% (w/w)	17,468
	Magnesium oxide (MgO)	Sanderson et al. (2015) [37]	Lab (1 mth)	10% (w/w)	446–8409
		Sanderson et al. (2015) [38]	Lab (12 mth)	0.5, 1, and 2% (w/w)	399–10,403
		Sanderson et al. (2014) [47]	Lab (12 mth)	2% (w/w)	2330–12,167
Iron (III) oxide (Fe ₂ O ₃) nanoparticles	Rodríguez-Seijo et al. (2020) [42]	Lab (10 d)	5% (w/w)	507–4452	
Iron grit (Fe ⁰), ferric oxyhydroxide (FeHO ₂) mixed with limestone	Okkenhaug et al. (2016) [45]	Field (4 y)	2 and 4%	356–1112	

* Range of soil total Pb concentration (mg kg⁻¹) min-max averages.

Table 2. Chemical reactions involved in the immobilization mechanisms of Pb^{2+} [13,37,38,50,51].

Material	Chemical Equation
Hydroxyapatite	$Ca_{10}(PO_4)_6(OH)_2(s) + 14H^+ \rightarrow 10Ca^{2+} + 6H_2PO_4^- + 2H_2O$ (dissolution of hydroxyapatite) $10Pb^{2+} + 6H_2PO_4^- + 2H_2O \rightarrow Pb_{10}(PO_4)_6(OH)_2(s) + 14H^+$ (precipitation of pyromorphite)
Phosphoric acid	$5Pb^{2+} + 3H_2PO_4^- + Cl^- \rightarrow Pb_5(PO_4)_3Cl(s) + 6H^+$
Phosphate rock	$Ca_{10}(PO_4)_6F_2 + 12H^+ \rightarrow 10Ca^{2+} + 6H_2PO_4^- + 2F^-$ (dissolution of phosphate rock) $10Pb^{2+} + 6H_2PO_4^- + 2F^- \rightarrow Pb_{10}(PO_4)_6F_2 + 12H^+$ (precipitation of fluoropyromorphite)
Diammonium phosphate	$(NH_4)_2HPO_4 + Ca^{2+} + 2H_2O \rightarrow CaHPO_4 \cdot 2H_2O + 2NH_4^+$ $4CaHPO_4 \cdot 2H_2O + H_2O \rightarrow Ca_4H(PO_4)_3 \cdot 3H_2O + H_3PO_4$ (phosphoric acid)
Magnesium oxide	$MgO + H_2O \rightarrow Mg(OH)_2$; $Mg(OH)_2 + H_2O \rightarrow Mg^{2+} + 2OH^-$ $Pb^{2+} + 2OH^- \rightarrow Pb(OH)_2$ $MgO + Pb^{2+} \rightarrow PbO + Mg^{2+}$
Calcium carbonate	$CaCO_3(s) + H_2O \rightarrow Ca^{2+} + CO_2(g) + 2OH^-$ $CaCO_3 + 2H_2O + Pb^{2+} \rightarrow Pb(CO_3)(\text{cerussite}) + Ca(OH)_2 + 2H^+$ $3Pb(CO_3) + 2H_2O \rightarrow Pb_3(CO_3)_2(OH)_2(s)$ (hydrocerussite) + H_2CO_3
Calcium oxide	$CaO(s) + H_2O \rightarrow Ca(OH)_2 \rightarrow Ca^{2+} + 2OH^-$ $Pb^{2+} + 2OH^- \rightarrow Pb(OH)_2$ $CaO + Pb^{2+} \rightarrow PbO(s)$ (litharge) + Ca^{2+} $PbO + CO_2(g) + 2H_2O \rightarrow PbCO_3(s)$ (cerussite) + $2OH^- + H_2$

More recently, industrial and agro-food by-products and wastes are being considered as soil ameliorants or amendments. According to the European Waste Framework Directive (WFD) (2008) [52], “waste” is any substance or object that the holder discards or intends or is required to discard and “by-product”, in contrast, is a material that is not deliberately generated in a production process, which may or may not be a waste and which can be used directly without any further processing other than normal industrial practice.

As a result of human economic activities, huge quantities of these materials are constantly being produced. In the absence of proper management, these materials can become a serious environmental problem. Many governments and industries throughout the world are becoming increasingly interested in diverting by-products and wastes from landfills, and instead are adopting the “reduce, reuse, repair, recycle” hierarchy of waste management. This has more recently been included in the principles of the European Circular Economy Strategy [53] as well as in the Sustainable Development Goals of the 2030 Agenda for Sustainable Development of the United Nations [54] Goal No. 12: “Responsible consumption and production”. The circular economy is considered to be an umbrella concept for an economy that fosters the sustainable and cyclical use of resources, thus minimizing waste generation [55]. More sustainable resource and waste management is also a strategic target of the European Green Deal [56–58].

The purpose of the present paper is to provide an overview of recent industrial and agro-food by-products and wastes that are being used as amendments for the remediation of Pb-contaminated shooting range soils, within the context of the circular economy paradigm. Likewise, it aims to assist decision-makers involved in the remediation of contaminated shooting ranges according to circular economy values. Finally, as an example, an experiment focused on the use of agro-food by-products and wastes as amendments for the remediation of a Pb-contaminated shooting range soil is presented. The main objective of this experiment was to evaluate the effectiveness of such amendments in terms of the reduction of soil extractable Pb under both laboratory and field conditions.

2. From Wastes to Resources: By-Products and Wastes as Pb-Immobilizing Agents in Shooting Range Soils

Heavy metal lead (Pb) ranks second, after As, on the priority lists of Hazardous Substances of the Agency for Toxic Substances and Disease Registry (ATSDR) and the US Environmental Protection Agency (US EPA) due to its widespread occurrence and, above all, its potential high ecotoxicity [13]. Lead is also recognized as a chemical of great concern in the European REACH Regulation (EC 1907/2006; Registration, Evaluation, Authorization, and Restriction of Chemicals) [59]. In shooting ranges, Pb toxicity is related to the constant weathering of ammunition, which increases its presence in the soil matrix [60,61]. Under certain soil conditions (i.e., depending on the values of soil pH, organic matter content, cation-exchange capacity, redox potential, clay mineral content, etc.) [62], soil Pb will undergo oxidation [PbO], carbonation [Pb(CO₃)₂] and/or hydration processes [Pb(CO₃)₂(OH)₂], leading to a possible increase in Pb mobility and bioavailability [63]. It is therefore essential to understand the geochemical behavior of Pb in shooting range soils in order to manage, and preferably reduce, potential risks for humans and the environment [19]. Soluble Pb can bioaccumulate in living organisms, thus causing toxic effects on terrestrial and aquatic biota at different trophic levels [4,64], and references therein]. It is widely recognized that Pb can affect plant performance [65] and reduce the biomass, activity and diversity of soil microorganisms, which are key to soil fertility and health [66]. The major impact on soil biota is expected to result from the unintended ingestion of Pb by consumption of soil, plants and water [4,67]. Moreover, as many shooting ranges are commonly located near urban and/or agricultural areas [9,10,68], they also represent a high risk of Pb migration to humans through metal-soil-crop interactions [61].

Lead has a prolonged persistence in the environment (it is non-degradable) [69], tends to form relatively stable complexes with organic and inorganic soil ligands, and is soluble only within a limited acidic pH range [4]. The water-soluble and exchangeable forms of metals in soils broadly determine their mobility and bioavailability (frequently considered as proxy for toxicity) [70]. Therefore, soil Pb generally shows restricted mobility, solubility and bioavailability [71,72]. However, its low solubility does not completely prevent groundwater contamination via Pb leaching [47,64,73,74], and references therein]. The abovementioned long residence time of Pb in shooting range soil makes it a long-term environmental threat for ecosystems and human health, as potential adverse effects on soil biological and physico-chemical properties can persist for centuries [73].

The environmental impact of Pb-contaminated shooting range soils usually varies from site to site, owing to variations in soil conditions and Pb speciation, as well as depending on the capacity of the local biota to absorb, bioaccumulate and biomagnify toxic Pb [47,75]. Risk assessment of shooting ranges has been undertaken in many studies [47,76,77] using selected representative (and sensitive) species to examine and quantify the potential toxic effects of Pb and co-contaminants. Recent studies have examined the beneficial effects of different remediation strategies on these biological endpoints [20,47,78]. After all, for a direct and proper determination of contaminant-induced adverse biological effects, biological measurements are needed, as chemical measurements of extractability and bioavailability (frequently considered as proxies for contaminant toxicity) are questionable estimations [79].

Among the remediation technologies that have been developed and traditionally implemented for the clean-up of metal-polluted soils, *in situ* strategies such as chemical immobilization through the use of industrial and agro-food by-products and wastes (preferably, of local origin) are most promising to achieve an environmentally sound, cost-effective remediation of Pb-contaminated shooting range soils. These by-products and wastes offer the following advantages: (1) they can make Pb less bioavailable and thus minimize the risks to environmental and human health; (2) they can improve soil physico-chemical and biological properties and, hence, soil fertility and vegetation re-establishment; (3) their use as soil amendments provides additional environmental (i.e., recycling and reutiliza-

tion) [80,81] and economic (i.e., avoids costs associated with treatment and landfill disposal) benefits.

As previously stated, industrial and agro-food by-products and wastes are currently considered valuable immobilizing agents for the remediation of Pb-contaminated shooting range soils [82]. Promising outcomes have been observed using inorganic wastes, such as fly ash, mud or red mud, and furnace slag, as soil amendments (Table 3A). Soil amendments from biological origins, such as oyster shells, egg shells, plant remains and biochar, have also been successfully used for the remediation of Pb contaminated soils (Table 3B). The fact that these materials are often easily available and/or have a relatively low cost, together with the abovementioned environmental and social benefits, has encouraged their use for the in situ immobilization of metal contaminants in shooting range soils.

Table 3. Industrial (A) and agro-food (B) by-products and wastes used as amendments for Pb immobilization in shooting range soils.

A. Industrial By-Products and Wastes Used as Inorganic Soil Amendments						
Type of Amendment	Activity	By-Product or Waste	Reference	Type of Study (Duration)	Treatment Rates	Range of Soil [Total Pb] * (mg kg ⁻¹)
Metal oxide-based amendments	Coal burning	Fly ash	Moon et al. (2013) [80]	Lab (7 d)	(L:S) ratio of 0.2	7996
	Alumina extraction	Red mud	Sanderson et al. (2015) [38]	Lab (12 mth)	0.5, 1, and 2%	399–10,403
			Sanderson et al. (2014) [47]		2% (<i>w/w</i>)	2330–12,167
Blast furnace	Furnace slag	Levonmäki and Hartikainen (2007) [49]	Lab (21 d)	5 t ha ⁻¹	22,600	
B. Agro-Food By-Products and Wastes Used as Soil Amendments of Biological Origin						
Lime-based amendments	Shellfish farming	Oyster shells (calcined)	Moon et al. (2013) [80]	Lab (7 d)	(L:S) ratio of 0.2	7996
			Moon et al. (2013) [83]	Lab (28 d)	3–10% (<i>w/w</i>)	11,900
			Moon et al. (2016) [84]	Lab (28 d)	0, 5, and 10% (<i>w/w</i>)	8588
			Lee et al. (2013) [85]	Lab (28 d)	5% (<i>w/w</i>)	7996
	Oyster and mussel shells	Mussel shell	Ahmad et al. (2012) [82]	Lab (1 d)	2.5, 5, 10, and 15%	4626
			Ahmad et al. (2012) [86]	Lab (7 d)	5% (<i>w/w</i>)	11,100
			Ahmad et al. (2014) [87]	Lab (175 d)	5% (<i>w/w</i>)	3970
	Poultry industry/farming	Eggshells (calcined)	Ahmad et al. (2012) [20]	Lab (28 d)	0, 2.5, 5, 10, 15, 20, 25, and 30%	4626
			Almaroai et al. (2014) [22]	Lab (21 d)	5% (<i>w/w</i>)	4626
		Poultry waste (incinerated)	Poultry litter waste	Ahmad et al. (2012) [82]	Lab (1 d)	2.5, 5, 10, and 15%
Hashimoto et al. (2008) [27]				Lab (15 d)	3% (<i>w/w</i>)	19,600
Hashimoto et al. (2009) [88]	Lab (90 min)	P/Pb molar ratio of 3:5	29,200			
Hashimoto et al. (2009) [89]	Lab (7 d)	0, 0.5, 1, 3, and 6% (<i>w/w</i>)	18,100			
Phosphate amendments	Livestock activity	Bone meal or powder	Almaroai et al. (2014) [22]	Lab (21 d)	5% (<i>w/w</i>)	4626
			Moon et al. (2013) [83]	Lab (28 d)	3–10% (<i>w/w</i>)	11,900
			Ahmad et al. (2012) [86]	Lab (7 d)	5% (<i>w/w</i>)	11,100
			Ahmad et al. (2014) [87]	Lab (175 d)	5% (<i>w/w</i>)	3970

Table 3. Cont.

B. Agro-Food By-Products and Wastes Used as Soil Amendments of Biological Origin						
Type of Amendment	Activity	By-Product or Waste	Reference	Type of Study (Duration)	Treatment Rates	Range of Soil [Total Pb] * (mg kg ⁻¹)
Phosphate amendments	Livestock activity	Bone biochar	Sanderson et al. (2015) [38]	Lab (12 mth)	1:1 proportion	399–10,403
		Chicken manure- and green waste-derived biochars	Park et al. (2011) [46]	Lab (14 d)	5% (w/w)	346
		Dairy manure biomass derived biochar	Cao et al. (2011) [90]	Lab (210 d)	2.5 and 5% (w/w)	70,000
Carbon-rich amendments	Livestock activity	Broiler litter biochars	Uchimiya et al. (2012) [91]	Lab (7 d)	0, 2, 5, 10, and 20% (w/w)	19,906
	Agroforestry	Buffalo weed (<i>Ambrosia trifida</i> L.) biomass derived biochar	Rajapaksha et al. (2015) [24]	Lab (12 mth)	5% (w/w)	17,468
		Charcoal mixed with compost made with sewage sludge, freshly chopped lop, grass and leaves, and soil and coarse wood branches	Lee et al. (2011) [92]	Lab (30 d)	0, 1, 3, 5, 10, 20, and 30%	4626
		Soybean (<i>Glycine max</i>) stover derived biochar	Moon et al. (2013) [93]	Lab (7 d)	1–20% (w/w)	11,885
		Oak wood biochar	Almaroai et al. (2014) [22]	Lab (21 d)	5% (w/w)	4626
			Ahmad et al. (2012) [86]	Lab (7 d)	5% (w/w)	11,100
			Ahmad et al. (2014) [87]	Lab (175 d)	5% (w/w)	3970
		Soybean stover and pine needles derived biochars	Ahmad et al. (2016) [94]	Lab (90 d)	10% (w/w)	17,468
Soybean stover, peanut (<i>Arachis hypogaea</i>) shell, and pine needle derived biochars	Ahmad et al. (2017) [95]	Lab (24 h)	10% (w/w)	17,468		
Soybean (<i>Glycine max</i>) stover derived biochar	Vithanage et al. (2017) [96]	Lab (1 mth)	0, 0.5, 1, and 2.5%	17,467		
Shredded timber waste derived biochar	Silvani et al. (2019) [97]	Lab (14 d)	0, 5, 10, and 20%	4300–6600		

* Range of soil total Pb concentration (mg kg⁻¹) min–max averages.

The selection of the most appropriate immobilizing amendment is critical for the intended remediation technique to be cost-effective and environmentally sound [62,98]. In the following sections, some of the aforementioned inorganic and organic amendments are discussed in detail in relation to their potential for the remediation of Pb-contaminated shooting range soils.

3. Industrial By-Products and Wastes as Inorganic Soil Amendments

The disposal of industrial by-products is facing increasing challenges derived from the high costs of operating landfills, in combination with the scarcity of landfill sites and the well-known potential risks of contamination linked to these sites. Alternatively, some industrial by-products, such as fly ash and red mud, can be used as soil amendments, thus offering a sustainable way for the management of contaminated areas in compliance with the EU Waste Framework Directive (Directive 2008/98/EC) [52] and its generic End-of-Waste (EoW) criteria.

Coal fly ash (CFA), also known as pulverized fuel ash, is the major solid by-product generated from coal burning in thermal power stations [62]. The total amount of fuel ash produced worldwide is enormous, in the region of 750 million tonnes per year [99]. Although CFA is one of the cheapest and most widely available waste materials suitable for the reclamation of degraded soils [99,100], the global average utilization rate of CFA is estimated to be less than 25% [101–103].

Coal fly ash is an alkaline material that is composed mainly of ferroaluminosilicates, with high adsorption capacity [104,105]. Numerous studies have documented the potential of CFAs to (i) increase the surface area available for cation adsorption; (ii) improve soil physical properties; (iii) neutralize the pH of acidic soils; (iv) render cationic metals less mobile; (v) enhance soil productivity, as they contain alkali (K^+) and alkaline earth metals (Ca^{2+} , Mg^{2+}), which are essential plant nutrients [99, and references therein,105]. The application of CFA as a soil ameliorant has great potential for Pb immobilization in acidic soils [62]. The pH of acidic soils usually needs to be increased to alleviate metal-induced toxic effects on plants and soil biota. Although limestone, composed mainly of $CaCO_3$ and/or $CaMg(CO_3)_2$, has been commonly used as an amendment to increase soil pH, many CFAs, especially those containing alkalizing agents (e.g., CaO and $Ca(OH)_2$), also have the potential to rectify soil acidity problems, while supplying vital plant nutrients [99]. In addition, the use of CFA instead of limestone can reduce CO_2 emissions (via surface adsorption and carbonation reaction), with concomitant beneficial effects in terms of mitigating climate change [103].

Although the effectiveness of CFA as a metal-immobilizing agent in soil has been demonstrated [103,106–108], few studies have actually used this material in Pb-contaminated soils and still fewer have used it for the remediation of shooting range soils. Moon et al. (2013) [80] reported that the combination of calcined oyster shells and CFA was a cost-effective treatment for Pb immobilization in contaminated military firing range soil. Dermatas and Meng (2003) [109] demonstrated that the addition of quicklime and CFA to Pb/Cr artificially contaminated soils effectively reduced Pb leachability below the non-hazardous regulatory limit. Ciccu et al. (2003) [110] and Moon and Dermatas (2006; 2007) [111,112] observed that a relatively small addition of CFA resulted in a significant decrease in the amount of Pb leached from contaminated soils. A greenhouse experiment found that the application of CFA significantly reduced soluble and exchangeable Pb (14.7%) in contaminated soil, leading to a significant reduction of Pb in rapeseed plant tissue (66.1%) [62,113]. Similarly, Kumpiene et al. (2007) [105] observed that the application of CFA and peat to a Cu/Pb-contaminated soil reduced Pb leaching by 97% during a two-year field observation period, which allowed for an increased seed germination rate, reduced metal accumulation in plant shoots, and decreased toxicity to plants and bacteria.

In summary, CFA is a feasible amendment to reduce Pb toxicity and improve the physico-chemical and biological properties of shooting range soils. There are, nonetheless, factors that restrict CFA application in agricultural and natural soils, such as the presence

of potentially toxic metalloids and metals (e.g., As, Se, and Cd), high salinity and capacity to reduce the solubility of some essential nutrients [114].

Red mud (also called bauxite residue) is an alkaline solid by-product generated during alumina extraction from bauxite in refineries [115–117]. As a result of the high demand for aluminum, the global inventory of red mud has reached an estimated 4 billion tons, based on its current rate of production, and is still rapidly increasing [117–120]. Currently, red mud is treated as a waste product and is stored in large lagoons or land-based disposal areas [121], which require continuous maintenance. However, its abundance has stimulated extensive research into possible uses [122,123], including its use as a soil amendment for the remediation of soils characterized by high levels of metal(loid)s and low pH values [116,124].

In general, regardless of the specific production process, red mud contains six major constituents, namely Fe_2O_3 (about 20–40% by mass), Al_2O_3 , SiO_2 , TiO_2 , Na_2O and CaO [125–128], as well as an array of minor elements, including Cr, Cu, Pb, and Zn [125,127,128]. Since red mud itself contains variable amounts of metal(loid)s and high pH and alkalinity, these characteristics should be carefully considered before reusing red mud for the remediation of contaminated soils [117,129].

Red mud exhibits a greater sorption capacity of Pb and other metals than fly ash, probably because it contains larger amounts of Fe- and Al-oxides and hydroxides, a larger surface area and pH stabilization ability [130]. Sanderson et al. (2014; 2015) [38,47] applied red mud at a rate of 2% in four shooting ranges and observed a substantial reduction in exchangeable Pb. Several studies have also shown that red mud can reduce metal bioavailability and thus plant metal uptake and accumulation, while enhancing soil microbial biomass and activity and increasing plant biomass production ([123], and references therein). Nonetheless, Sanderson et al. (2014) [47] reported that red-mud-amended soils can be toxic to plants and earthworms, probably due to the resulting highly alkaline soil (pH 9–12). The high alkaline pH of red mud can be neutralized through its treatment with seawater, gypsum, etc. [129]. Tandy et al. (2017) [131] demonstrated that the lower pH of neutralized red mud made it more suitable for plant growth and led to lower values of dissolved organic carbon, thereby preventing loss of soil organic matter and, hence, retaining soil fertility and structure.

Despite their abovementioned benefits for the remediation of shooting range soils, the use of industrial by-products is currently limited, mainly due to high transportation costs, the need for prior processing and/or pretreatment, and data scarcity on their potential environmental impacts [132]. Another practical limitation is the lack of specific guidance or legislation regarding the application of fly ash [133] and red mud [116] in relation to their use for contaminated soil remediation.

4. Agro-Food By-Products and Wastes as Soil Amendments of Biological Origin

With the increasing global demand for food production, agricultural and food industries must deal with extremely large amounts of organic waste generated along the food supply chain (production, processing and consumption) [134–136]. Food waste is defined as “fractions of both edible and inedible parts of food removed from the food supply chain” [134]. Current estimates for the EU show that 75–100 million tons of food waste is produced along the food supply chain, which amounts to an average of about 170 kg per capita per year [134,137]. At present, the majority of food waste is treated as general waste and is therefore sent to landfill or incineration [136,138].

The food waste produced by the agro-food industry, in addition to being a great loss of valuable material, also raises serious problems both from an economic and environmental point of view [136,139]. In this respect, food waste emits over 20% of the total global production of greenhouse gases, such as methane (CH_4), nitrous oxide (N_2O), and carbon dioxide (CO_2) [136,140,141]. Additionally, food waste leachates from landfills constitute a high potential risk for contamination of surface waters and groundwater [23]. As a result of the incineration of food waste, emissions to air of particulate matter (especially

dust), acid gases (HCl, HF, SO₂⁻, CO, NO_x), dioxins and VOCs [142,143] are produced, with concomitant negative impacts on human and ecosystem health. On the other hand, incineration often involves high energy consumption [136,144,145].

Food waste must be urgently reduced and diverted from disposal in landfills, as advocated by the Integrated Product Policy (IPP) [146], the Roadmap to a Resource Efficient Europe [147], now embodied in the Resource Efficiency Flagship of the Europe 2020 Strategy [148], the Bioeconomy Strategy [149], Target 12.3 of the UN Sustainable Development Goals (which aims at halving wastage by 2030), and the European Green Deal Strategy [56]. The agro-food sector is particularly committed to adopting strategies and challenges within circular economy principles, and therefore, actions are being taken towards the valorization and recycling of food waste, with a view to transforming it into a valuable resource. Food waste can be valorized into a range of high-value-added products, including biofertilizers and soil amendments, to, for instance, reverse land degradation in agricultural areas and assist in the remediation of contaminated soils [136]. For acidic and clayey soils in shooting ranges close to forested areas, the US EPA (2005) [5] traditionally recommends the application of lime-based amendments. However, limestone mining can be a destructive process, with cascading environmental impacts [150]. As a more sustainable alternative, biogenic carbonate materials, such as treated oyster and mussel shells and eggshells, are also being used for metal removal and immobilization in shooting range soils (Table 3B). The disposal and utilization of products of animal origin and animal by-products in soils not intended for agricultural use is regulated by Commission Regulation (EU) No. 142/2011, which sets out methods for disposal or use defined by Regulation (EC) No. 1069/2009 in order to minimize potential risk to public and animal health as well as to avoid any threat of pathogens and disease dispersion [151].

Shellfish farming is an expanding economic activity worldwide and generates a large volume of waste, including an abundance of mollusk shells [152]. In China, the largest producer of shellfish in the world, about 10 million tons of waste seashells are disposed of in landfills annually [153]. Seashell waste, which includes oyster and mussel shells, is particularly rich in calcite (CaCO₃) and quicklime (CaO) components, which might form insoluble metal hydroxides at alkaline pH values [154–156]. To convert the relatively less reactive CaCO₃ into highly reactive CaO, a calcination process is usually adopted. Moon et al. [80,83] stabilized Pb and Cu in a shooting range soil located in Busan Metropolitan City (Korea) using calcined oyster shells mixed with bone meal or fly ash as amendments. These authors tested the effectiveness of the stabilization process according to the Toxicity Characteristic Leaching Procedure using the 0.1 M HCl extraction method and observed a drastic reduction of Pb (98%) and Cu (96%) leaching when soils were treated with calcined oyster shell, whether it was mixed with bone meal (5% each on a weight basis) or with fly ash (10% and 5%, respectively). From the results of their batch- and column-leaching tests, Lee et al. (2013) [85] concluded that the application of calcined oyster shell waste, together with coal mine drainage sludge, was an effective treatment for the simultaneous immobilization of As, Pb and Cu in soils from a Korean military shooting range. In a similar study, scanning electron microscopy-energy dispersive X-ray spectroscopy analyses performed by Moon et al. (2016) [84] indicated that As immobilization may be associated with the formation of Ca-As and Fe-As precipitates, while Pb and Cu immobilization was most probably linked to calcium silicate hydrates and calcium aluminum hydrates.

Oyster shells, mussel shells and eggshells were assessed by Ahmad et al. (2012) [82] for their effectiveness towards Pb and Cu immobilization in highly contaminated shooting range soil. The rise in soil pH due to amendment application resulted in the formation of relatively insoluble Pb-hydroxide. However, all lime-based waste materials failed to immobilize Cu. This is not surprising, since it is a well-known fact that soil composition can significantly affect the performance of these amendments.

Eggshell is also one of the most important food wastes, with an annual worldwide production of ca. 250,000 tons [157]. Eggshell is an alkaline and nitrogen rich material with

a high content of CaCO_3 (>90%), embedded in an organic framework of protein fibers [151]. Ahmad et al. (2012) [86] investigated Pb immobilization in shooting range soil amended with raw and calcined eggshell waste. Both amendments decreased the exchangeable Pb fraction to 1% of the total soil Pb content (>4600 mg kg^{-1}), while the carbonate-associated Pb fraction was increased to 40.0–47.1% at >15% application rates. These authors proposed that their eggshell amendment was immobilizing soil Pb via the formation of $\text{Pb}(\text{OH})_2$ or $\text{Pb}_2\text{O}(\text{SO}_4)$, adsorption on Al-containing minerals, and co-precipitation as carbonate. In turn, Pb in soil amended with calcined eggshell was associated with Si and Ca^{2+} and then possibly immobilized by entrapping into calcium-silicate-hydrate [86].

Among the best management practices to prevent soil Pb migration, phosphate application, alone or in conjunction with lime, is recommended by the US EPA. Immobilization of Pb with phosphates in contaminated soils relies on the formation of the highly insoluble mineral pyromorphite $[\text{Pb}_5(\text{PO}_4)_3\text{X}]$, where $\text{X} = \text{Cl}^-, \text{OH}^-, \text{F}^-$ [158]. Given the relatively high cost of phosphate treatments, bone meal (calcium phosphate) can also be considered as a readily available phosphate source for the remediation of Pb-contaminated soils [159]. In fact, the bioavailable fraction of Pb in shooting range soils can be reduced effectively by the addition of bone meal as an amendment, thereby decreasing the risk of ecotoxicity [20,22,83]. Ahmad et al. (2014) [87] reported that stable Pb species, such as Pb-hydroxide, Pb sorbed on kaolinite and chloropyromorphite, were formed in shooting range soils amended with mussel shells, biochar and bone meal, respectively.

Over the last years, one of the most promising options for achieving the so-called “zero-waste-to-landfill” goal in agriculture and forestry is to convert wastes into biochars. The term biochar refers to a porous carbonaceous solid produced by thermochemical conversion of organic materials in the absence of oxygen [160]. Biochar is commonly produced from pyrolysis of animal manures, crop residues, grasses and weeds, wood biomass, sewage sludge and municipal solid waste at a temperature ranging from 200 to 800 °C [161,162]. The unique physico-chemical and sorptive properties of biochar, such as large surface area, high porosity, dominance of negatively charged surface functional groups, high cation exchange capacity and long-term stability [163], make it highly recommended as an amendment for contaminated soils. Biochar amendments can immobilize positive Pb cations through electrostatic attraction, precipitation as Pb-phosphate, surface complexation, and increasing soil pH [24,87,90,94,95]. Nonetheless, the effectiveness of biochar for metal immobilization is heavily determined by the source of biomass feedstock and the specific pyrolysis conditions [19,164,165].

A number of studies have examined the suitability of biochars for metal immobilization in shooting range soils, as summarized in Table 3B. In most of these studies, short-term batch experiments were conducted using biochars derived from manures [46,90], crop residues [24,85,93–97] and oak wood [20,22,87]. In most cases, biochar proved to be a versatile functional material, not only because it promoted soil Pb immobilization, but also because its application led to the incorporation (and retention) of labile organic carbon and nutrients, which in turn improved soil fertility and enhanced plant growth and soil microbial activity [166,167]. Even though the majority of studies showed the beneficial effects of biochar, the lack of field-scale studies [168], scarce ecotoxicological data [169,170] and legal restrictions [171] still limit its application in soil amendment.

The selection of the most appropriate amendment among those described in Sections 3 and 4 for the in situ remediation of a specific shooting range soil is a complex but essential task. When selecting industrial or agro-food by-products (or other wastes) as soil amendments, the following aspects should be simultaneously taken into consideration: (i) site-specific edaphoclimatic conditions, including both soil physicochemical and biological properties; (ii) contamination type and the extension of the affected area; (iii) the production rate and availability of the by-product or waste as well as the availability of space for its proper storage; (iv) physical characteristics and chemical composition of the by-product or waste as well as potential the environmental risks associated with high conductivity (salinity) and/or very high/low pH values; (v) cost of the transformation processes required

prior to amendment application as well as transport costs; (vi) available information on application procedures and the effectiveness for metal (Pb) immobilization, obtained from both microcosm and field experiments; (vii) public acceptance regarding the use of industrial and agro-food by-products as soil amendments.

5. Effectiveness of Agro-Industrial Wastes as Amendments for the Remediation of Pb-Contaminated Shooting Range Soil: A Practical Example from Basque Country (Spain)

In Spain, where it is estimated that hunting and shooting are responsible for the dispersion in nature of about 6000 tonnes of Pb per year [172,173], there are over 770 shooting ranges [10,174]. According to a local hunting foundation [175], hunting is practiced by 1.6% of the population of the Basque Country (northern Spain). In many cases, recreational shooting ranges are located in meadows nearby forest plots and close to farm houses and/or agricultural lands, making these environments vulnerable to Pb contamination. Since shooting has been classified as a potentially contaminating activity by Spanish legislation (Spanish Royal Decree (RD) 9/2005), several initiatives are being conducted to remediate shooting range soils impacted by Pb contamination [10,42,174].

Phytomanagement practices have positioned themselves as cost-effective technologies for the sustainable long-term remediation of large areas affected by mild to medium levels of soil contamination [176–180]. Phytomanagement usually involves the cultivation of profitable plants (mainly non-food crops) in organically amended soils to control the bioavailable fraction of contaminants, thereby minimizing environmental risks while maximizing ecological, social and economic revenues [70,176,179]. It is necessary to conduct preliminary research to gather data on the effectiveness of the available amendments for metal immobilization before their use for on-site phytomanagement.

The main objective of the practical experience detailed below was to evaluate the effectiveness of three different agro-industrial wastes for soil Pb immobilization at a shooting range situated in the municipality of Oiartzun (Basque Country, Spain). Specifically, we compared the effect of the application of spent mushroom substrate (SMS), forest wood-derived biochar (BC) and bone meal (BM) on the CaCl_2 -extractable Pb fraction under laboratory and field conditions. A detailed description of materials and methods for this practical experience can be found in the Supplementary Materials. Briefly, a microcosm experiment was carried out using topsoil (0–20 cm) with a total Pb concentration of 2058 mg kg^{-1} DW soil. This Pb concentration was approximately 17 times higher than the Critical Reference Concentration established by Basque legislation [181] and 7 times higher than the maximum allowable concentration for agricultural settings set by European legislation [17]. The CaCl_2 -extractable Pb concentration of soil samples from this shooting range was approximately 12 mg kg^{-1} DW soil, accounting for 0.6% of the total Pb concentration. This shooting range soil, therefore, was severely contaminated with Pb and required the implementation of an effective remediation strategy. To this end, first, a microcosm experiment was conducted in order to evaluate the effectiveness of three agro-industrial wastes (SMS, BC and BM: 5% *w/w*) under controlled laboratory conditions. A control treatment (without amendment) was also studied. Four independent pots for each treatment were arranged. Lead immobilization was evaluated in terms of the reduction of CaCl_2 -extractable Pb. Blank samples were included in all the analyses to verify any possible cross-contamination. Soil pH was also measured. Following the microcosm experiment, a field experiment was established in the shooting range. Biochar and BM (5% *w/w*) were applied as soil amendments in three independent plots. An unamended control treatment was also included in this on-site experiment. Soil samples from each plot were collected at 0, 7 and 17 days after amendment application to determine soil extractable Pb and pH.

In the microcosm study, immediately after their application, BC and BM amendments significantly ($p < 0.05$) decreased extractable Pb concentrations compared to the control and SMS-amended soils (Figure 1). The addition of BC and BM to the shooting range soil caused a pronounced reduction (90 and 95%, respectively) of extractable Pb compared

to the control treatment. This effect remained for up to 7 days for BC and BM. For less effective treatments (C, SMS), a significant increase in extractable Pb concentration was observed at this time. Under our experimental conditions, treated soils had a short aging period (7 days). Other abiotic factors (redox reactions, interactions with soil components, etc.) and/or the activity of soil microorganisms could be responsible for the observed increase in extractable Pb levels.

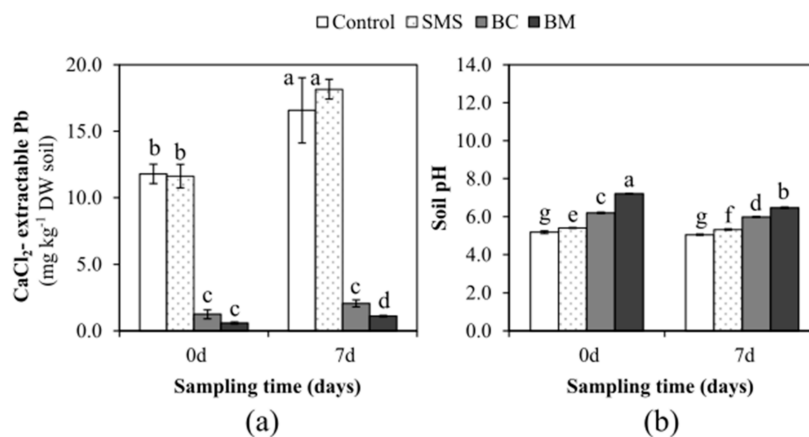


Figure 1. (a) CaCl₂-extractable Pb concentrations and (b) pH of control soil and SMS-, BC- and BM-amended soils at 0 and 7 days after amendment application. Error bars represent standard errors. Different lower case letters above error bars indicate significant differences at $p < 0.05$ among means of different treatments.

In addition to the effective interaction of Pb with specific chemical components of BC and BM amendments (see Section 4 and reviews [9,19,28]), the higher pH values reported in BC- and BM-amended soils (pH 6.2 and 7.2, respectively) in comparison with control soils (pH 5.2) (Figure 1b) could also contribute to the reduction in extractable Pb concentrations. In fact, a statistically significant negative correlation between extractable Pb in amended soils and pH (0 d: $\rho = -0.853$; 7 d: $\rho = -0.778$, both $p < 0.05$) was observed. The induction of soil alkalization by the application of amendments is an alternative for promoting soil Pb immobilization, as previously pointed out by Ahmad et al. (2012; 2014) [20,87]. The addition of SMS to soil under our experimental conditions, however, did not alter extractable Pb concentrations.

Our field experiment confirmed the above-mentioned effectiveness of BC and BM amendments by significantly reducing soil extractable Pb concentrations (Figure 2a). In the amended soil, Pb remained immobilized, even 17 days after the application of the amendments. Our findings indicate that a 5% BC and BM application rate is appropriate for Pb immobilization in shooting range soil. When measuring soil microbial activity, BM-amended soils exhibited higher values of CO₂ per unit biomass and time ($\approx 18 \mu\text{g CO}_2\text{-C g}^{-1} \text{ soil h}^{-1}$) than control and BC-amended soils ($\approx 2 \mu\text{g CO}_2\text{-C g}^{-1} \text{ soil h}^{-1}$). This higher CO₂ production could be due to decarbonation processes.

Our findings are in good agreement with previous studies by Ahmad et al. (2012; 2014) [20,87], which related metal retention onto soil particles (via metal phosphate formation, adsorption/complexation and ion exchange) with an increase of pH in shooting range soils treated with 5% BC and BM. The high efficiency of both BC and BM for soil Pb immobilization may be attributed to the adsorption of Pb [46] and precipitation of Pb-phosphate [159], respectively. The immobilization of Pb after the incorporation of biochar to soil has been associated with elevated concentrations of soluble carbon [46] (Table S1, Supplementary Materials), its high-sorption properties due to its relatively high-specific surface area (porous structure) [22], and the abundance of carboxyl functional groups [91]. The BM is usually characterized by a high content of Ca-bearing minerals and P in the

form of P_2O_5 [87] (Table S1). These components may have contributed to the formation of geochemically stable Pb species, leading to a lower leachability [83].

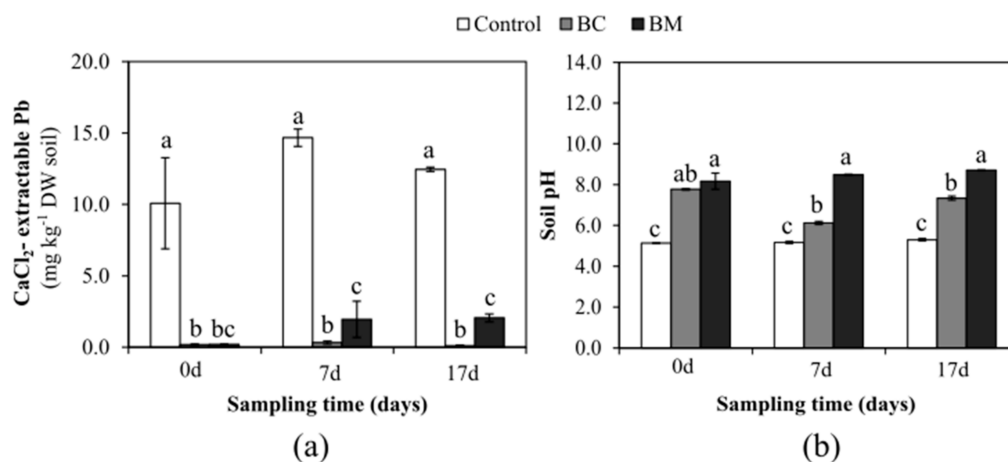


Figure 2. (a) $CaCl_2$ -extractable Pb concentrations and (b) pH of control soil and BC- and BM-amended soils at 0, 7 and 17 days after amendment application, under field conditions. Error bars represent standard errors. Different lower case letters above error bars indicate significant differences at $p < 0.05$ among means of different treatments.

In view of the high Pb immobilization capacity of BC and BM, we concluded that these wastes are suitable amendments for the phytomanagement of soils affected by shooting activity. The immobilization effect of amendments can be initially assessed with a simple and cheap approach at a microcosm scale in the laboratory. The presented practical experience under field conditions shows the immediate (0 days) and short-term (7 days, 17 days) effects of amendment application on Pb extractability in shooting range soil. Nonetheless, further on-site studies are required to determine the longer term effects of these soil amendments, not only on Pb immobilization but also on soil health.

6. Concluding Remarks

Shooting ranges have been identified as sources of Pb contamination. Chemical immobilization, through the use of inorganic and organic soil amendments, is one of the most effective strategies for reducing Pb extractability and bioavailability and, hence, ecotoxicity. Amendments from agro-food and industrial by-products and wastes have great potential for soil remediation purposes. The practical experience presented here opens the door to the application of forest wood biochar and bone meal for the remediation of shooting range soils, thus fostering the implementation of circular economy principles for a sustainable waste management. Despite increasing research on the application of soil amendments to immobilize soil Pb, there are still some aspects that need to be developed. For instance, ongoing research is mostly focused on short-term laboratory experiments or leaching tests. Results from these studies should be verified by field studies. Field-scale studies not only allow researchers to confirm the effectiveness of by-products and wastes as metal immobilizing agents, but also to optimize their application rate and assess the effect of site-specific edaphoclimatic conditions on extractable metal concentrations. It is also important to investigate how these amendments can interact with other co-contaminants of shooting ranges, such as Sb, as well as their long-term effects on soil biota and vegetation. Finally, remediation studies of shooting range soils should address, when possible, the links and trade-offs between environmental and socio-economic gains.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/pr9040572/s1>, detailed description of materials and methods for Section 5 and Table S1: Physico-chemical characteristics of spent mushroom substrate (SMS), biochar (BC) and animal bone meal (BM) on a dry weight basis.

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