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► **To cite this version:**

Ashley Schnackenberg, Géraldine Bidar, Valérie Bert, Patrice Cannavo, Sebastien Detriche, et al.. Effects of Inorganic and Organic Amendments on the Predicted Bioavailability of As, Cd, Pb and Zn in Kitchen Garden Soils. *Advances in Environmental and Engineering Research*, 2022, 3, pp.1 - 1. 10.21926/aeer.2201004 . hal-03583334

HAL Id: hal-03583334

<https://hal.science/hal-03583334v1>

Submitted on 21 Feb 2022

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Original Research

Effects of Inorganic and Organic Amendments on the Predicted Bioavailability of As, Cd, Pb and Zn in Kitchen Garden Soils

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Academic Editor: Amanda Laca

Special Issue: [Environmental Impact Assessment](#)

Adv Environ Eng Res

2022, volume 3, issue 1

doi:10.21926/aeer.2201004

Received: November 11, 2021

Accepted: January 23, 2022

Published: February 08, 2022

Abstract

Moderately contaminated garden soils can benefit from gentle remediation options such as soil amendments, which improve soil functions and agronomic potentialities while decreasing



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environmental and human risk. This study aimed to analyze the effects of doses of various common soil amendments generally applied by gardeners on the predicted bioavailability (i.e., extractability) of metal(loid)s (i.e., As, Cd, Pb, and Zn) in contaminated kitchen garden soils. Fourteen different amendment mixes (i.e., a green waste compost with two degrees of maturity used alone and in combination with zeolite, three organic fertilizers, two calcareous amendments, two natural siliceous or alumino-silicate amendments, and one potting soil) were tested on three different garden soils with diverse sources of contamination and physico-chemical characteristics. Chemically extractable metal(loid)s were analyzed using 0.05 M EDTA extraction and 1 M NH_4NO_3 extraction. In one soil sample, potting soil showed significant potential to reduce the availability of As, as analyzed by both extractants. This amendment also effectively reduced the Pb extractability in the geogenic-contaminated soil, as did other high-organic matter amendments such as various application rates of composts. Zeolite and zeolite-compost mixes demonstrated success on various metal(loid)s and therefore could be a promising emerging amendment mix. Other efficient amendments include crushed horn, which effectively reduced available Zn in all soils, as well as available Pb. The application of bone meal similarly reduced the extractable As, Pb, and Zn in various soils. The two applications of limes were effective against Cd, As, Pb, and Zn in the different soils studied. This study provided evidence that it is possible to reduce the extractability and thus the environmental availability of the metal(loid)s applied with available and affordable amendments. The results depended on the physico-chemical soil parameters and metal(loid)s considered. There is no single solution, which implies that tests must be carried out before any implementation activities on the kitchen gardens.

Keywords

Kitchen garden soils; metal(loid)s; amendments; extractability; immobilization

1. Introduction

Urban gardening is a growing trend in several cities in the world. The need for nature in the city, the growing movement towards healthy eating, and difficult economic and societal contexts have led to a re-emergence of gardening activities. This applies to both private gardens—most commonly adjoining homes—as well as community gardens. Several cities and urban communities are required to install and support community gardens, a term that includes allotments, shared, or integration gardens. It is common to see these gardens on urban wastelands, along roads or railways, near previously or currently active industrial sites, or even on former industrial or urban wastelands. In comparison with agricultural or forest soils, urban soils are more complex, often very heterogeneous, and which, depending on their history and/or their environment, may have poor agronomic qualities [1, 2]. These contexts also suggest the presence of inorganic contaminants (such as As, Cd, Pb, and Zn) of various origins and in variable concentrations [3, 4]. In addition to a geogenic origin (linked to the geochemical background) of the contaminants, soil contamination may result from past and/or present discharges from industrial or urban activities (e.g., combustion of fossil fuels, automobile traffic, incineration of household waste) [5-8], or from improper

gardening practices (e.g., use of phytosanitary products, mineral fertilizers, contaminated irrigation water) [9-11].

Human exposure to metal(loid)s in soils can present risks that vary by the element, exposure pathway, vector, and receptor [12]. Considering kitchen gardens, humans may be exposed via ingestion/inhalation of soil particles and consumption of contaminated vegetables. Ingestion may be considered the main exposure route associated with the cultural practices of gardening [13]. Commercially sold vegetables are regulated by the European Union (European Directive of 25 June modifying the European Directive no. 1881/2006), which defines the maximum authorized concentrations of metals such as Cd and Pb in marketable food. These regulations do not apply to kitchen garden vegetables cultivated by individuals [14]. The accumulation of metal(loid)s in crops could vary as per the physico-chemical parameters of the soils, the metal(loid) speciation, the cultural practices (amendments, irrigation), the vegetable species, and the cultivar. The behavior of metal(loid)s in the soil relies on certain soil parameters, including but not limited to pH, cation exchange capacity, organic matter, nitrogen, phosphorous, carbonates, clay content, and biological activities [15-17]. According to these parameters and their temporal evolution, only a fraction of the metals is available for the soil biota. In general, reducing the availability of pollutants can play a key role in limiting their phytoavailability and potential toxicity via ingestion (and to a lesser extent, inhalation) of dust/soil particles and via consumption of vegetables [18-21].

There are various potential remediation strategies to limit the availability of metal(loid)s in soils. Regardless of the strategy utilized to manage kitchen garden soils, soil functions must be restored and maintained, and their agronomic potentialities improved. A feasible way to do this is via gentle remediation options [22]. One such method is using cost-effective, accessible, and easily applicable soil amendments [23-27]. Inorganic and organic soil amendments can act as *in-situ* metal(loid)-immobilizers and stabilizers [28]. Five types of products can be identified, namely, organic amendments (e.g., manure, compost, peat), natural organic fertilizers (e.g., crushed horn, bone powder), calcareous amendments (e.g., lime flower, magnesian lime), natural siliceous or alumino-silicate amendments (e.g., diatomaceous earth, natural zeolites), and expanded alumino-siliceous amendments (e.g., perlite, vermiculite).

Compost is the most commonly used among the organic amendments used by gardeners [29]. It is rich in nutrients, contributes to the carbon supply, increases the water holding capacity of the soil, improves biological functioning, and is an alternative to chemical fertilizers [30-32]. Its production is relatively simple to set up, and it is most often self-produced using green waste from the garden or cities. In addition to improving the agronomic potential of soils, compost can favorably influence the behavior of metal(loid)s present in the soil [33-35] by reducing their mobility and thereby toxicity to the biosphere. These pollutants can be immobilized via sorption, complexation, precipitation, and redox reactions, especially using amendments containing humic substances and inorganic compounds. However, research on the effects of compost on the transfer of pollutants from soil to vegetable production is still limited [29, 36-40]. Results are occasionally contradictory, although most often, the addition of composts to garden soils reduces the concentration of metal(loid) in vegetable production. However, these effects depend on the stage of maturity of the composts, their nature, their application rate, their soluble organic matter content, and the physico-chemical parameters of soil [29, 34].

Inorganic amendments have various origins and natures [41, 42]. They can be natural products, byproducts, or industrial wastes that can sometimes be combined with organic amendments. The

physico-chemical processes involved in amended soils are a result of interactions between metal(loid)s, the soil constituents, and amendments. The referent mechanisms are those of chemical reactions such as adsorption or surface complexation (physisorption and chemisorption), precipitation, or coprecipitation. These reactions depend on the physical and/or chemical parameters of the soils, the biogeochemical cycles of the metal(loid)s, and biological activities. Phosphate/hydroxyapatite products are among the most studied mineral amendments [43, 44]. Their effects are dependent on not only the physico-chemical properties of the soils but also on the speciation of the phosphate ions and their respective counter ions. Carbonate and lime-based amendments are exploited for their alkaline character and are used to increase the pH of acid soils. The effective reduction of phytoavailability has been demonstrated for several metal(loid)s [45-53]. Aluminosilicates such as natural zeolites are efficient when employed alone for Cd and Pb [54, 55] and in a mixture with organic amendments in the case of Cd and Pb as well as Zn [56, 57].

The overall aim of the study was to examine a range of amendments, in particular either self-produced or cost-effective, commercially accessible and easily applicable composts and mineral amendments, to manage urban kitchen garden soils with moderate geogenic and/or nature of anthropogenic contamination. The effects of these amendments were evaluated on (i) the physico-chemical characteristics of soils, (ii) the extractability of As, Cd, Pb, and Zn using both chemical extractants (EDTA and ammonium nitrate), and (iii) their efficiency to reduce the metal(loid)s' extractability. An *ex-situ* experiment was carried out to compare the effects induced by a collection of 10 organic and mineral amendments, used alone or in combination, on three contaminated garden soils with different physical and chemical characteristics. One organic amendment (a green waste compost with two degrees of maturity), three organic fertilizers (crushed horn, bone powder, and organic complete fertilizer made from poultry manure), two calcareous amendments (lime flower and magnesian lime), two natural siliceous or alumino-silicate amendments (diatomaceous earth and natural zeolite), and potting soil (a mixture of peat, sand, perlite, and dolomite) were selected for the study. These are well-known products, commonly used by gardeners. Particular attention was paid to the rate of application of the amendments. The doses typically reported in studies are often very high and do not always reflect the cultivation practices. A specific objective of this study was to use appropriate application rates for each amendment tested, i.e., as close to cultural seeding practices as possible. The aim was to assess the potential of the amendments to sustainably manage the availability of metal(loid)s to limit human exposure.

2. Material and Methods

2.1 Experimental Kitchen Garden Soils

Experimental *ex-situ* assays were conducted using soils from three metal(loid)-contaminated kitchen gardens in France, representing a range of physico-chemical parameters, as well as origins and levels of moderate contamination.

One soil sample was collected from a private kitchen garden "PKG" (50° 26' 4 N and 3° 2' 18 E), in Evin Malmaison in the former coal-mining area of northern France. Here, considerable atmospheric dust emissions of Cd, Pb, and Zn were recorded, generated by the former lead smelter Metaleurop Nord. The second soil sample was collected from a community kitchen garden "CKG-N" (47° 16' 1 N and 1° 34' 30 W) in Nantes (in western France), in the vicinity of a highway. This area is associated with past agricultural activities. The arsenic and lead contamination in this soil is

primarily of geogenic origin due to the presence of a mineralized vein of micaschist rich in As- and Pb-bearing minerals. The third soil sample was collected from a community garden “CKG-L” (50° 37' 14 N and 3° 1' 39 E) in Lille (in northern France). The soil of this garden is derived from backfill and is influenced by atmospheric fallout from former industrial activities and proximity to a highway. This soil is primarily contaminated with Pb.

For each site, bulk soil was sampled from the topsoil horizon tilled by the gardeners (0–25 cm deep).

2.2 Treatment and Experimental Design

The three soils samples (PKG, CKG-N, and CKG-L) were first air-dried at a temperature below 40°C, homogenized, and gently crushed to pass through a 2-mm stainless steel sieve as per the ISO 11464 standard (AFNOR, 1995). For each soil sample, 15 modalities, one of which is a control, and three repetitions of each were considered. In total, 126 amended soil samples and 9 control soil samples (i.e., unamended soils) were tested in polypropylene pots of 500 ml. 500 g of dry soil sieved to 2 mm were amended using the appropriate application rate with each of the amendments. The amendments involved using unaltered hydrated lime or diatomaceous earth, 2 mm sized compost and potting soil, and the 250 µm sized organic fertilizer, magnesium lime, zeolite, bone meal, and crushed horn. For the composts, two different degrees of maturation (i.e., 6 and 8 months qualified as young and mature, respectively) and two amendment rates (i.e., 20 and 40 t ha⁻¹) were tested. The compost was also tested as a mix with 10% of zeolite by mass. Amendment rates were selected to mimic cultural seeding practices that a gardener typically applies. The details are presented in Table 1. The physico-chemical parameters and concentrations of the metal(loid)s studied (As, Cd, Pb, and Zn), covering all the amendments, are presented in Table S1 in the Supplementary Material. All the amendments selected are commercialized and conform to European regulations.

Table 1 Amendment rates (mixed with soil) with applications A1 and A2 used in the experiment chosen according to the cultural seeding and gardening practices and compared to the scientific literature (where doses are based upon maximum doses of a given amendment tested for metal(loid) immobilization).

	Name	A1	A2	Gardening practices	Literature
Young compost (6 months)				0.6%/2 year	33% ^a
Single dose	C6–20	0.6%	1.2%	-	-
Double dose	C6–40	1.2%	2.4%	-	-
Mature compost (8 months)				0.6%/2 year	33% ^a
Single dose	C8–20	0.6%	1.2%	-	-
Double dose	C8–40	1.2%	2.4%	-	-
Composts + zeolite				10:1	-
Young compost + zeolite	C6+Z	0.6% + 0.06%	1.2% + 0.12%	-	-
Mature compost + zeolite	C8+Z	0.6% + 0.06%	1.2% + 0.12%	-	-
Other amendments					
Natural zeolite	Z	2%	4%	1.8%	3% ^b
Organic fertilizer (chicken manure)	OF	0.5%	1%	0.1%	4.5% ^c

Magnesium lime	ML	0.1%	0.2%	0.036%	3% ^b
Hydrated lime	HL	0.05%	0.1%	0.03%	1% ^{b,d}
Diatomaceous earth	DE	0.1%	0.2%	1%	4% ^e
Potting soil (for seeding)	PS	3%	6%	33%	33% ^f
Bone meal	BM	0.05%	0.1%	0.03%	4% ^g
Crushed horn	CH	0.05%	0.1%	0.024%	-
Unamended control	U	-	-	-	-

Calculations are based on dry weights. Gardening practice rates are based on suggested rates of application on commercialized amendment packaging. ^a [58]; ^b [59]; ^c [60]; ^d [61]; ^e [30]; ^f [62]; ^g [63].

Dried soil-amendment mixes were thoroughly mixed, humidified to 60% (pH of tap water at 7.0), and placed into pots. The pots were kept in the dark area at a constant temperature (18°C). The humidity was maintained at 60% of the field water capacity during the experiment. After six weeks of incubation with the first application A1 (Table 1), the soil-amendment mixes were removed from the pots and left to dry at ambient temperature. 150 g was then removed and sieved to 2 mm and ground to 250 µm for further analysis. The samples ($n = 135$) were analyzed as “A1” soils. 300 g of the remaining soil was further re-amended with the same application rate of the amendment (i.e., A2; Table 1), rehumidified to 60% humidity, repotted to continue incubation for eight additional weeks in the same conditions, and dried and ground similarly. These samples ($n = 135$) were analyzed as “A2” soils. The experimental design is presented in Figure S1 in Supplementary Material.

2.3 Amended and Unamended Soil Analysis

2.3.1 Physicochemical Characteristics

The soil pH was measured using a 1:5 volume ratio of 2 mm sieved soil and osmosis water, according to NF ISO 10390. The total carbonate content was obtained by measuring the volume of CO₂ released after a reaction with HCl (NF ISO 10693). The conductivity was measured using a 1:5 ratio of soil sieved to 2 mm and double-distilled water, rotated for 2 h, and then centrifuged at 1000 RPM for 10 min before being measured in the filtrate using a conductivity meter (Cond 330i/SET-2C20-0011) with automatic temperature compensation. The available P (expressed in g P₂O₅ kg⁻¹ of soil) was measured after extraction with an ammonium oxalate solution and spectrophotometric determination (NF X31-161). The cation exchange capacity (CEC) was analyzed according to NF EN ISO 11260 and was then quantified using an AA-6800 Shimadzu atomic absorption spectrometer (AAS).

2.3.2 Degree of Contamination

The pseudo-total metal(loid) concentrations (As, Cd, Pb, and Zn) in the 270 soil subsamples (i.e., unamended and amended with A1 and A2) were obtained using the Hot Block system-assisted digestion (Environmental Express® SC100, Charleston, SC, USA) and determined by AAS for Cd, Pb, and Zn. For As, the analyses were performed using inductively coupled plasma mass spectrometry (ICP-MS, Serie X2, Thermo Scientific) by the INRAE Soil Analysis Laboratory (Arras, France) accredited by COFRAC according to the ISO 17025 standard. More specifically, 300 mg of each

ground sample was digested in a mixture of 1.5 mL HNO₃ (70%) and 4.5 mL HCl (37%) at 95°C for 90 min [64]. For the carbonate-rich CKG-L soil, this step was followed by the addition of 3 mL of 70% HNO₃, which was allowed to react for 1.5 h. This was followed by the addition of 3 mL of hydrogen peroxide, which was allowed to sit for 12 h before being heated at 95°C for 2 h. After mineralization, the digestion products were completed to 25 mL with distilled water (resistivity 10 MΩ cm⁻¹), filtered using a 0.45 μm cellulose acetate filter, and stored at 4°C before analysis. The quality control was based on the use of internal reference material, which demonstrated good recoveries for As, Cd, Pb, and Zn (96.4–100.5%, 99.8–101.3%, 97.8–100.3%, and 88.1–90.2%, respectively).

2.3.3 Predicted Bioavailability of Metal(loid)s

For each of the 270 soil samples (i.e., unamended and amended with A1 and A2), the ammonium nitrate (NH₄NO₃)-extractable metal(loid)s were analyzed using 10 g of soil sieved to 2 mm and agitated with 25 mL of a 1 mol L⁻¹ NH₄NO₃ solution for 2 h. The samples were centrifuged at 1500 g for 10 min and then filtered with a 0.45 μm cellulose acetate filter (NF ISO 19730). The EDTA-extractable metal(loid)s were quantified using 4 g of soil sieved to 2 mm and agitated with 40 mL of a 0.05 mol L⁻¹ EDTA solution with a pH of 7 for 1 h [65]. Samples were then centrifuged at 4500 g for 20 min and filtered with a syringe and 0.45 μm cellulose acetate filter. The metal(loid) concentrations in the extracts were measured using AAS for Cd, Pb, and Zn, using ICP-MS for As. The use of EDTA as a strong extractant (able to chelate ions and dissolve amorphous oxyhydroxides) allows the estimation of the potential influence of these amendments in the long term. It is, however, to be noted that NH₄NO₃-extractable metal(loid)s are more relevant in terms of environmental availability of metallic elements in soils in the short term (as NH₄NO₃ displaces ions from exchange sites).

To compare their origin of contamination more specifically, only the Pb in the soil samples was fractionated using the modified sequential extraction procedure from the SM&T program [66], described by Waterlot et al. [64]. Each fraction was noted as fraction F1, F2, F3, or F4 and defined, respectively as (i) exchangeable, water- and acid-soluble (40 mL of 0.11 mol L⁻¹ acetic acid was added to 1 g of soil sample, and the mixture was shaken at room temperature for 16 h); (ii) reducible (40 mL of 0.5 mol L⁻¹ hydroxylammonium chloride was added to the residue from the previous step and the mixture was shaken for 16 h); (iii) oxidizable (8.8 mol L⁻¹ H₂O₂, followed by 50 mL of 1.0 mol L⁻¹ ammonium acetate at pH 2 and mixing for 16 h); and (iv) residual (*aqua regia*). The Pb concentrations in each fraction were measured using AAS.

2.4 Calculation of Amendment Efficiency

The influence of amendments on metal(loid) extractability (using EDTA and NH₄NO₃) was evaluated by calculating an efficiency factor (EF), which corresponds to the ratio of metal(loid) extractability with an amendment to its extractability without amendment (i.e., control). The amendments were ranked as efficient if the EF values were less than 1 and inefficient if the EF values were greater than 1.

2.5 Statistical Analysis

All the data were analyzed using three replicates of unamended or amended soil. The differences between amended and unamended soils were calculated using Student's t-test or Wilcoxon's test depending upon the parametricity of the data. A one-way ANOVA and a Bonferroni test were utilized to analyze the differences in data between A1 and A2 collected over time. The analyses were performed using XLSTAT 2019.3.2 and Microsoft Excel 2016.0.4966.

3. Results

3.1 Characteristics of the Three Soils Studied

On average, all three soils studied were found to have soil texture ranging from silt loam to loam according to the USDA textural soil classification (Table 2). According to the soils' physico-chemical parameters, the total carbonate and organic matter contents were higher in CKG-L than in PKG and CKG-N, while the available phosphorus content and cationic exchange capacity (CEC) were higher in PKG (Table 2). The pH values were slightly acidic in the PKG and CKG-N soils and slightly alkaline in CKG-L. Regarding the metal(loid)s present in the soils studied, (i) the PKG soil was found to be associated with moderately elevated anthropogenic levels of Cd, Pb, and Zn; (ii) the CKG-N soil detected high geogenic levels of As and Pb; and (iii) the CKG-L soil was associated with elevated point levels of Pb and Zn (Table 2). The median geochemical background values in the topsoil in northern France were found to be 0.4 mg kg⁻¹, 29.7 mg kg⁻¹, and 67.1 mg kg⁻¹, respectively for Cd, Pb, and Zn [67], while the median geochemical values in a sampling of the local site in the topsoil in western France was 29 mg kg⁻¹ and 84 mg kg⁻¹, respectively for As and Pb [68].

Table 2 Physicochemical parameters and degree of contamination of the topsoil in the three sites (PKG, CKG-N, and CKG-L; n = 1).

		PKG	CKG-N	CKG-L
Clay	%	25	4	16
Silt	%	50	60	50
Sand	%	25	36	34
pH _{water}		6.0	6.8	7.7
CaCO ₃ tot	g kg ⁻¹	3	2	119
OM	g kg ⁻¹	51.1	49.6	94.2
C/N		21.3	12.3	32.7
Available P	g kg ⁻¹	1.3	0.7	0.4
CEC	cmol ⁺ kg ⁻¹	32.5	12.6	15.5
As	mg kg ⁻¹	16	32	12
Cd	mg kg ⁻¹	6.3	0.24	0.73
Pb	mg kg ⁻¹	264	238	384
Zn	mg kg ⁻¹	337	77	399

CaCO₃ tot: total carbonates; OM: organic matter; C/N: ratio of organic carbon on total nitrogen; Available P: available phosphorus expressed in g of P₂O₅ per kg of soil; CEC: cationic exchange capacity.

Overall, the three soils presented a similar degree of contamination in terms of Pb (from 238 to 384 mg kg⁻¹). To specifically compare the origin of contamination, sequential extractions were used to evaluate the metal distribution within soils and to provide information on the affinity of Pb to the soil components and the strength with which it is bound to the matrix. The distribution of Pb in fractions F1, F2, F3, and F4 are presented in Figure S2 in the Supplementary Material. Overall, the PKG and CKG-L soils were found to be associated with anthropogenic contamination, and the distribution of metallic elements in the different soil fractions was found to be identical, suggesting similar behavior. Logically, for the PKG and CKG-L soils, for which pseudo-total concentrations of Pb were 264 and 384 mg kg⁻¹, respectively, Pb was primarily found in the reducible fraction (respectively, 77.6% and 78.5%). Negligible amounts of Pb were found in the exchangeable fraction (respectively, 2.6 and 2.3%), in the oxidizable fraction (respectively, 10.3 and 4.0%), and the residual fraction (respectively, 9.5 and 15.0%). For the CKG-N soil (238 mg of Pb kg⁻¹), 21.6% of Pb was linked to Fe/Mn oxides, and the remainder was found in the residual fraction (i.e., 78.4%). This high percentage of Pb in the latter fraction explains the geogenic origin of this contamination.

3.2 Effects of Amendments and Application Rates on Physicochemical Characteristics of Soils

Table 3 presents the effects of the amendments and application rates (A1 and A2) on the physico-chemical characteristics of soils (i.e., pH, electric conductivity, contents of total carbonates, and available phosphorus) for the three soils studied (PKG, CKG-N, and CKG-L).

Table 3 Effects of amendments on the physico-chemical soil characteristics with applications A1 and A2 for the three soils studied (PKG, CKG-N and CKG-L).

	pH		CaCO ₃ (g kg ⁻¹ DW)		EC (μS cm ⁻¹)		P ₂ O ₅ (g kg ⁻¹ DW)	
	A1	A2	A1	A2	A1	A2	A1	A2
PKG								
U	5.87	6.08*	2.27	1.24	1,769	1,716	0.66	0.69
OF	5.86	5.87 §	1.00	2.04 §	1,780	2,208 §	0.71	0.83 §
PS	5.90	6.12*	2.42	1.58	1,750	1,858	0.68	0.71
C6–20	5.85	6.11*	2.04	0.81*	1,781	1,709	0.66	0.77 §
C6–40	5.87	6.11*	2.81	1.74*	1,636	1,686	0.71	0.80 §
C8–20	5.93	6.07*	2.23	1.13	2,285	1,824	0.68	0.75
C8–40	5.84	6.08	2.88	1.95 §	1,783	1,620	0.74	0.78
C6+Z	5.80	6.03*	2.03	1.39	1,562	2,018	0.69	0.75 §
C8+Z	5.86	5.98	1.51 §	1.99 §	1,691	1,871	0.63	0.78*
Z	5.80	6.03*	1.68 §	1.96	2,083	1,597*	0.68	0.72
HL	5.77	6.09*	1.63	1.65 §	2,016	1,618	0.66	0.77
ML	5.87	6.23*	2.11	1.69	1,646	1,681	0.62	0.72
DE	5.96	6.06	2.36	1.62	1,694	1,762	0.63	0.77
BM	5.99	6.11	1.82	1.00	1,605	1,788	0.61	0.74
CH	5.84	6.00*	1.88	1.58	1,629	1,714	0.63	0.69
CKG-N								
U	7.12	7.28*	1.10	1.44	96	98	0.34	0.31
OF	7.30 §	7.40*	1.91 §	1.85	152 §	234 §*	0.44 §	0.60 §*

PS	6.80 §	7.12*	1.10	1.13	192 §	207 §	0.35	0.34
C6–20	7.34 §	7.28	1.17	2.44*	113	109 §	0.36	0.40
C6–40	7.31 §	7.33 §	1.47	2.29	112	140 §*	0.42	0.46 §
C8–20	7.28 §	7.30	1.52	1.36	106	116 §	0.39	0.42 §
C8–40	7.21 §	7.36 §	1.16	1.50	106	152 §	0.42 §	0.53 §
C6+Z	7.29 §	7.31	0.92	1.50	101	120 §	0.38	0.44 §
C8+Z	7.24 §	7.48 §*	1.52	1.06	104	129 §	0.42 §	0.55 §*
Z	7.25 §	7.45 §*	1.47	0.89	89	105	0.39	0.43 §
HL	7.20	7.49 §	1.73 §	1.61	92	104	0.38	0.46 §
ML	7.45 §	7.69 §*	2.20 §	1.23	96	102	0.37	0.47 §
DE	7.30 §	7.44 §*	1.90 §	1.70	92	109 §	0.39	0.47 §
BM	7.13	7.21	1.54	1.39	96	98	0.39	0.42 §
CH	7.14	7.18 §	0.84	0.75	100	84	0.36	0.37 §
CKG-L								
U	7.86	7.91	68.9	75.4	231	149	0.42	0.48*
OF	7.66 §	7.71 §	69.5	71.8 §	433 §	340 §*	0.54 §	0.49
PS	7.78	7.60 §*	68.3	73.0	270	326 §	0.48	0.40 §*
C6–20	7.68 §	7.85*	51.6 §	72.7*	239	196 §	0.47 §	0.48
C6–40	7.70 §	7.89*	60.8	73.7*	272	255 §	0.49 §	0.51
C8–20	7.70 §	7.88*	67.2	76.4*	258	201 §	0.47 §	0.51
C8–40	7.70 §	7.86*	60.4 §	74.4*	337 §	271 §*	0.49 §	0.58 §*
C6+Z	7.73 §	8.00*	67.1	73.1	250	201 §	0.48 §	0.47
C8+Z	7.79	7.77 §	67.1	74.1	238	211 §	0.50 §	0.56*
Z	7.89	7.79	66.6	71.5 §	204	211 §	0.47 §	0.44
HL	7.79	7.79	68.6	74.0	235	177	0.49 §	0.42
ML	7.88	8.09 §	71.1	76.2	262	174	0.49 §	0.44*
DE	7.76	7.62 §*	67.7	77.1	258	190 §	0.48 §	0.43
BM	7.85	7.84	68.2	76.2	219	154	0.49 §	0.50
CH	7.78	7.78	72.9	77.8	266	172	0.44	0.52*

DW: dry wet; P₂O₅: available phosphorus; EC: electric conductivity; U: unamended soil; OF: organic fertilizer; PS: potting soil; C6–20: young compost (6 months) at 20 t ha⁻¹; C6–40: young compost (6 months) at 40 t ha⁻¹; C8–20: mature compost (8 months) at 20 t ha⁻¹; C8–40: mature compost (8 months) at 40 t ha⁻¹; C6+Z: a mixture of young compost and zeolite; C8+Z: a mixture of mature compost and zeolite; Z: zeolite; HL: hydrated lime; ML: magnesium lime; DE: diatomaceous earth; BM: bone meal; CH: crushed horn. § denotes significant differences between the unamended control soil and a given amended soil, with A1 and A2, and * denotes a significant change between A1 and A2 ($p < 0.05$).

For each experiment conducted with A1 and A2, the pH values in the unamended control soils (U) were on average 6.0 for PKG, 7.2 for CKG-N, and 7.9 for CKG-L. Compared to U, the results for the PKG soil showed: (i) no significant change in pH associated with amendment addition with A1 and (ii) the addition of organic fertilizer (OF) was associated with a significant decrease in pH with A2 (0.2 pH units). For the CKG-N soil, the results highlighted: (i) with A1, a significant decrease (0.3 pH units) in pH with the addition of potting soil (PS). In contrast, OF, C6, and C8 composts (for both

20 and 40 t ha⁻¹) alone and with zeolite (Z), Z, magnesium lime (ML), and diatomaceous earth (DE) were associated with a significant increase in pH (between 0.1 and 0.2 pH units); and (ii) with A2, PS was associated with a significant decrease in pH (0.1 pH units), while a significant increase was observed with the addition of C8+Z, Z, HL, and ML (between 0.2 and 0.4 pH units). In terms of the CKG-L soil, the results showed (i) a significant decrease (about 0.2 pH units) in pH with the addition of C6 and C8 (for both 20 and 40 t ha⁻¹), C6+Z, and OF with A1, which corresponds to all amendments with high organic matter content and (ii) a significant decrease (between 0.2 and 0.3 pH units) in pH associated with the addition of OF, PS, C8+Z, and DE, and a significant increase (about 0.2 pH units) in pH was observed with the addition of ML with A2. Most of the changes in pH were observed in the soils, which were initially in the basic pH range. Moreover, composts tended to decrease the pH in the three soils with A1. Soil pH increased significantly between A1 and A2 with the addition of some amendments to the PKG soil (PS, C6–20, C6–40, C8–20, C6+Z, Z, HL, ML, and CH), CKG-N (OF, PS, C8+Z, Z, ML, and DE) and the CKG-L soil (PS, C6–20, C6–40, C8–20, C8–40, C6+Z, and DE). A slight increase in pH was observed in the control soils in the cases of PKG and CKG-N soils. This may be attributed to the water used to maintain the soil at a stable humidity over time. For CKG-L, the most alkaline soil, no change was associated with the irrigation process.

Regarding the electric conductivity (EC) in the control U soils, the values were approximately 1,700 $\mu\text{S cm}^{-1}$, 100 $\mu\text{S cm}^{-1}$, and 200 $\mu\text{S cm}^{-1}$, respectively for PKG, CKG-N, and CKG-L (Table 3). The addition of amendments to the PKG soil showed: (i) no significant change in the EC with A1; (ii) a significant increase in the EC associated with OF with A2; and (iii) a significant decrease in the EC between A1 and A2 of Z. For CKG-N, the results highlighted: (i) that the addition of OF and PS with A1 was associated with a significant increase in the EC in comparison to the control; (ii) a significant increase with A2 in the EC with the addition of OF, PS, and C6 and C8 composts alone and with Z, and DE and (iii) a significant increase in the EC between A1 and A2 with an increased dose of OF and C6–40. Regarding the CKG-L soil, the results showed (i) a significant increase in the EC associated with the addition of OF and C8–40 with A1; (ii) a significant increase with A2 in the EC associated with the addition of OF, PS, C6, and C8 alone and with Z, Z, and DE, and (iii) a significant decrease in the EC over the time associated with an increased dose of OF and C8–40. The most significant changes in the EC were observed in the soils having a lower EC (i.e., CKG-N and CKG-L). More specifically, the increases in EC may be linked to the organic matter content, specifically with the addition of compost, chicken manure, and potting soil.

In the control U soils, the mean content of total carbonates was relatively low for PKG, with 1.8 g kg⁻¹, and for CKG-N, with 1.3 g kg⁻¹, while the values were higher for CKG-L, with 72 g kg⁻¹. When compared to U, the results for the PKG soil showed (i) a significant decrease in the CaCO₃ content associated with the addition of C8+Z and Z with A1 but a significant increase in terms of A2 with OF, C8–40, C8+Z, and HL; and (ii) a significant decrease in the CaCO₃ content with increased amendment doses of C6 (for both 20 and 40 t ha⁻¹). For the CKG-N soil, it was observed that (i) the addition of OF, HL, ML, and DE with A1 resulted in a significant increase in the CaCO₃ in comparison to the control; (ii) addition of amendments with A2 did not show any significant changes in the CaCO₃ content; and (iii) a significant increase in the CaCO₃ was observed overtime linked to an increased dose of C6–20. Regarding the CKG-L soil, the results showed (i) a significant decrease in the CaCO₃ content associated with the addition of C6–20 and C8–40 with A1 (ii) that the addition of OF and Z resulted in a significant decrease in the CaCO₃ content with A2 and (iii) a significant increase over time with an increased dose of C6 and C8 (for both 20 and 40 t ha⁻¹).

Regarding the content of available phosphorus in the control soils, the values were approximately 0.7 g kg^{-1} , 0.3 g kg^{-1} , and 0.4 g kg^{-1} , respectively, for PKG, CKG-N, and CKG-L (Table 3). More specifically, the addition of amendments to PKG showed (i) no significant changes in the P_2O_5 content with A1; (ii) a significant increase in the available phosphorus with the addition of OF, C6–20, C6–40, and C6+Z with A2; and (iii) a significant increase between A1 and A2 with increased C8+Z in P_2O_5 in comparison to the control. For CKG-N, the results highlighted that (i) the addition of OF, C8–40, and C8+Z with A1 resulted in a significant increase in the P_2O_5 content; (ii) the addition of all the amendments (except PS and C6–20) with A2 was associated with a significant increase in the available phosphorus and (iii) a significant increase in P_2O_5 was associated with an increased dose of OF and C8+Z. With CKG-L, the results showed (i) a significant increase with A1 in P_2O_5 with the addition of all the amendments (except PS and CH) (ii) a significant increase in P_2O_5 with A2 associated with an increased dose of C8–40 and C8+Z, but a significant decrease with PS and (iii) a significant increase associated with an increased dose of PS, C8–40, C8+Z, ML, and CH.

3.3 Effects of Amendments and Application Rates on Extractability of Metal(loid)s

3.3.1 EDTA-Extractable Metal(loid)s

Figure 1 presents the extractability of metal(loid)s by EDTA in the three unamended and amended soils studied (PKG, CKG-N, and CKG-L) considering the two applications of amendments (A1 and A2). The extractability of the metal(loid)s studied in the control soils (i.e., unamended soils U) were expressed as a percentage of the pseudo-total concentrations and were approximately (i) 70%, 66%, and 50%, respectively for Cd, Pb, and Zn in the PKG soil (ii) 7%, 14%, and 17%, respectively for As, Pb, and Zn in the CKG-N soil and (iii) 48% and 14% for Pb and Zn, respectively in the CKG-L soil. Overall, the results showed that the extractability of the geogenous Pb (in CKG-N) was lower (14%) compared to the anthropogenic Pb (in PKG and CKG-L), where the percentages were 66% and 48%, respectively. Compared to the control sample for the PKG soil, the results showed: (i) a significant increase in extractable Cd in the case of A1 and A2 with C6 (20% at both 20 and 40 t ha^{-1}), and only A1 with CH (16%), and DE (14%); (ii) no significant change with A1 in the Pb extractability associated with amendment addition, but a slight decrease with A2 accompanied with the addition of OF and PS (of 11% and 9%, respectively) and (iii) that the addition of C6–20 (with A1 and A2) and ML (only with A2) was associated with a significant increase in extractable Zn (20% and 9%, respectively), while PS was linked to a decrease of 6%. For the CKG-N soil, the addition of PS, C6+Z, Z, DE, and BM with A1 was associated with a significant decrease (between 13% and 68%) in extractable As, while no significant change in the Pb or Zn extractability with any other amendment. However, several changes were recorded in the case of A2 for this soil: (i) a significant increase of As extractability associated with the addition of OF, C6–40, C8–20, C6+Z, C8+Z, Z, ML, and CH (between 38% and 86%), but a decrease by 63% with the addition of PS; and (iii) an increase (between 22% and 42%) in Zn extractability associated with the addition of C6 and C8 at 40 t ha^{-1} and C8+Z. For the CKG-L soil, no significant change was observed in Pb extractability with the addition of amendments with A1 and A2. On the other hand, the addition of C6–20 and C6–40 with A1 and that of OF, C8–20, C8–40, HL, and ML with A2 demonstrated an increase between 12% and 25% in Zn extractability.

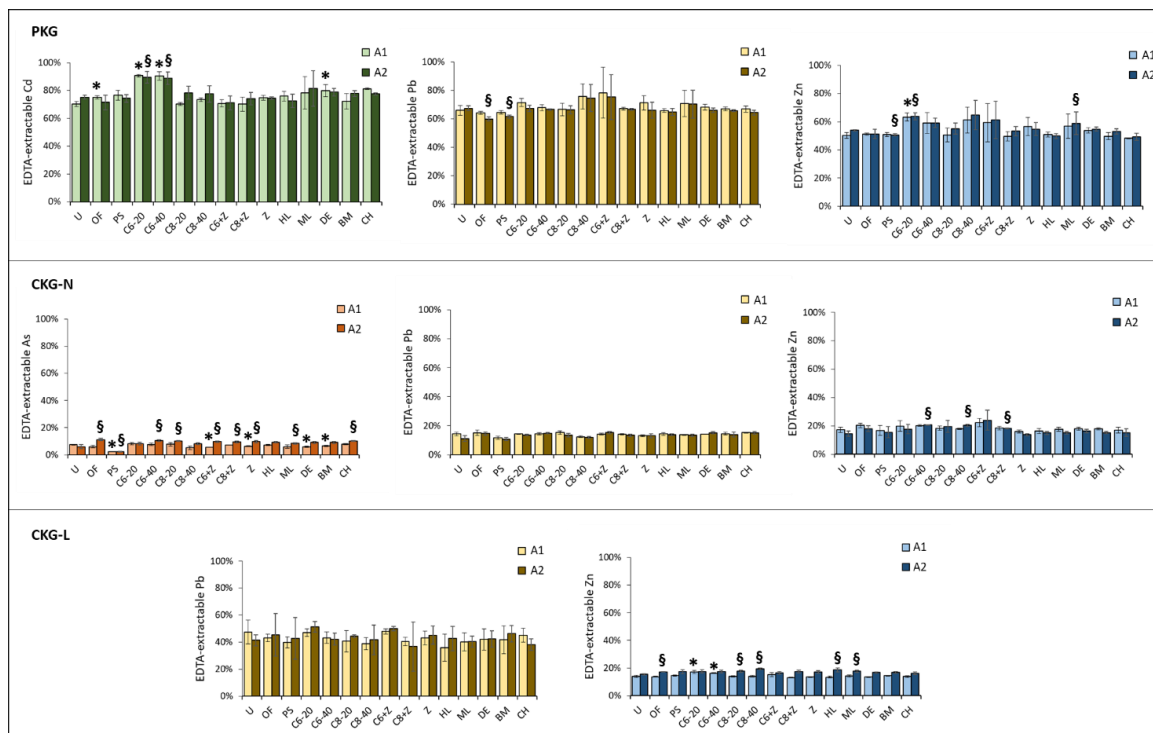


Figure 1 Comparison of metal(loid)s’ extractability by EDTA for several amendments at application rates A1 and A2 for the three soils studied (PKG, CKG-N, and CKG-L). U: unamended soil; OF: organic fertilizer; PS: potting soil; C6–20: young compost (6 months) at 20 t ha⁻¹; C6–40: young compost (6 months) at 40 t ha⁻¹; C8–20: mature compost (8 months) at 20 t ha⁻¹; C8–40: mature compost (8 months) at 40 t ha⁻¹; C6+Z: a mixture of young compost and zeolite; C8+Z: a mixture of mature compost and zeolite; Z: zeolite; HL: hydrated lime; ML: magnesium lime; DE: diatomaceous earth; BM: bone meal; CH: crushed horn. * and § denote significant differences between a given amended soil and the unamended control soil, respectively with A1 and A2 ($p < 0.05$).

3.3.2 NH₄NO₃-Extractable Metal(loid)s

Figure 2 presents the extractability of metal(loid)s as evaluated by an NH₄NO₃-extraction performed on the three unamended and amended soils studied (PKG, CKG-N, and CKG-L) with the application of A1 and A2. Overall, in the control soils (i.e., unamended soils), the NH₄NO₃-extractable metal(loid) concentrations were very low. The extractability of As, Cd, Pb, and Zn is expressed as a percentage of the pseudo-total concentrations.

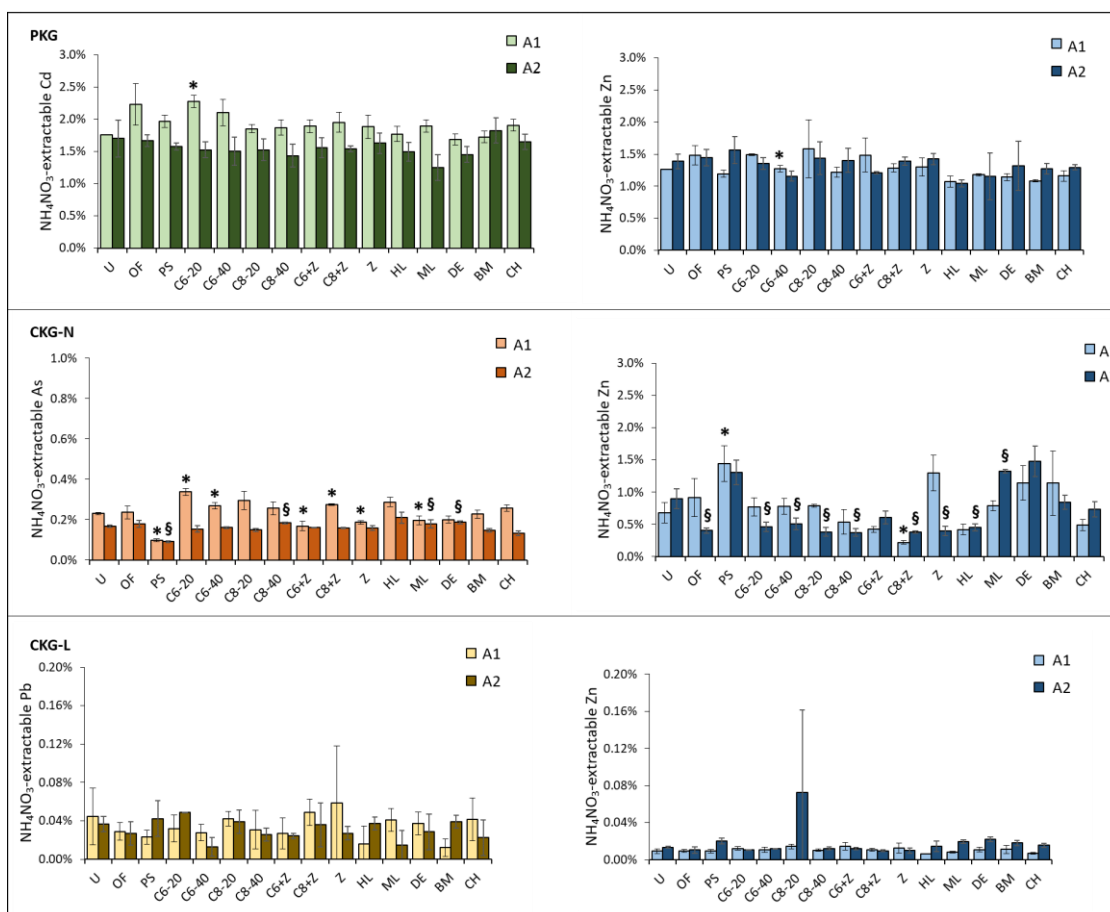


Figure 2 Comparison of metal(loid)s' extractability by ammonium nitrate for the amendments studied at applications A1 and A2 for the three soils studied (PKG, CKG-N, and CKG-L). U: unamended soil; OF: organic fertilizer; PS: potting soil; C6–20: young compost (6 months) at 20 t ha⁻¹; C6–40: young compost (6 months) at 40 t ha⁻¹; C8–20: mature compost (8 months) at 20 t ha⁻¹; C8–40: mature compost (8 months) at 40 t ha⁻¹; C6+Z: a mixture of young compost and zeolite; C8+Z: a mixture of mature compost and zeolite; Z: zeolite; HL: hydrated lime; ML: magnesium lime; DE: diatomaceous earth; BM: bone meal; CH: crushed horn. * and § denote significant differences between a given amended soil and the unamended control soil, respectively with A1 and A2 ($p < 0.05$).

The extractability of metal(loid)s were recorded as approximately: (i) 1.8% and 1.3%, respectively for Cd and Zn in the PKG soil (ii) 0.2% and 0.7%, respectively for As and Zn in the CKG-N soil and (iii) 0.04% and 0.009%, respectively for Pb and Zn in the CKG-L soil. The results of Pb extractability in the CKG-N and PKG soils are not presented because the extractable values by NH₄NO₃ were under the limit of detection.

Compared to the unamended PKG soil (control), the results showed no significant change in Cd or Zn extractability associated with the cases of both A1 and A2. However, there was an exception in the case of A1 with C6–20 for Cd and C6–40 for Zn. For CKG-N, the addition of amendments was associated with (i) a significant increase (between 6% and 46%) in extractable As with C6 (for both 20 and 40 t ha⁻¹), and C8+Z with A1 and with C8–40, ML and DE with A2. PS was associated with a significant decrease (of 50% on average) in the case of both applications; and (ii) a 100% increase in extractable Zn with PS and a significant decrease of 67% with C8+Z in the case of A1. In the case of

A2, ML was associated with a significant increase (47%) in extractable Zn, and OF, C6, C8 (for both 20 and 40 t ha⁻¹), C8+Z, Z, and HL were associated with a significant decrease (between 43% and 58%). For the CKG-L soil, no significant changes were observed in Pb or Zn extractability in the cases of both A1 and A2, whatever the type of amendment studied.

3.4 Efficiency of Amendments on Metal(loid) Extractability

The influence of the applied amendments was evaluated by the calculation of an efficiency factor (EF). An amendment was classified as efficient if it reduced the metal(loid) extractability in comparison to its given control soil, thereby being assigned an EF value of less than 1. Inefficient amendments are, on the other hand, associated with EF values greater than 1. Figure 3 presents both the efficiency and inefficiency of amendments with regards to the EDTA- and NH₄NO₃-extraction of metal(loid)s for the three soils. The focus was primarily on the efficiency of amendments at reducing EDTA- and NH₄NO₃-extractable metal(loid)s.

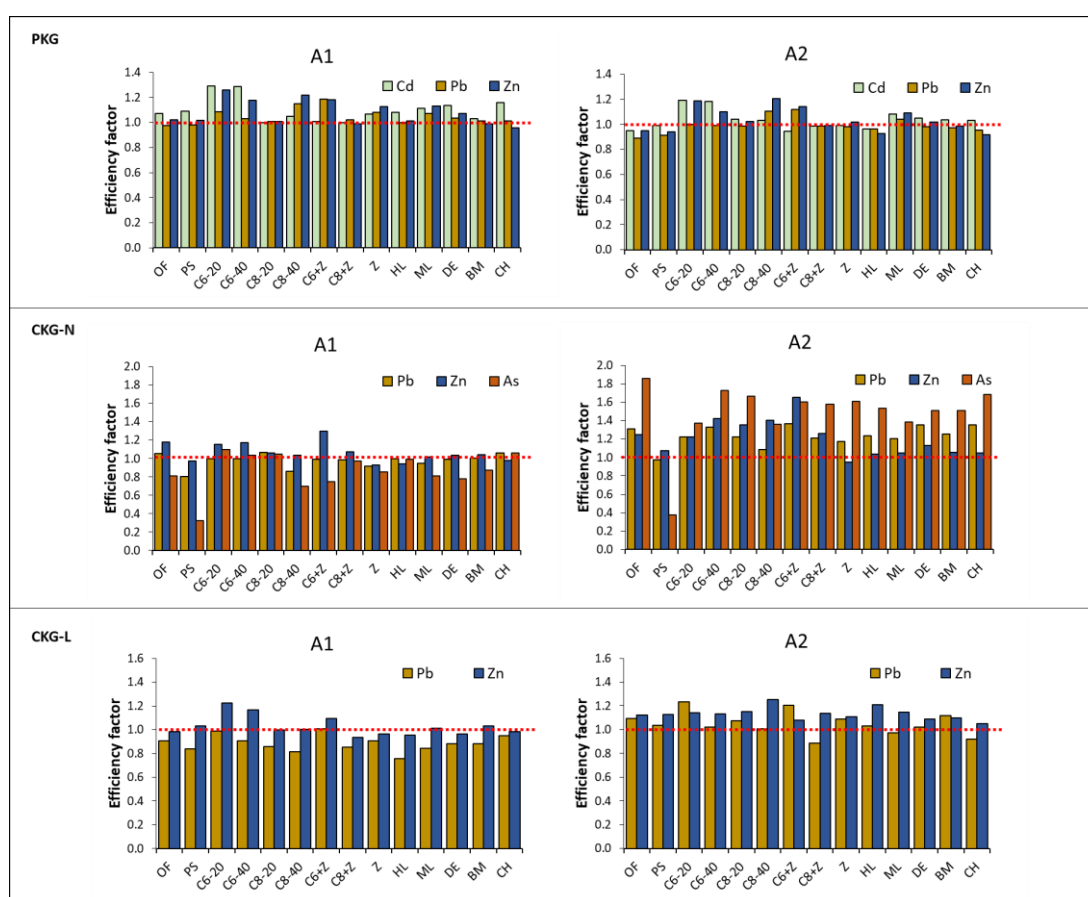


Figure 3 Immobilization efficiency of amendments on EDTA-extractable metal(loid)s for the three soils studied (PKG, CKG-N, and CKG-L); OF: organic fertilizer; PS: potting soil; C6–20: young compost (6 months) at 20 t ha⁻¹; C6–40: young compost (6 months) at 40 t ha⁻¹; C8–20: mature compost (8 months) at 20 t ha⁻¹; C8–40: mature compost (8 months) at 40 t ha⁻¹; C6+Z: a mixture of young compost and zeolite; C8+Z: a mixture of mature compost and zeolite; Z: zeolite; HL: hydrated lime; ML: magnesium lime; DE: diatomaceous earth; BM: bone meal; CH: crushed horn. Amendments were ranked as efficient if EF < 1 and inefficient if EF > 1.

Regarding the EDTA-extraction for the PKG soil (Figure 3), (i) none of the amendments resulted in a significant efficiency at reducing both Cd and Pb extractability in the case of A1 (EF between 1.0 and 1.3), while with A2, OF, C6+Z and HL were associated with relatively less extractable Cd (EF of 0.9 on average) and OF, PS, HL, DE, BM, and CH resulted in relatively less extractable Pb (EF of 0.9 on average) (ii) for Zn, in the case of A1, only the addition of CH allowed a reduction in its extractability (EF of 0.9), while in the case of A2, OF, PS, HL, BM, and CH resulted in less extractable Zn (EF of 0.9 on average). For the CKG-N soil, it was observed that PS, C8–40, Z, and ML with A1 were efficient (EF between 0.8 and 0.9), and their addition was associated with less extractable Pb, while with A2, none of the amendments resulted in significant efficiency at reducing Pb extractability (EF between 1.0 and 1.4). A1 with PS, Z, HL, and CH, resulted in decreased extractability (EF of 0.9 on average) in the case of Zn, while with A2, only Z was associated with relatively less extractable Zn (EF of 0.9). The amendments that efficiently reduced extractable As with A1 included OF (EF of 0.8), PS (EF of 0.3), C8–40 (EF of 0.7), C6+Z (EF of 0.9), Z (EF of 0.9), ML (EF of 0.8), DE (EF of 0.8), and BM (EF of 0.9), while only PS reduced extractable As in the case of A2 (with EF of 0.4). In the case of A1 in the CKG-L soil, the addition of all the amendments other than C6–20 and C6+Z was associated with efficiency at reducing the extractable Pb (EF between 0.7 and 0.9). A2 with only C8+Z and CH resulted in a similar reduction. In the case of Zn with A1, the addition of C8+Z, Z, HL, and DE resulted in less extractable Zn (EF of 0.9 on average). However, none of the amendments were efficient after the second application (EF between 1.0 and 1.3).

Regarding the efficiency factors for PKG from the NH_4NO_3 -extraction (Figure 4), only DE and BM in the case of A1 are associated with relatively less extractable Cd (EF of 0.9), while with A2, the addition of all the amendments except BM reduced Cd extractability (EF between 0.7 and 0.9). For Zn, several amendments with both doses seemed efficient to reduce its extractability, more specifically PS, C6–40, C8–40, HL, ML, DE, BM, and CH with A1 (EF of 0.9 on average), and C6–20, C6–40, C6+Z, HL, ML, DE, BM, and CH with A2 (EF between 0.8 and 0.9). For the CKG-N soil (Figure 4), the addition of C8–40 (EF of 0.8), C6+Z (EF of 0.6), C8+Z (EF of 0.3), HL (EF of 0.6), and CH (EF of 0.7) with A1 was associated with less extractable Zn. A2 with OF (EF of 0.4), composts with and without zeolite (EF of 0.5 on average), Z (EF of 0.4), HL (EF of 0.5), BM (EF of 0.9), and CH (EF of 0.8) also efficiently reduced extractable Zn. In the case of A1 with PS (EF of 0.4), C6+Z (EF of 0.7), and to a lesser extent Z, ML, and DE (with EF between 0.8 and 0.9), and the A2 of PS (EF of 0.5), and to lesser extent C6–20, C6–40, C8–20, C6+Z, C8+Z, Z, BM, and CH (EF between 0.8 and 0.9) reduced extractable As. For CKG-L (Figure 4), all amendments with A1 (excepted C8+Z and Z) were efficient at reducing Pb extractability (EF between 0.3 and 0.9). In the case of A2, several amendments were found to be efficient, more specifically OF, C6–40, C8–40, C6+Z, Z, ML, DE, and CH (EF between 0.4 and 0.9). For Zn, A1 with only HL, ML, and CH resulted in less metal extracted (EF of 0.8 on average), while A2 with OF, C6–20, C6–40, C8–40, C6+Z, C8+Z, and Z reduced extractable Zn (EF between 0.7 and 0.9).

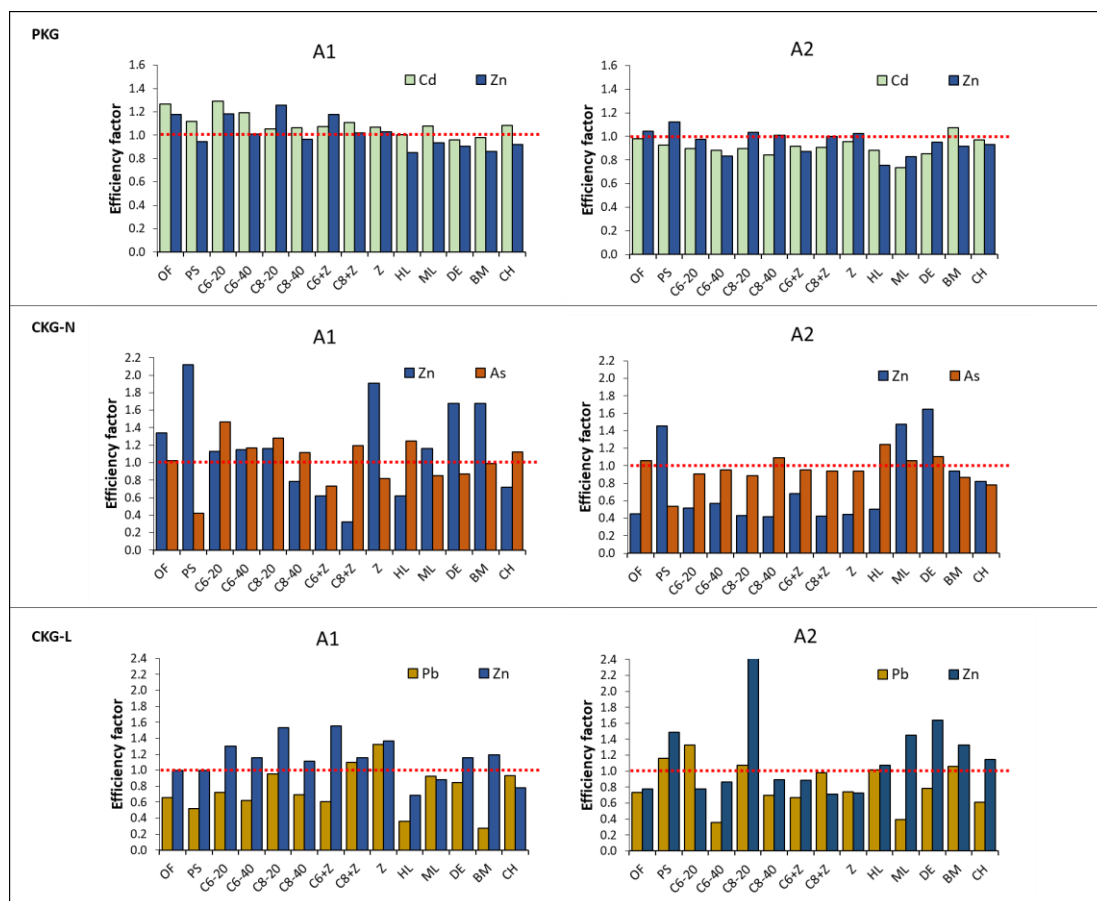


Figure 4 Immobilization efficiency of amendments on NH_4NO_3 -extractable metal(loid)s for the three soils studied (PKG, CKG-N, and CKG-L); OF: organic fertilizer; PS: potting soil; C6–20: young compost (6 months) at 20 t ha^{-1} ; C6–40: young compost (6 months) at 40 t ha^{-1} ; C8–20: mature compost (8 months) at 20 t ha^{-1} ; C8–40: mature compost (8 months) at 40 t ha^{-1} ; C6+Z: a mixture of young compost and zeolite; C8+Z: a mixture of mature compost and zeolite; Z: zeolite; HL: hydrated lime; ML: magnesium lime; DE: diatomaceous earth; BM: bone meal; CH: crushed horn. Amendments were ranked as efficient if $\text{EF} < 1$ and inefficient if $\text{EF} > 1$.

4. Discussion

This study aimed to evaluate the effects of soil management practices based on the use of amendments on three contaminated kitchen garden soils under *ex-situ* conditions. Particular attention was paid to the influence and efficiency of organic and inorganic amendments with different application rates on metal(loid) extractability. The novelty of the study is based on (i) the comparison of a collection of amendments on soils with contrasting characteristics and sources of contamination and (ii) the use of doses commonly used by gardeners.

4.1 Organic Amendments

The organic amendment studied comprised of a commercialized green waste compost with two degrees of maturity. In terms of EDTA extractability of metal(loid)s for all three soils, A2 with composts is associated with a statistically significant decreased amendment efficiency in terms of

metal(loid) reduction. These doses correspond to at least double that of what is recommended by gardening practices. This trend is echoed for extractable As in the CKG-N soil. Regarding the extractability of NH_4NO_3 , it was observed that for all applications of composts, there was an increase in the available As in the CKG-N soil in comparison to the unamended soil. Though compost amendments are globally inefficient for this metalloid and soil, it was improved upon with the higher dose. Notably, the addition of compost amendments was also associated with a significant increase in pH in comparison to the control with A1 for the As-contaminated CKG-N soil and with A2 for C6–40 and C8–40. Though classified as an inefficient amendment for As immobilization in the CKG-N soil, the higher doses of some composts significantly decreased Cd availability in the PKG soil in comparison to A1. It has been shown that organic humidified materials can immobilize metals via adsorption or complexation, as the increase in soil organic matter content increases binding sites, thereby potentially reducing metal(loid) availability in soils [69]. OM increases Cd stability via adsorption and/or organic-metal complex processes, which can be improved with the addition of organic amendments, converting soluble and exchangeable fractions of metals to the less-available organic-bound fractions [70-72]. Additionally, the functional groups in organic materials show a high affinity for Cd ions [73-75]. For the CKG-L soil, many of the efficient, high-OM amendments with both A1 and A2 are associated with significant decreases in pH in comparison to the control soil, which had an alkaline pH of 7.9. However, the addition of C8–40 was associated with a decrease in NH_4NO_3 -extractable Pb and Zn and significant increases in EC for this soil. A study by Alvarenga et al. [76] found that various soil amendments led to significant increases in the EC values, particularly for composts, which effectively reduced CaCl_2 -extractable Cu, Pb, and Zn. CaCl_2 is an extractant that acts upon weaker bonds such as that of NH_4NO_3 .

4.2 Natural Fertilizers

Three organic fertilizers were considered in this study—crushed horn (CH), bone meal (BM), and organic complete fertilizer made from poultry manure (OF).

CH is a bovine byproduct rich in keratin, N, P, K, and Mg, which decomposes slowly over time. The application of CH effectively reduced the extractable Pb and Zn from the soils in this study. This product, when hydrolyzed, has been associated with the reduction of available aqueous Cd up to 50% depending on its particle size [77]. This effect was attributed to the production of peptides, oligopeptides, and free amino acids with strong ion binding capacities upon hydrolysis. However, there is limited research and information on the effects of CH on soil metal(loid)s. However, the effect of the degradation of keratin complexed with the pollutants (i.e., the release of pollutant-bound keratin) in the medium term is also to be noted

BM has a composition close to CH and is a mixture of finely and coarsely ground animal bones and slaughterhouse waste products, rich in phosphorus and calcium. BM efficiently reduced the metal(loid)s (As, Pb, and Zn) with both applications when evaluated with the NH_4NO_3 extraction in this study. Similarly, a study by Sneddon et al. [78] found that the (relatively weak) CaCl_2 (0.01 M) extractions predicted a reduction in more metal availabilities than DTPA extractions conducted on the same BM-treated soils. Generally, DTPA extracts more metal from soils than CaCl_2 , and this trend is the same for EDTA and NH_4NO_3 extractions, respectively. Thus, the results support the fact that BM can reduce the concentration of metal(loid) ions held on exchange sites, more so than on chelatable metals. BM demonstrated varied results for the different soils on reducing EDTA-

extractable Pb, As, and Zn. The effective reduction of Pb in the CKG-L soil may be linked to sorption processes and a subsequent formation of metal phosphate, which retains Pb in the soil [79]. Other studies have suggested that bone char apatite, another source of phosphate, could remediate metal-contaminated soils, causing excessive P runoff [80, 81]. This amendment removes metals using adsorption and promotes ion-exchange reactions between ions in soil solution and calcium ions [82]. However, because of the low application rate of BM and the limited time of contact with the soils, the formation of a metal-phosphate phase is unlikely to be due to substitution, an ionic process that typically takes place slowly [79].

OF, another amendment rich in organic matter and N, P, K, resulted in a significant reduction of Cd in the case of A2 in the PKG soil, and Pb and Zn in the case of A1 in the CKG-L soil. Organic matter-rich amendments immobilize the metal(loid)s in the soil by increasing the pH, negatively charged surface sorption areas, and by forming hydroxyl forms of metal cations due to a subsequent increase in soil CEC [80, 81]. Specifically, organic fertilizer in the form of chicken manure is rich in humic substances, which reduces metal(loid) availability via adsorption or the formation of stable organic fractions [83, 84]. Chicken manure has been demonstrated to effectively reduce metals, especially Cd, alone and in composted manure [59, 72, 85, 86].

4.3 Calcareous Amendments

Two calcareous amendments (calcium-containing inorganic minerals composed primarily of oxides and hydroxides, usually calcium oxide and/or calcium hydroxide) were investigated, including lime flower (i.e., hydrated lime, HL) and magnesium lime (ML). Both applications (A1, A2) of the limes (HL, ML) were effective in immobilizing Cd, As, Pb, and Zn in the soils and were the most efficient among the amendments studied for the three soils tested. The addition of lime to soil can result in the precipitation of exchangeable soil Al, which can act as a cementing agent that binds soil particles, and improves soil structure. Liming can also significantly increase the concentration of exchangeable Ca and Mg in soils, a reaction that leads to an increase in CEC and the content of available soil phosphorous. Notably, lime can influence and increase soil pH, which is linked with the reduction of various bioavailable metal(loid)s, including Cd and Zn, via complexation and precipitation [87-92]. However, contrary to what was observed in this study, As is fairly available at the pH range in which other metals such as Cd and Pb may be immobilized [93].

4.4 Natural Siliceous and Alumino-Silicate Amendments

Two natural siliceous or alumino-silicate amendments were selected, including diatomaceous earth (DE) and natural zeolite (Z). DE demonstrated some success in reducing the extractability of As and Pb. A significant decrease in soil pH (from 7.9 to 7.6) was associated with the second application of DE in the CKG-L soil. This decrease may be due to the accumulation and retention of carbon dioxide (CO₂) and the creation of carbonic acids in the soil in the presence of moisture, which is retained by diatomite. A study by Prakash et al. [94] found that rates of application of diatomite as small as 0.3 t ha⁻¹ result in a decrease in pH in alkaline soils and an increase in acidic soils at field water capacity. DE, a highly porous silicate, has a high specific surface area and is often used by gardeners as a pest deterrent and means of increasing soil water retention and improving soil quality. A study by Lu et al. [95] found that doses of calcium silicate, sodium silicate, and potassium silicate (1% by weight each) effectively reduced the amount of Cd accumulated in kale grown in

garden soil (control) with a slightly acidic pH. This was similarly accurate for a 1.5% dose of silicate slag tested on cabbage for Cd uptake [96]. However, this was attributed to increased Cd adsorption by amended soil, leading to decreased Cd uptake by vegetables into the soil solution due to the increase in relative dissolved concentrations of cations competing with Cd^{2+} . In terms of soil Pb, studies have shown that active silicate materials can be useful in remediating Pb-polluted soils, as they can transform lead into oxidizable and residual soil fractions at neutral pH [97]. Little is known about the effects of silicates, particularly diatomaceous earth, on soil Pb and As.

Zeolites were successful on various metal(loid)s, at given applications and effectively reduced available As and Zn in the case of A1 and A2, respectively, in the CKG-N soil. Zeolites (Z) that contain calcium oxide and/or sodium hydroxide can also increase soil pH [98, 99]. Specifically, minerals added to the soil by zeolites can increase a soil's pH and CEC, and prevent the decrease in organic matter [100]. Similar studies have found that sepiolite (a magnesium hydro silicate clay mineral) significantly decreased HCl-extractable Cd due to an increase in soil pH, Cd complexation reactions, Cd diffusion into the clay lattice structure, and Cd substrate surface retention [101, 102]. At a pH value between 4 and 6, like in the PKG soil, zeolites are typically cited as immobilizing metals mainly through ion exchange [103]. Other studies cite sorption as the main mechanism that occurs at moderately alkaline pH's, like those seen in the CKG-L and CKG-N soils [98].

4.5 Mixtures Containing Organic Products

Two mixtures of products were tested—potting soil (PS) and a mixture of young or mature composts with zeolite (C6+Z and C8+Z). Among the amendments that were found to be efficient with both extractants, PS showed a statistically significant potential in reducing the availability of As and Pb with both applications, making it a potentially effective tool for risk reduction in As- and Pb-pollution kitchen garden soils (CKG-N). For the CKG-L soil, a significant decrease in soil pH was associated with the addition of the second application of PS. The addition of 6% PS by mass could result in the decrease of soil pH by way of dilution. Potting soils often consist of a mixture of peat, dolomite, vermiculite, or perlite, and are designed to hold water and nutrients in the soil. A typical mix contains 33–66% of peat rich in organic matter [104]. Peat and vermiculite are cited as heavy metal immobilizers [105, 106]. The addition of 3% vermiculite and peat separately to contaminated and slightly acidic soil can provoke a reduction in water-soluble and extractable soil Cd and Pb [106]. Vermiculite, particularly that which has been modified by cationic surfactants, has also been cited as an effective sorbent of As(V) and As(III) in aqueous solutions [107]. The addition of PS at 33% by mass resulted in the reduction of available Pb in garden soil with acidic pH, but an increase in available As [62]. The observed reduction in the available Pb could be due to mechanisms of complexation and adsorption but also due to dilution at the high application dose. In this study, PS very effectively reduced extractable As when applied at 3% (A1) and 6% (A2) by mass to alkaline soil. However, since the constituents of PS were not tested separately in this experiment, it is difficult to determine if it was the entire mixture or a single constituent that demonstrated the effects.

Young or mature composts mixed with zeolite were overall determined as efficient amendments as evaluated by both extractants. C6+Z with A1 significantly reduced available As in comparison to the control soil, and C6+Z and C8+Z with A2 reduced extractable Cd in the PKG soil. The application of C8+Z also reduced extractable Pb in the CKG-L soil. To reiterate, zeolites are natural or synthetic crystalline frameworks of alkaline-based aluminosilicates that can act as molecular sieves because

of their adsorption capacity [98, 103]. They are linked to metal(loid) immobilization in conjunction with composts and have an immobilizing effect when applied to soil alone [57].

Table 4 presents a summary of the amendments applied on the three soils having the efficiency to reduce the extractability of metal(loid)s.

Table 4 Summary of the amendments for both applications A1 and A2 on the three soils studied (PKG, CKG-N, and CKG-L) having an efficiency (i.e., EF<1) to reduce the extractability of metal(loid)s.

Soil	Metal(loid)	Dose	Amendment
PKG	Cd	A1	DE, BM
		A2	OF, PS, C6–20, C6–40, C8–20, C8–40, C6+Z, C8+Z, Z, HL, ML, DE, CH
	Pb	A1	-
		A2	OF, PS , HL, ML*, DE, BM, CH
	Zn	A1	PS, C6–40*, C8–40, HL, ML, DE, BM, CH
		A2	OF, PS , C6–20, C6–40, C6+Z, HL, ML , DE, BM, CH
CKG-N	As	A1	OF, PS* , C8–40, C6+Z* , Z* , ML*, DE, BM
		A2	PS* , C6–20, C6–40, C8–20, C6+Z, C8+Z, Z, BM, CH
	Pb	A1	PS, C8–40, Z, ML
		A2	-
	Zn	A1	PS, C8–40, C6+Z, C8+Z*, Z, HL, CH
		A2	OF*, C6–20*, C6–40*, C8–20*, C8–40*, C6+Z, C8+Z*, Z*, HL*, BM, CH
CKG-L	Pb	A1	OF, PS, C6–20, C6–40, C8–20, C8–40, C6+Z, C8+Z, Z, HL, ML, DE, BM, CH
		A2	OF, C6–40, C8–40, C6+Z, C8+Z, Z, ML, DE, CH
	Zn	A1	C8+Z, Z, HL, ML, DE, CH
		A2	OF, C6–20*, C6–40*, C8–40, C6+Z, C8+Z*, Z

OF: organic fertilizer; PS: potting soil; C6–20: young compost (6 months) at 20 t ha⁻¹; C6–40: young compost (6 months) at 40 t ha⁻¹; C8–20: mature compost (8 months) at 20 t ha⁻¹; C8–40: mature compost (8 months) at 40 t ha⁻¹; C6+Z: a mixture of young compost and zeolite; C8+Z: a mixture of mature compost and zeolite; Z: zeolite; HL: hydrated lime; ML: magnesium lime; DE: diatomaceous earth; BM: bone meal; CH: crushed horn. An asterisk denotes a significant decrease in extractability in comparison to the given control soil for the NH₄NO₃ extraction. Amendments in bold denote a significant decrease in extractability in comparison to the given control soil as evaluated by the EDTA extraction.

5. Conclusion

Exposure to metal(loid)-contaminated kitchen garden soils can pose a great risk to human health. This study conducted an assessment of various amendments (at doses commonly used by gardeners), particularly composts and other organic and mineral amendments, used to manage urban kitchen garden soils with moderate geogenous and/or anthropogenic contamination. The impact of the amendments was evaluated via extractability of metal(loid)s by the use of two chemical extractants. An *ex-situ* experiment was performed, which provided evidence of the possibility of reducing the extractability of the metal(loid)s considered in the study. The results were, however, dependent on the physico-chemical soil parameters and the metal(loid) considered.

Overall, the efficiency of amendments at reducing the extractability of the selected metal(loid)s' was more marked for the ammonium nitrate extraction than for the EDTA extraction. It also varied greatly on the metal(loid) studied and the garden soil on which the amendment was applied to. Moreover, for the strong EDTA-extractant, several amendments (e.g., PS, OF, C8–40, C6+Z, Z, DE, BM) were efficient at reducing metal(loid)s' extractability with the first application in the cases of CKG-N and CKG-L. On the other hand, for PKG, the most acidic soil, reamendment (i.e., second application A2) was necessary to observe a significant efficiency for EDTA-extractable metal(loid)s. With the weakest NH_4NO_3 -extractant, the behavior of the metal(loid)s varied with the soil: (i) while the effectiveness of certain amendments (e.g., DE, BM, C6–40) to the PKG soil was demonstrated in the case of A1, the positive effects were significantly more marked in the case of A2; (ii) in the CKG-N soil, A1 demonstrated some positive effects at reducing metal(loid)s' extractability. The effects were significantly more marked in the case A2 (e.g., PS, OF, C6, C8); and (iii) in the CKG-L soil, the effectiveness of certain amendments (e.g., OF, C6, C8, C6+Z, C8+Z) was demonstrated in both doses. For all the soils, the results also depended on the type of product studied. Considering the three soils are inherently different based on their agronomic characteristics, their origin, and the nature of their metallic contamination, there is no single optimal solution. Therefore, tests must be carried out before any implementation activities on the kitchen gardens. The durability of the effects obtained is also to be noted, specifically in the case of organic amendments.

In this study, the influence of amendments on the availability of metal(loid)s in soils was monitored using chemical extractions. The results should be validated by examining the phytoavailability of these pollutants through biological models (i.e., vegetables) in *ex-situ* conditions, followed by *in situ* tests. Further investigation is also required to assess the effects of amendments on agronomic parameters, including the cycle of elements and the evolution of organic matter in the medium term.

Acknowledgments

The authors wish to thank Agriopale and more specifically Camille Dusannier for providing the compost samples. Special thanks to Arnaud Lefebvre and Julien Dassonneville from LGCgE for their contributions to the setting up of the *ex-situ* experiment. We also thank gardeners for allowing us to collect soil samples in their gardens and cities of Nantes and Lille for giving us access to the community gardens.

Additional Materials

The following additional materials are uploaded at the page of this paper.

1. Table S1: Soil amendment parameters.
2. Figure S1: Experimental design - Description of the different steps and conditions of the experiment.
3. Figure S2: Fractionation of Pb in the three soils studied (PKG, CKG-N and CKG-L); F1: exchangeable, water- and acid-soluble fraction, F2: reducible fraction, F3: oxidizable fraction, F4: residual fraction.

Author Contributions

Ashley Schnackenberg: conceptualization, methodology, data acquisition and analysis, statistical analysis, writing original draft; **Géraldine Bidar:** conceptualization, methodology, supervision, writing review editing; **Laure Beaudet:** conceptualization, methodology, writing review editing; **Valérie Bert:** conceptualization, methodology, writing review editing; **Patrice Cannavo:** methodology, writing review editing; **Sébastien Détriché:** sampling and mapping, writing review editing; **Francis Douay:** conceptualization, supervision, methodology, writing review editing; **René Guenon:** conceptualization, methodology, writing review editing; **Liliane Jean-Soro:** conceptualization, methodology, writing review editing; **Alice Kohli:** methodology, writing review editing; **Thierry Lebeau:** conceptualization, methodology, writing review editing; **Karen Perronet:** conceptualization, methodology, writing review editing; **Christophe Waterlot:** methodology, writing review editing; **Aurélié Pelfrêne:** conceptualization (lead), supervision, methodology, data acquisition and analysis, writing original draft.

Funding

The authors wish to thank ADEME (Agency for ecological transition), and more specifically Franck Marot, for the financial support of this research (contract n°1972C0005).

Competing Interests

The authors have declared that no competing interests exist.

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