Journal of Soils and Sediments (2021) 21:2273–2286
https://doi.org/10.1007/s11368-021-02933-y

SOILS, SEC 3 • REMEDIATION AND MANAGEMENT OF CONTAMINATED OR DEGRADED LANDS • RESEARCH ARTICLE

A promising amendment for the immobilization of heavy metal(loid)s in agricultural soil, northwest China

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Received: 22 November 2020 / Accepted: 14 March 2021 / Published online: 21 March 2021

Abstract

Purpose In situ remediation techniques are currently limited for agricultural soil contaminated by heavy metal(loid)s, particularly for alkaline soil. In this study, we applied the amendments (Chinese loess, attapulgites, and composted sludge) to a contaminated alkaline agricultural soil to compare their effectiveness in immobilizing heavy metal(loid)s.

Materials and methods Soil was collected from the uppermost 20 cm of a heavily contaminated cornfield in Baiyin, China. The amendments were added to the soil at the ratios of 1%, 5%, 10%, and 15%. Controls without amendment were also included. The effectiveness of amendments was evaluated using single extractions and Tessier sequential extraction procedure. The extractants in the single-step extraction were H_2O, calcium chloride ($CaCl_2$), diethylenetriaminepentaacetic acid (DTPA), and hydrochloric acid (HCl). Seed germination was performed with the soil after immobilization for 180 days.

Results Chinese loess effectively reduced the toxicity of heavy metal(loid)s and improve seed germination rates compared with attapulgites (ATPs) and composted sludge. Composted sludge effectively transformed heavy metal(loid)s, particularly Cu, from the carbonate fraction into the organic matter or the residual fraction. The concentrations of heavy metal(loid)s extracted with DTPA were positively correlated with the summation of the exchangeable and carbonate fractions ($P < 0.05$). A high seed germination rate could be obtained when the four amendments were added at the ratios between 5 and 10%.

Conclusions Chinese loess was effective and safe to immobilize heavy metal(loid)s in contaminated alkaline agricultural soil. DTPA performed the best in predicting the availabilities of As, Cd, Pb, Cu, and Zn in the soil.

Keywords Contaminated soil · Heavy metal(loid)s · Raw amendment · Toxicity · Arid region

1 Introduction

Heavy metal(loid)s contamination of agricultural soil is a major environmental problem of global concern because of their high toxicity, persistence, and bioaccumulation, which pose an indirect threat to human health via the food chain (Zhao et al. 2015; Sun et al. 2019; Dong et al. 2020; Mamat et al. 2020). The industrial wastewater used for irrigation, mining and smelting, solid wastes, abuse of pesticides and fertilizers, and deposition are primary sources of agricultural soil pollution (Zhao et al. 2015; Dong et al. 2019). According to the 2014 State of Environmental Bulletin, it can be concluded that 19.4% of arable land exceeds the soil environmental quality limits of China based on sampling points, and 21.7% has been contaminated by heavy metal(loid)s (MEPPRC (Ministry of Environmental Protection of the People's Republic of China) and MLRPRC (Ministry of Land and Resources of the People's Republic of China), 2014). Recently, it is reported that nearly 20,000,000 ha of agricultural soils has been polluted by heavy metals in China (Sun et al. 2019). Contamination of agricultural soil polluted by heavy metal(loid)s affects plant growth by interfering with nutrient absorption and cell metabolic activity, leading to a significant reduction in crop yields (Xu et al. 2019; Dong et al. 2020). He et al. (2020) estimated

Responsible Editor: Girish Choppala

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that 12,000,000 t of grains were contaminated with heavy metals per year in China, resulting in a reduction in the production of more than 10,000,000 t and economic losses of more than 20 billion dollars. Additionally, the World Health Organization (WHO 2016) estimated that 12,600,000 deaths each year were attributed to environmental pollution such as contaminated soil in 2012. Therefore, it is imperative to explore an environmentally friendly and cost-effective remediation technology to remediate the agricultural soil contaminated by heavy metal(loid)s to ensure food safety and human health.

The in situ immobilization of heavy metal(loid)s with amendments has been found to be an environmentally friendly remediation technique with low cost (Guo et al. 2006; He et al. 2020). It has been reported that this technique can reduce the mobility and bioavailability of heavy metal(loid)s in soil through adsorption, precipitation, and complexation (Hamid et al. 2019; He et al. 2020). The amendments frequently used to immobilize heavy metal(loid) include lime and limestone, clay minerals (attapulgite, bentonite, zeolite, and loess), metal oxide, phosphates, and organic substance (biochar, manure, composted sludge) (Appel and Ma 2002; Xu et al. 2017; Palansooriya et al. 2020). Loess and attapulgite clays (ATPs) have been found to reduce the mobility and bioavailability fractions of heavy metals in weak acid soil and sediment due to their high sorption capacity and alkaline characteristics (Álvarez-Ayuso and Garcıa-Sánchez 2003; Wang et al. 2015; Zang et al. 2017). Chinese loess covers an area of about 640,000 km^2 in northwest China, and ATPs are preliminary predicted to have reserves of about 1,000,000 t in Gansu Province, China. Application of composted sludge has also been reported to reduce the mobility of heavy metals by buffering soil pH, adsorbing metals, or complexing with organic active groups (Arif et al. 2018; Palansooriya et al. 2020). At high metal loading rate, biosolids (municipal byproduct) are recommended as an immobilizing agent for Cd, Pb, Cu, and Zn in soil (Shaheen et al. 2017). However, few attentions have been paid to remediation of heavy metal(loid)s contamination in alkaline soils compared to acidic soils (Liang et al. 2019), particularly in the arid oases of northwest China.

The bioavailability is defined as the potential of living organisms to take up chemicals from the external environment (Bolan et al. 2014). The bioavailable fractions of heavy metal(loid)s in soil are considered to be more toxic for water living beings, plants, and human (Liang et al. 2014). To examine the bioavailability of metal(loid)s in soil, chemical extracts and bioassay tests have been used in previous studies (Bolan et al. 2014; He et al. 2020). Both single and sequential extractions have been performed to examine the uptake of heavy metal(loid)s in a short-term or long-term experiment (Bakircioglu et al. 2011; Liu et al. 2017). Currently, the commonly used single extractants include salt solutions ($CaCl_2$, $MgCl_2$, $NaNO_3$, NH_4NO_3), buffer solutions (CH_3COONH_4), chelating agents (DTPA, EDTA), and acid solutions (HCl, HNO_3, CH_3COOH) (Pueyo et al. 2004; Feng et al. 2005; Wang et al. 2009; Liang et al. 2014; Zhang et al. 2016; Zang et al. 2017). The sequential extraction procedures mainly used in literature include Tessier method (Tessier et al. 1979; Xu et al. 2019), BCR method (He et al. 2020), Wenzel method (Wenzel et al. 2001), and Javed method (Javed et al. 2013). Additionally, the bioavailability of heavy metal(loid)s is predicted by thin films diffusion gradient and isotopic dilution techniques (Nolan et al. 2005). The indicators of bioassay included animals, plants, and microorganisms (Bolan et al. 2014). However, there is no standard evaluation method for remediation effect due to the different soil properties. The single extractions and Tessier extraction procedure were used to explore the bioavailability of heavy metal(loid)s in this study.

Baiyin is located in the central part of Gansu Province in northwest China and is known for its non-ferrous metal mining mineral resources, such as Cu, Pb, and Zn. Dongdagou stream is an important suburban drainage stream in Baiyin, which receives treated and untreated domestic wastewater and industrial sewage. The local farmers have been using wastewater from Dongdagou stream to irrigate or partially irrigate cropland over the past 50 years, resulting in severe contamination of the soil with heavy metals such as As, Cd, Pb, Cu, and Zn (Nan et al. 2002; Zhang et al. 2018). However, little attention is generally paid to study the remediation of metal-polluted agricultural soil in this area.

Therefore, the aim of this study was to screen for amendments suitable for alkaline polymetallic contaminated agricultural soil. Specifically, this study attempted to (1) evaluate the effectiveness of Chinese loess, ATPs, and composted sludge for the heavy metal(loid)s immobilization in polymetallic contaminated agricultural soil; (2) identify one or more extractants to predict the impact of applying three amendments in contaminated soil; and (3) discuss the possible mechanisms of heavy metal(loid)s immobilized by Chinese loess, ATPs, and composted sludge.

2 Materials and methods

2.1 Preparation of soil and raw amendments

The topsoil (0–20 cm) was collected from a cornfield (36° 28′ 23″ N, 104° 18′ 42″ E) in the Dongdagou watershed of Baiyin, China, following the Decision Unit-Multi Increment Sampling method (Mao et al. 2020). The soil belongs to the sierozem. Soil samples were homogenized, air-dried, and sieved through 2 mm to remove coarse gravel and visible plant debris.

Chinese loess belongs to Malan loess, collected in Yuzhong County, Gansu Province (Zang et al. 2017).

Attapulgites (ATPs) were obtained from Linze County (L-ATP) and Baiyin City (B-ATP), Gansu Province, respectively, provided by the College of Chemistry and Chemical Engineering, Lanzhou University. Composted sludge was collected at Baiyin Power Company's wastewater treatment plant in June 2019. Chinese loess, ATPs, and composted sludge were air dried, ground, and screened through a 10-mesh (2 mm) nylon sieve before they were used in the following experiment.

2.2 Chemical analysis

Soil pH, electrical conductively (EC), and dissolved organic carbon (DOC) were determined in 1:2.5 suspension at 25°C using a pH meter (Leici, PHS-3E, China), EC meter (YOKE INSTRUMENT, DDS-307A, China), and total organic carbon analyzer (Ma et al. 2021), respectively. The methods described by (Lu, 2000) were carried out to obtain the content of $CaCO_3$ and organic matter (OM) in soil. The total metal concentrations in the soil, amendments, and residual fraction were obtained by digesting with a fresh 3:1(v/v) HCl-HNO_3 mixture in a microwave digestion system (MARS6, CEM, US). The Cd, Pb, Cu, and Zn concentrations in filtrates were measured using an atomic absorption spectrophotometer (Thermo Fisher, ICE3000, US), and the As concentrations in filtrates were determined using an atomic fluorescence spectrophotometer (AFS-8220, Beijing Jitian Instrument Co., Ltd., China). The basic physicochemical properties and the metal concentrations of the soil and amendments were shown in Table S1. The mineral compositions of soil, Chinese loess, ATPs, and composted sludge were identified by the X-ray diffraction analyzer (XRD, D/max-2400, Rigaku). The surface morphologies of soil, Chinese loess, ATPs, and composted sludge were investigated by scanning electron microscope (SEM, MIRA3, TESCAN).

2.3 Soil immobilizing experiment

Chinese loess, composted sludge, and ATPs were added to soil at the rate of 1%, 5%, 10%, and 15% (dry weight) and mixed thoroughly with soil. Soil without any addition was included as controls (CK). The mixture was loaded in a plastic container (height 14.8 cm × diameter 10 cm). Subsequently, all plastic containers were placed in 25 °C for 180 days for soil incubation. The moisture was kept at 70% of the maximum field water capacity. All soils were carried out with six parallel treatments. At the end of incubation, the soil samples were removed from the plastic container and homogenized, air-dried, and sift through 2 mm for future analyses.

2.4 Immobilization and toxicity of heavy metal(loid)s

The bioavailability of heavy metal(loid)s in the control and treated soil was evaluated using single extraction including H_2O, 0.01 mol L^{-1} $CaCl_2$, 1 mol L^{-1} HCl, and 0.005 mol L^{-1} DTPA (pH 7.3) (Zang et al. 2017; Wang et al. 2018). Briefly, 1.0 g soil sample was added into 10 mL extract solution, and then the suspension was shaken in a thermostatic shaker at 250 r min^{-1} and 25 °C for 2 h. After that, the mixture was centrifuged, and the suspension was filtered with a 0.45 μm syringe filter for the analyses As, Cd, Pb, Cu, and Zn. The Tessier sequential extraction method was carried out to estimate the variations of fractionation of As, Cd, Pb, Cu, and Zn in the soil (Tessier et al. 1979). The five fractions were exchangeable (F1), carbonate (F2), organic (F3), Fe-Mn oxides (F4), and residual fractions (F5).

The seed germination test was often used to asses heavy metal(loid) toxicity due to its short experimental cycle and simple procedure (Xu et al. 2019). Briefly, 30 seeds (*Brassica chinensis*, *Chinese cabbage* and *spinach*) were selected and sowed in each plastic container after soil immobilization for 180 days. Soil moisture was adjusted to 70% of maximum field water capacity with tap water. The germination rate of the seeds was counted after 7 days of incubation at room temperature (16–30 °C).

2.5 Quality assurance and data analysis

For analytical precision, we repeated the chemical analysis in triplicate for each sample. The standard reference sample (GSS-8) and reagent blank were included in the whole procedure. The average recoveries and the relative standard deviations of the As, Cd, Pb, Cu, Zn, Fe, Mn, K, Ca, Na, and Mg were 90–110% and <10%, respectively. The fraction recoveries of As, Cd, Pb, Cu, and Zn were 92 ± 5%, 90 ± 6.9%, 95 ± 10%, 100 ± 10%, and 98 ± 10%, respectively.

The statistical analysis of all data was performed with Microsoft Excel 2010 and SPSS 22.0 software. The statistical differences between different amendments or between different ratios for each amendment were assessed using one-way analysis of variance (ANOVA) followed by Tukey's test at $p < 0.05$.

3 Results and discussion

3.1 Soil and raw amendments characteristics

The basic physicochemical parameters and metal concentrations of soil and four raw amendments used in this study were shown in Table S1. The pH of studied soil, Chinese loess, L-ATP, and B-ATP were 8.09, 8.25, 7.74, and 7.76, respectively, which were alkaline (Liang et al. 2019). The composted sludge was slightly acidic, with pH at 6.26. The EC of studied soil, Chinese loess, L-ATP, B-ATP, and composted sludge were 0.18 mS cm^{-1}, 2.08 mS cm^{-1}, 3.04 mS cm^{-1}, 3.94 mS cm^{-1}, and 5.69 mS cm^{-1}, respectively. The OM content followed the order composted sludge (58.88%) > soil (1.48%) > L-ATP (0.32%) > B-ATP (0.21%) > Chinese loess (0.14%), but the carbonate content ($CaCO_3$) exhibited a reverse order as follows: Chinese loess (14.93%) > B-ATP (13.04%) > soil (10.27%) > L-ATP (9.88%) > composted sludge (5.66%). The concentrations of heavy metal(loid)s in the soil were as follows: Zn (1,359.61 mg kg^{-1}) > Pb (523.00 mg kg^{-1}) > As (237.92 mg kg^{-1}) > Cu (171.01 mg kg^{-1}) > Cd (26.19 mg kg^{-1}), which were approximately 20 times, 28 times, 19 times, 7 times, and 218 times higher than the soil background values in Gansu Province (Zn: 68.5 mg kg^{-1}; Pb: 18.8 mg kg^{-1}; As: 12.6 mg kg^{-1}; Cu: 24.1 mg kg^{-1}; Cd: 0.12 mg kg^{-1}), respectively (China National Environmental Monitoring Center 1990), and were highly elevated compared to the risk screening values for soil contamination of agriculture land (Zn: 300 mg kg^{-1}; Pb: 170 mg kg^{-1}; As: 25 mg kg^{-1}; Cu: 100 mg kg^{-1}; Cd: 0.6 mg kg^{-1}). The concentrations of As and Cd in soil were approximately 2 times and 58 times higher, respectively, than the risk intervention values for soil contamination of agriculture land (As: 100 mg kg^{-1} and Cd: 4 mg kg^{-1}) (Ministry of Ecology and Environment of the People's Republic of China 2018). The results indicate that the studied soil was heavily contaminated by heavy metal(loid)s, particularly by Cd and As, and that a soil remediation approach was urgently needed. Previous studies have also identified wastewater irrigation from mining and smelting as a major cause of heavy metal(loid) contamination of farmland in this region (Zang et al. 2017; Dong et al. 2019).

The concentrations of As, Cd, Pb, Cu, and Zn in the Chinese loess, L-ATP, and B-ATP were below or slightly above the Gansu soil background value, whereas the contents of heavy metals in composted sludge were lower than the category A standards limits of the control standards of pollutants in sludge for agricultural use (Zn: 1200 mg kg^{-1}; Cu: 500 mg kg^{-1}; Pb: 300 mg kg^{-1}; As: 30 mg kg^{-1}; Cd: 3 mg kg^{-1}) (State Administration for Market Regulation 2018). Chinese loess, ATPs, and composted sludge can be used as amendments in agricultural soils. The concentrations of Fe, Mn, K, Ca, Na, and Mg were also abundant in Chinese loess, composted sludge, and ATPs (L-ATP and B-ATP) (Table S1). Calcium-, magnesium-, and ferromanganese-rich materials can adsorb and immobilize heavy metals due to their large specific surface area and higher ion exchange capacity (Liang et al. 2014; Zang et al. 2017; Hamid et al. 2019; He et al. 2020), whereas composted sludge has high organic matter, which improves the biochemical characteristics of contaminated soil (Palansooriya et al. 2020).

3.2 Toxicity of heavy metal(loid)s in the soil

3.2.1 The bioavailability of heavy metal(loid)s in the soil

The single extraction is an effective method to assess the toxicity of heavy metal(loid)s (Wang et al. 2009; Liang et al. 2014). In this study, we selected four extractants including H_2O, $CaCl_2$, DTPA, and HCl to predict heavy metal(loid) availability after immobilization (Fig. 1A~E). The four extractants differed in their abilities to extract heavy metal(-loid)s: HCl > DTPA > H_2O > $CaCl_2$. The desorption behavior of heavy metal(loid)s in soil was associated with soil properties, extractants, and metal types (Wang et al. 2009). The tremendous extraction capacity of HCl was attributed to the dissolution of the metal carbonate fraction and the organic complexed fraction (Bakircioglu et al. 2011), while DTPA was a buffered chelating agent that refrains dissolution of carbonate fraction and extracts only the complexed and adsorbed with organic matter and exchangeable fraction of metals (Xiao et al. 2015). Metals extracted by $CaCl_2$ were easily exchangeable fraction (Pueyo et al. 2004). H_2O showed higher capability than $CaCl_2$ for the extractions of As, Pb, and Cu in our study, which is in line with Bakircioglu et al. (2011) who found that the concentrations of Cu, Mn, and Ni extracted by H_2O were higher than those by $CaCl_2$ in a basic soil. The extraction concentrations of heavy metal(loid)s by the extractants were also different. The concentrations of heavy metal(loid)s extracted with DTPA and HCl extractants followed the order Zn > Pb > Cu > As > Cd, which could be attributed to the high concentrations of Zn and Pb in soil (Table S1). For H_2O and $CaCl_2$, the extractable concentrations of heavy metal(loid)s decreased as follows: As >Zn > Cu > Pb (Cd).

The amendments caused a variation of H_2O-, $CaCl_2$-, HCl-, and DTPA-extractable concentrations of heavy metal(loid)s compared to the control, and the magnitude of impacts depended on the types of metal, extractant, and amendment. Compared with the CK, the concentrations of As, Cd, Pb, Cu, and Zn extracted by the four extractants pronouncedly decreased with increasing ratio of Chinese loess, which is in well agreement with Zang et al. (2017). The effective immobilization of Cd, Pb, Cu, and Zn could be attributed to the abundant cations and the primary and secondary minerals in loess (Zang et al. 2017). Nazari et al. (2017) noted that AsO_4^{3-} and Ca^{2+} formed calcium arsenate precipitates in soil solution in alkaline environment, causing the reduction of As mobility. For ATPs, the concentrations of As, Pb, Cu, and Zn extracted by the four extractants decreased with the elevated ratio, which was likely due to the reduction of metal contents in the mixture (Zang et al. 2017) and the transformation from active fractions to inactive ones (Liang et al. 2019). Xu et al. (2019) also observed that ATP significantly decreased the concentrations of Cd, Pb, and Cr extracted by $CaCl_2$ with the increase in addition ratios and promoted the

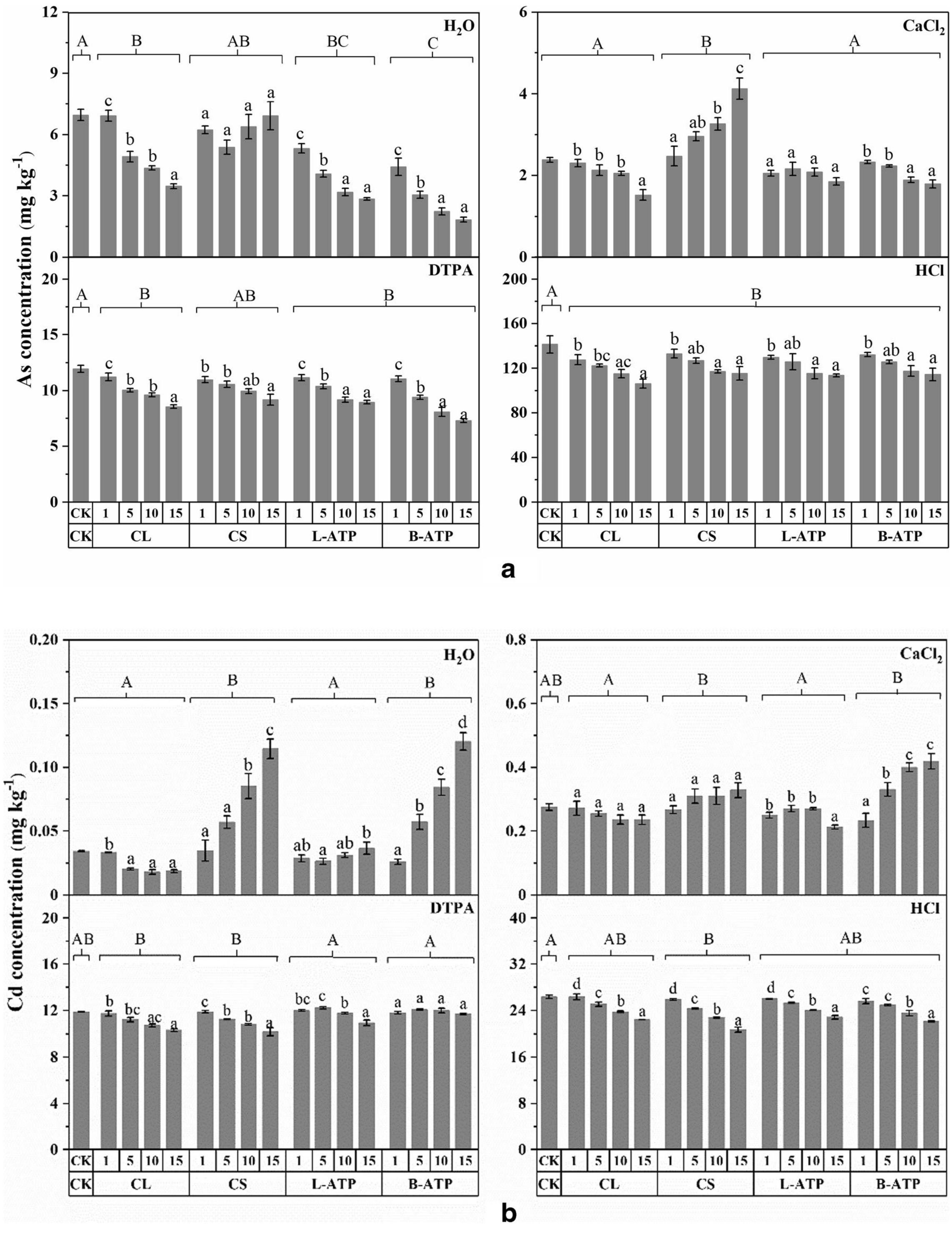
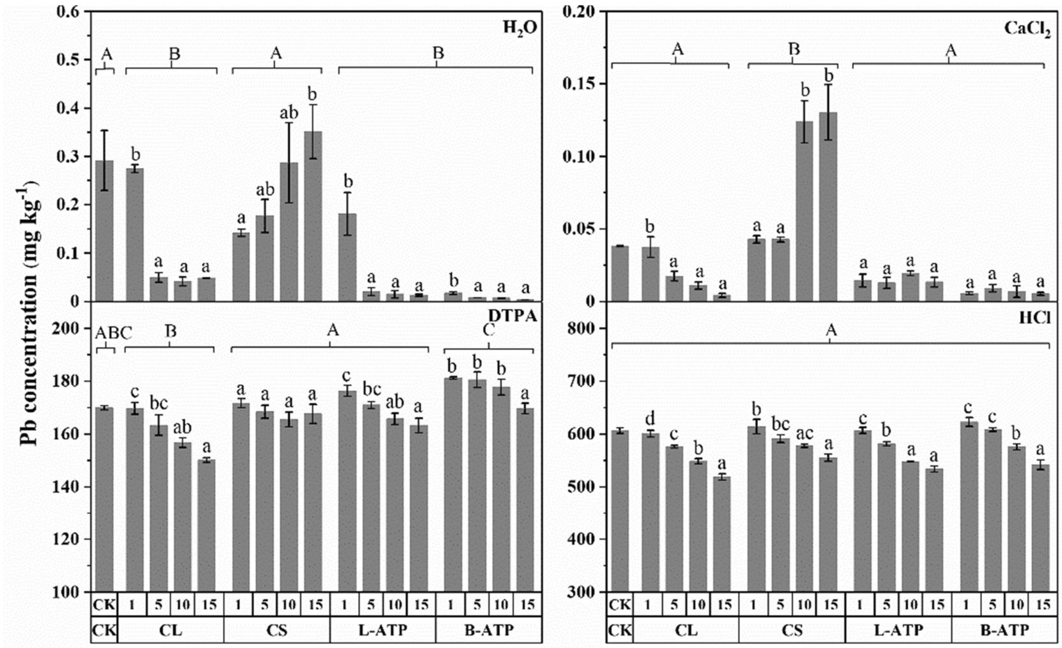


Fig. 1 Concentrations of As (**A**), Cd (**B**), Pb (**C**), Cu (**D**), and Zn (**E**) extracted by H_2O, $CaCl_2$, HCl, and DTPA from the amended soil. CK, untreated soil; CL, Chinese loess; CS, composted sludge; L-ATP, attapulgite obtained from Linze; B-ATP, attapulgite obtained from Baiyin. 1, 5, 10 to 15 denoted the application dose rates of Chinese loess, composted sludge and attapulgites (L-ATP and B-ATP). Error bars represented standard deviation (n = 6). Different lowercase letters on the columns indicated significant statistically differences among different dose of the same amendment at $p < 0.05$, while different capital letters on the columns indicated significant statistically differences among same amendment groups and control group at $p < 0.05$. The concentration of Zn extracted by H_2O was below the limit of detection (LOD: 0.1mg L^{-1}) of atomic absorption spectrophotometer (AAS)

transformation of metal species from exchangeable fraction to other relatively stable states in acidic soil. In contrast, the H_2O- and $CaCl_2$-Cd increased with the increase in the addition of ATPs, which may be due to large amounts of free ions in



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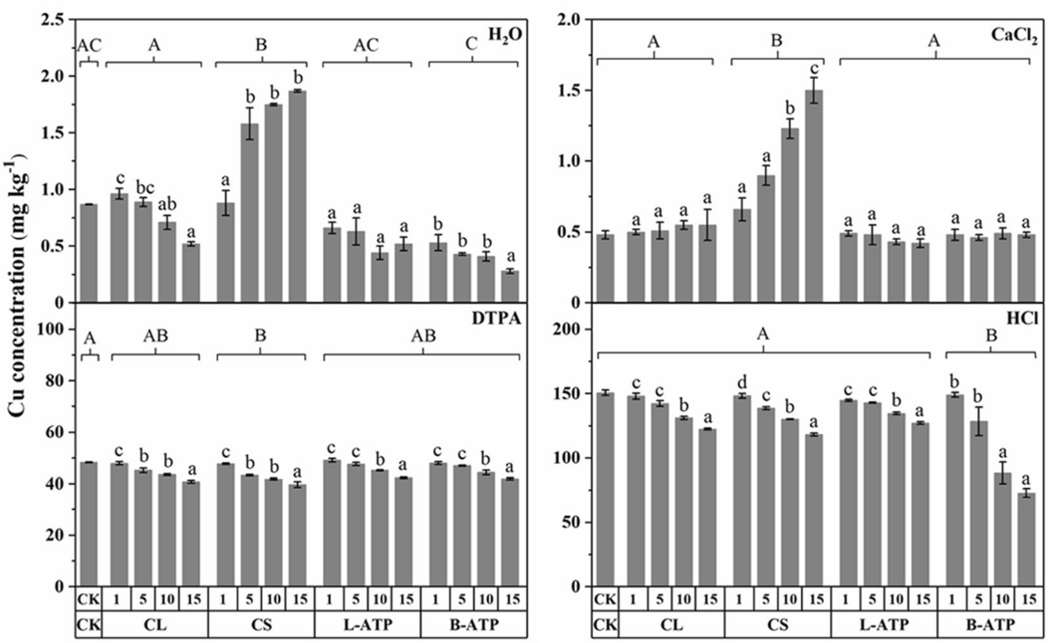

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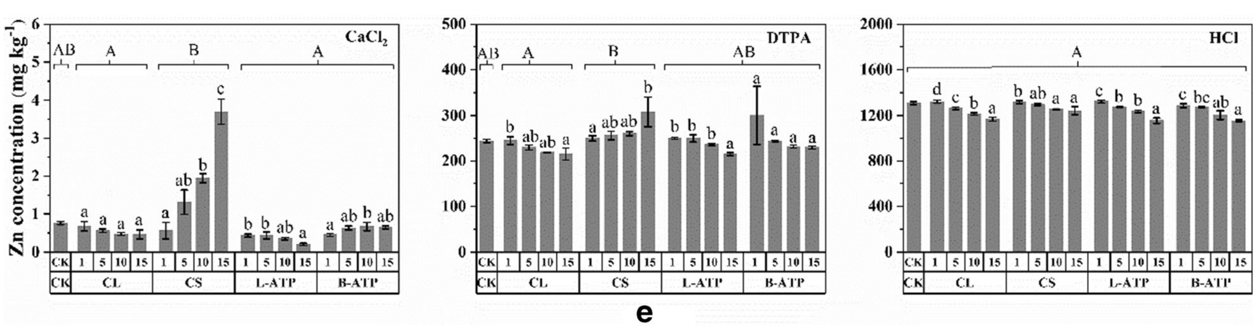


e

Fig. 1 (continued)

the ATPs (Table S1). Ma et al. (2021) found that the soil EC was negatively correlated with Cd bioavailability ($CaCl_2$-Cd), which could be attributed to the competitive adsorption of the heavy metals with free ions such as Ca, Na occurring at soil surface (Li 2018). The immobilization of metal cation by minerals was mainly through ion-exchange and adsorption to carbonate compounds hydroxides (He et al. 2020). However, Cd precipitation in the form of carbonate or hydroxide induced by natural sepiolite was impossible to occur during alkaline soil remediation (Liang et al. 2019). The concentrations of As, Cd, Pb, Cu, and Zn in soil extracted by H_2O and $CaCl_2$ increased with increasing composted sludge addition. This phenomenon can be explained by the fact that the addition of composted sludge caused partial dissolution of the carbonate fraction in soil due to its low pH and high EC, and the high heavy metals of the composted sludge itself (Table S1). For the HCl and DTPA extraction methods, the concentrations of As, Cd, Pb, Cu, and Zn in amended soil decreased with increasing ratio of composted sludge, but the DTPA-extractable Zn increased with increasing composted sludge ratio, which may be related with the increase in DOC in composted sludge amended soil (Table 2). The concentrations in the leachates of Zn and Ni were higher than those of Cu, Cd, and Pb in the soil amended with sewage sludge compost, which was explained by the dissolution of DOC at alkaline environment (Fang et al. 2016). However, the abundance of organic functional groups such as hydroxyl, carboxyl, carbonyl, and phenyl groups in compost can immobilize Pb, Cu, Cd, Ni, and Zn (Milojković et al. 2016). Sewage sludge compost had a high ability of acid-base buffering and contained large amounts of iron oxides, aluminum oxides, and organic matter (Fang et al. 2016), which are beneficial to heavy metal retention and immobilization. Overall, Chinese loess is safer than composted sludge and ATPs due to the fact that the addition of composted sludge increased the soluble and $CaCl_2$-extractable fractions of Zn, Cd, Pb, Cu, and As, and ATPs increased those of Cd.

The heavy metal(loid)s fractions from the sequential extraction method showed differences in mobility and bioavailability (Srithongkul et al. 2019; Xu et al. 2019). The extractable heavy metal(loid)s in F1and F2 from the Tessier method were considered available for plants (Liang et al. 2014; Zang et al. 2017); thus we used the sum of F1 and F2 to evaluate the bioavailability of heavy metal(loid)s. The results showed that the application of amendments was promising in decreasing the bioavailable As, Cd, Pb, Cu, and Zn (Fig. 2). Composted sludge showed a greater potential to reduce the bioavailable Cd, Pb, and Cu than Chinese loess and ATP when the ratio was >1%. For example, the summation of Cd, Pb, and Cu in F1 and F2 were reduced from 21.3 ± 0.6 to 16.6 ± 0.6 mg kg^{-1}, from 182.9 ± 1.1 to 113.9 ± 11.5 mg kg^{-1}, and from 23.7 ± 0.2 to 7.8 ± 0.5 mg kg^{-1}, respectively. B-ATP offered the best effectiveness for As, with 22.1% converted from 28.4 ± 0.32 mg kg^{-1} in the control to 22.1 ± 1.7 mg kg^{-1} in the soil treated with 15% B-ATP. A good performance of 10% Chinese loess was observed for the reduction of bioavailable Zn (27.5%). The specific heavy metal(loid) fractions in amended soil were discussed in the next section. To explore the relationship between the heavy metal(loid)s extracted by single extractants and the bioavailable metals, their Pearson's relationship is presented in Table 1. The concentrations of bioavailable As and Zn were positively correlated with the metal(loid)s extracted with H_2O, $CaCl_2$, DTPA and HCl. The concentrations of bioavailable Cd, Pb, and Cu were negatively correlated with the metals extracted with H_2O and $CaCl_2$ but were positively correlated with the metals extracted with DTPA and HCl. The heavy metal(loid)s extracted with DTPA correlated better with the summation of bioavailable heavy metal(loid)s in amended soil compared to HCl, $CaCl_2$, and H_2O. These showed that DTPA extraction procedure was applicable for simultaneous prediction of As, Cd, Pb, Cu, and Zn in amended soil.

3.2.2 The germination test

The seed germination of Brassica chinensis, Chinese cabbage, and spinach after amendment with Chinese loess, composted sludge, and ATPs are shown in Fig. 3. The seed germination rate decreased in the following order: brassica chinensis > Chinese cabbage > spinach, which indicates spinach was more sensitive to polymetallic contamination of soil compared to brassica chinensis and Chinese cabbage. After addition of amendments, the seed germination rate of brassica chinensis, Chinese cabbage, and spinach in the treatment group was higher than those in the controls. The addition of amendments could prevent heavy metal(loid)s uptake by plants and improve soil nutrient conditions, which in turn promoted plant growth (Arif et al. 2018; Xu et al. 2019). However, we found that seed germination rate decreased in the soils treated with the amendments at 15%, except the Chinese loess. ATPs applied at 15% brought about the soil compaction, which was not conducive to seed germination. Another reason could be the increased salinity after the application of composted sludge and ATPs (see Table 2, EC), which was found to limit plant growth (Masondo et al. 2018).

3.3 Fraction of heavy metal(loid)s in soil

The chemistry fractions of As, Cd, Pb, Zn, and Cu in the soil by the Tessier sequential extraction are presented in Fig. 4. The predominant fraction of As in the control was F5 (47%), followed by F3 (36%) and F2 (11%), while F1 and F4 accounted for only 4% and 2% of total As. The primary fraction of Cd in the control soil was F2 (47%), followed by F1 (27%), the F3 (24%), F4 (1%), and F5 (1%). The dominant fraction of Pb in the soil without any addition was F3 (49%), followed by F2 (26%), F5 (16%),

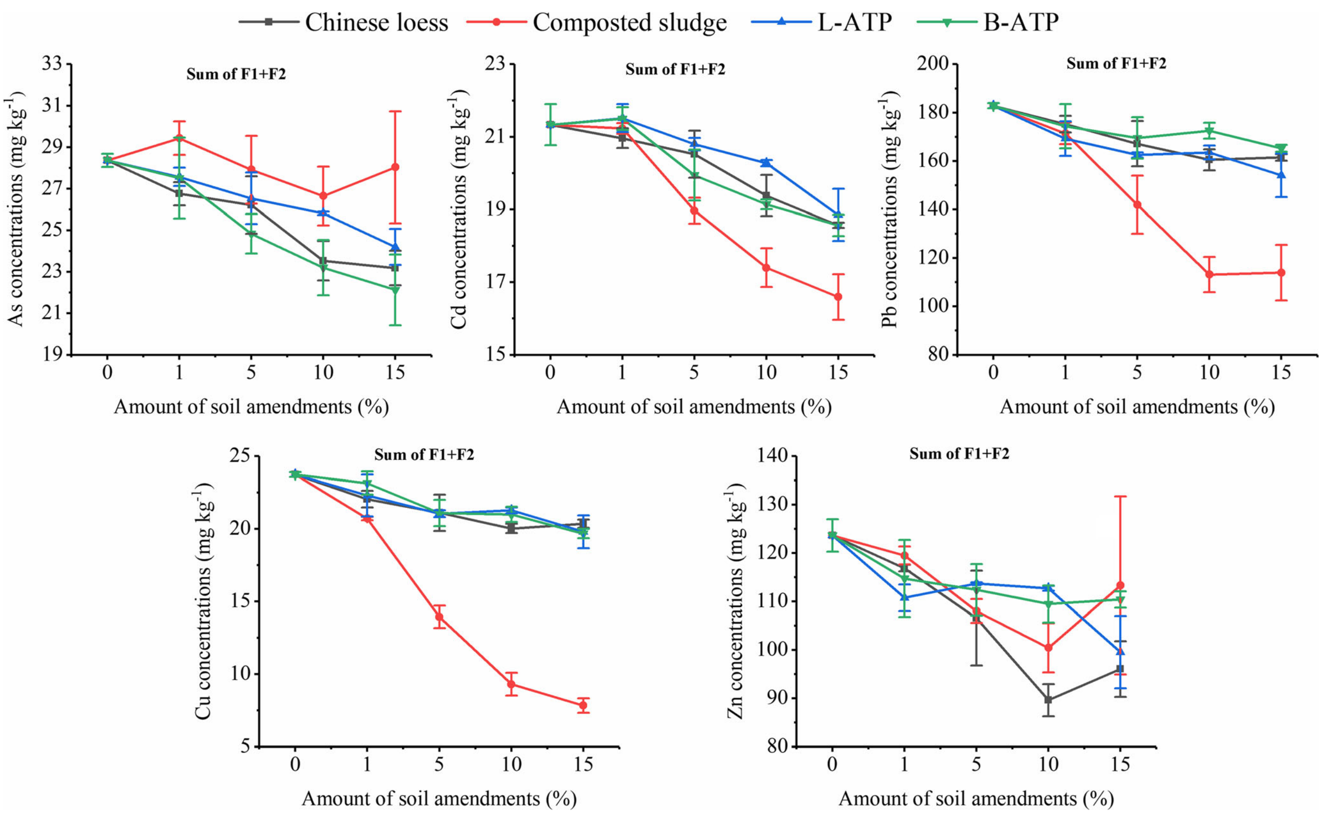


Fig. 2 Summation of F1 (exchangeable fractions) and F2 (carbonate fractions) of heavy metal(loid)s fractions in soil samples after treated with Chinese loess, Composted sludge, L-ATP and B-ATP at different application rates for 180 d of incubation. Error bars represent standard deviation (n = 6)

F4 (7%), and F1 (2%). The Cu in unamended soil was mainly F5 (34%), followed by F3 (31%), F4 (23%), F2 (11%), and F1 (0.4%). The Zn fraction of unamended contaminated soil was predominantly F5 (62%), followed by F3 (21%), F2 (9%), F4(7%), and F2 (2%). It is notable that the bioavailability of Cd was higher than As, Pb, Cu, and Zn in this study. However, the bioavailability of As, Pb, Cu, and Zn should not be neglected due to the high total concentration in this area. Compared with CK, the application of amendments led to a decrease of 2.4–9.3% in As in F3 in soil, while the proportion of F5 increased by 1.6–9.6% (Fig 3a). These also explained the reduced concentration of As with single extractions. The addition of minerals such as bentonite, zeolite, and dolomite has been found to lead to a decrease in reducible As, but an increase in oxidizable As in agriculture soil (He et al. 2020). Figure 3b showed that the Chinese loess and composted sludge promoted the transformation of Cd in soil from the F2 to F3. It was noteworthy that composted sludge and ATPs led to the transformation of Cd in soil from F2 to F1. This also explains that the Cd concentrations increase in the H_2O and $CaCl_2$ extracts. However, the application of palygorskite shifted Cd species from F1 to F2 and F5 in an acidic soil (Liang et al. 2014). The application of composted sludge led to the transformation of Pb in the soil from the F2 to F5 (Fig. 3c). Composted sludge could dramatically increase organic fraction of Cu due to the decrease in F2 and F3 of Cu. The F5 of Cu was slightly elevated by Chinese loess and ATPs in the soil (Fig. 3d). It proved that Cu had the greater affinity for OM than other metals (Shaheen et al. 2017). Zang et al. (2017) also found that Chinese loess effectively elevated residual Cu with the reduce of carbonate Cu in sediment. The amendments dramatically decreased F2 and F3 of Zn but elevated the F5 in the L15, S5 and LA10 samples (Fig. 3e). This study provides an analysis of the fractions of heavy metal(loid)s in amended soil, and the unique

Table 1 Pearson correlation coefficients of heavy metal(loid)s extracted by different extractants and available metal(loid)s in the amended soil

Available metals[a]	H_2O	$CaCl_2$	DTPA	HCl
As	0.788**	0.603*	0.834**	0.734**
Cd	−0.723**	−0.422	0. 742**	0.927**
Pb	−0.464	−0.787**	0.484**	0.331
Cu	−0.840**	−0.954**	0.703**	0.208
Zn	/	0.664**	0. 683**	0.234

** Correlation is significant at the 0.01 level (2-tailed)

* Correlation is significant at the 0.05 level (2-tailed)

[a] Sum of the heavy metal(loid) concentrations in the exchangeable and carbonate fractions from the Tessier method

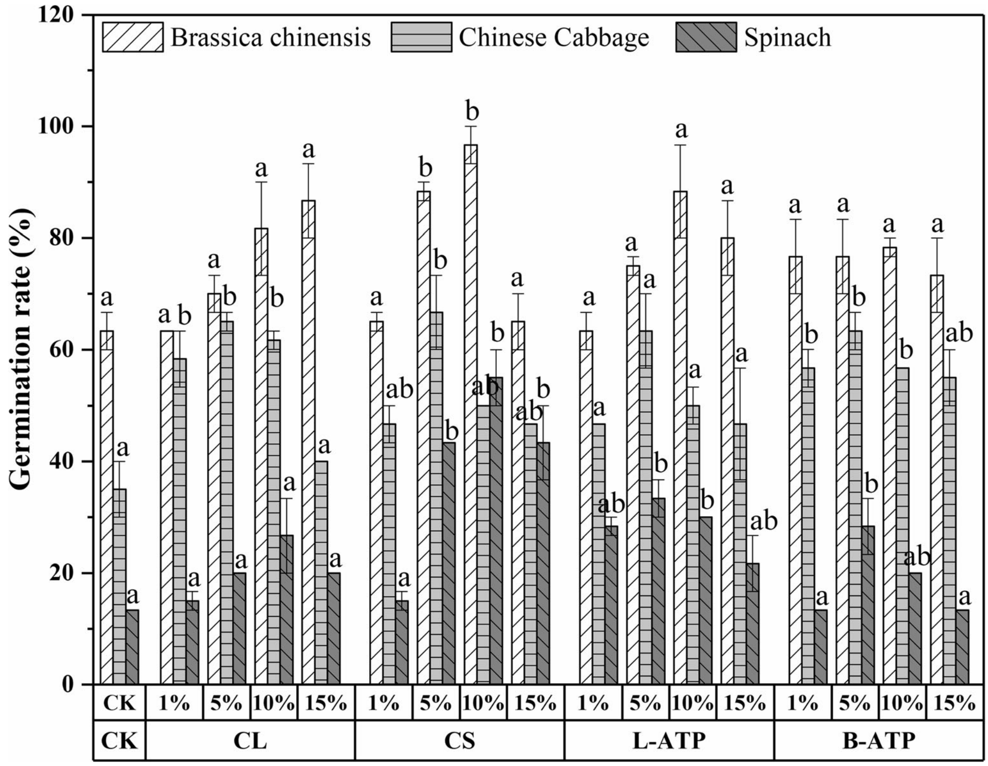


Fig. 3 The germination rate of *Brassica chinensis*, *Chinese cabbage,* and *Spinach* at amended soil. CK, untreated soil; CL, Chinese loess; CS, composted sludge; L-ATP, attapulgite obtained from Linze; B-ATP, attapulgite obtained from Baiyin. 1 to 15% denoted the application dose of Chinese loess, composted sludge, and attapulgites (L-ATP and B-ATP). Error bars represented standard deviation (n = 6). Different letters on the columns indicated significant statistically differences among different treatments of the same vegetable at $p < 0.05$

Table 2 Variations in the physicochemical properties in soils amended with Chinese loess (CL), composted sludge (CS), and attapulgites (ATPs) (L-ATP and B-ATP)

Sample	pH	EC (mS cm^{-1})	$CaCO_3$ (%)	OM (%)	DOC (mg kg)
CK	7.75 ± 0.04 a	0.25 ± 0.01a	10.2 ± 0.4ab	1.7 ± 0.2a	28.3 ± 2.2a
CL					
L1	7.70 ± 0.07 a	0.28 ± 0.01a	10.3 ± 0.1a	1.6 ± 0.1a	23.1 ± 1.3ab
L5	7.81 ± 0.02 a	0.42 ± 0.04b	10.7 ± 0.4a	1.7 ± 0.2a	20.0 ± 0.1b
L10	7.74 ± 0.02 a	0.57 ± 0.06c	11.1 ± 0.2a	1.5 ± 0.0a	19.9 ± 0.1b
L15	7.75 ± 0.02 a	0.76 ± 0.01d	10.5 ± 0.6a	1.7 ± 0.1a	21.6 ±1.9ab
CS					
S1	7.77 ± 0.05 a	0.38 ± 0.02a	10.2 ± 0.3a	2.1 ± 0.2a	37.3 ± 0.1a
S5	7.40 ± 0.03 b	0.78 ± 0.05b	10.0 ± 0.3a	2.9 ± 0.1b	62.7 ± 2.9b
S10	7.07 ± 0.02 c	1.13 ± 0.07c	9.8 ± 0.6a	4.5 ± 0.2c	115.4 ± 1.0c
S15	6.77 ± 0.04 d	1.69 ± 0.16d	9.3 ± 0.0c	5.6 ± 0.2d	127.4 ± 0.4d
L-ATP					
LA1	7.61 ± 0.03 a	0.33 ± 0.03a	9.9 ± 0.1a	1.7 ± 0.2a	25.0 ± 2.1ab
LA5	7.59 ± 0.01 b	0.67 ± 0.02b	9.8 ± 0.0a	1.6 ± 0.0a	19.8 ± 1.1ab
LA10	7.41 ± 0.07 b	1.01 ± 0.03c	10.3 ± 0.4a	1.4 ± 0.0a	16.5 ± 0.5b
LA15	7.40 ± 0.01 b	1.22 ± 0.04d	10.4 ± 0.1a	1.6 ± 0.2a	17.2 ± 2.0b
B-ATP					
BA1	7.49 ± 0.03c	0.60 ± 0.08a	10.0 ± 0.1a	1.5 ± 0.2a	17.5 ± 1.9b
BA5	7.37 ± 0.03bc	1.23 ± 0.13b	10.5 ± 0.1ab	1.5 ± 0.0a	18.6 ± 0.7b
BA10	7.30 ± 0.03b	1.65 ± 0.14c	10.8 ± 0.1b	1.5 ± 0.0a	17.1 ± 0.3b
BA15	7.29 ± 0.09b	1.97 ± 0.17c	10.5 ± 0.3ab	1.5 ± 0.1a	16.6 ± 1.0b

The results in this table represent "mean ± standard deviation (n = 6)". Different lowercase letters indicated significant statistically differences among different dose of the same amendment and control group at $p < 0.05$

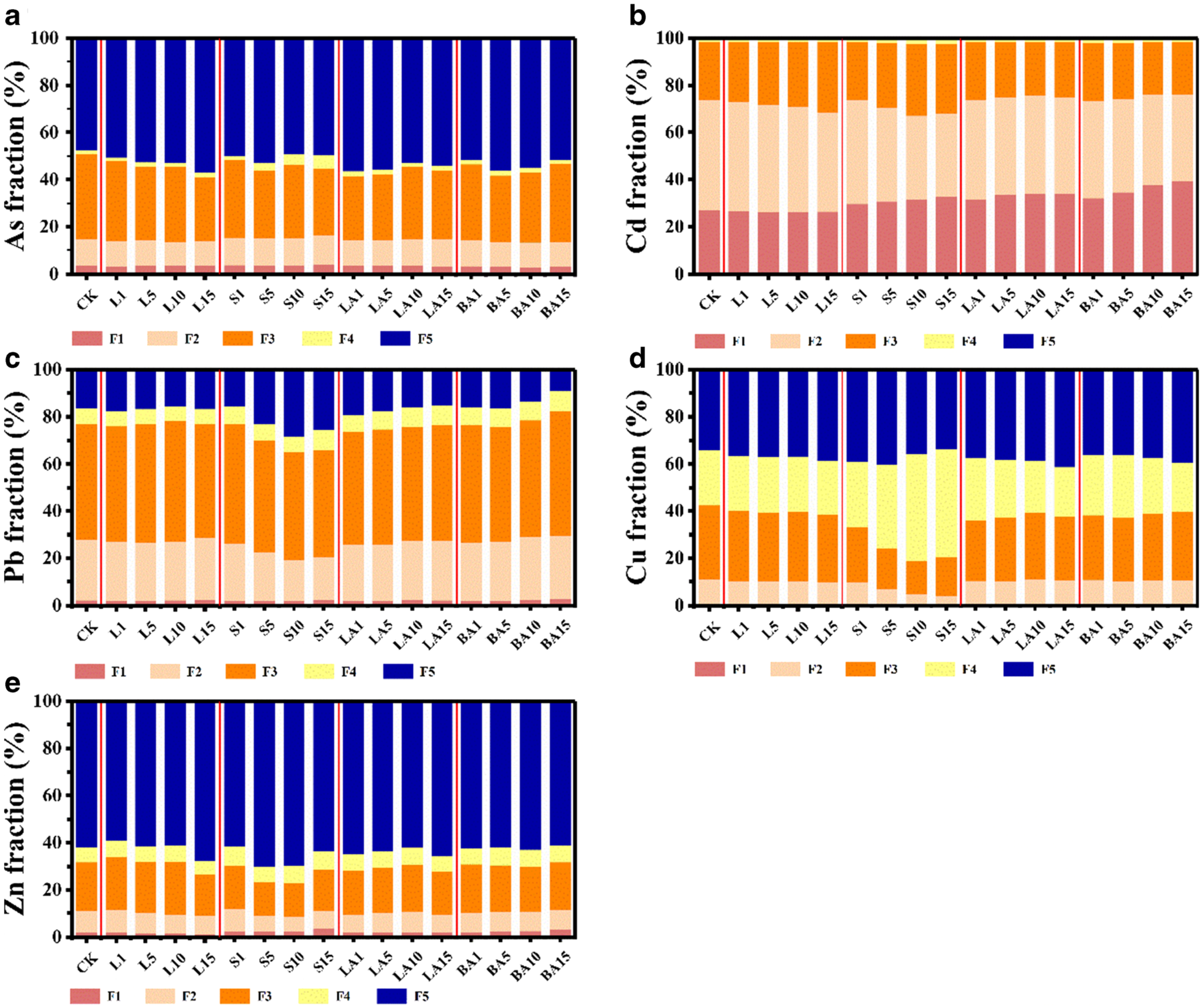


Fig. 4 Chemistry fractions of As (**a**), Cd (**b**), Pb (**c**), Zn (**d**), and Cu (**e**) in amended soil by the Tessier sequential extraction. CK: untreated soil. L1 to L15 denote 1%, 5%, 10%, and 15% loess application dose, respectively. S1 to S15 denote 1%, 5%, 10%, and 15% composted sludge application dose, respectively. LA1 to LA15 denote 1%, 5%, 10%, and 15% L-ATP (attapulgite obtained from Linze) application dose, respectively. BA1 to BA15 denote 1%, 5%, 10%, and 15% B-ATP (attapulgite obtained from Baiyin) application dose, respectively.

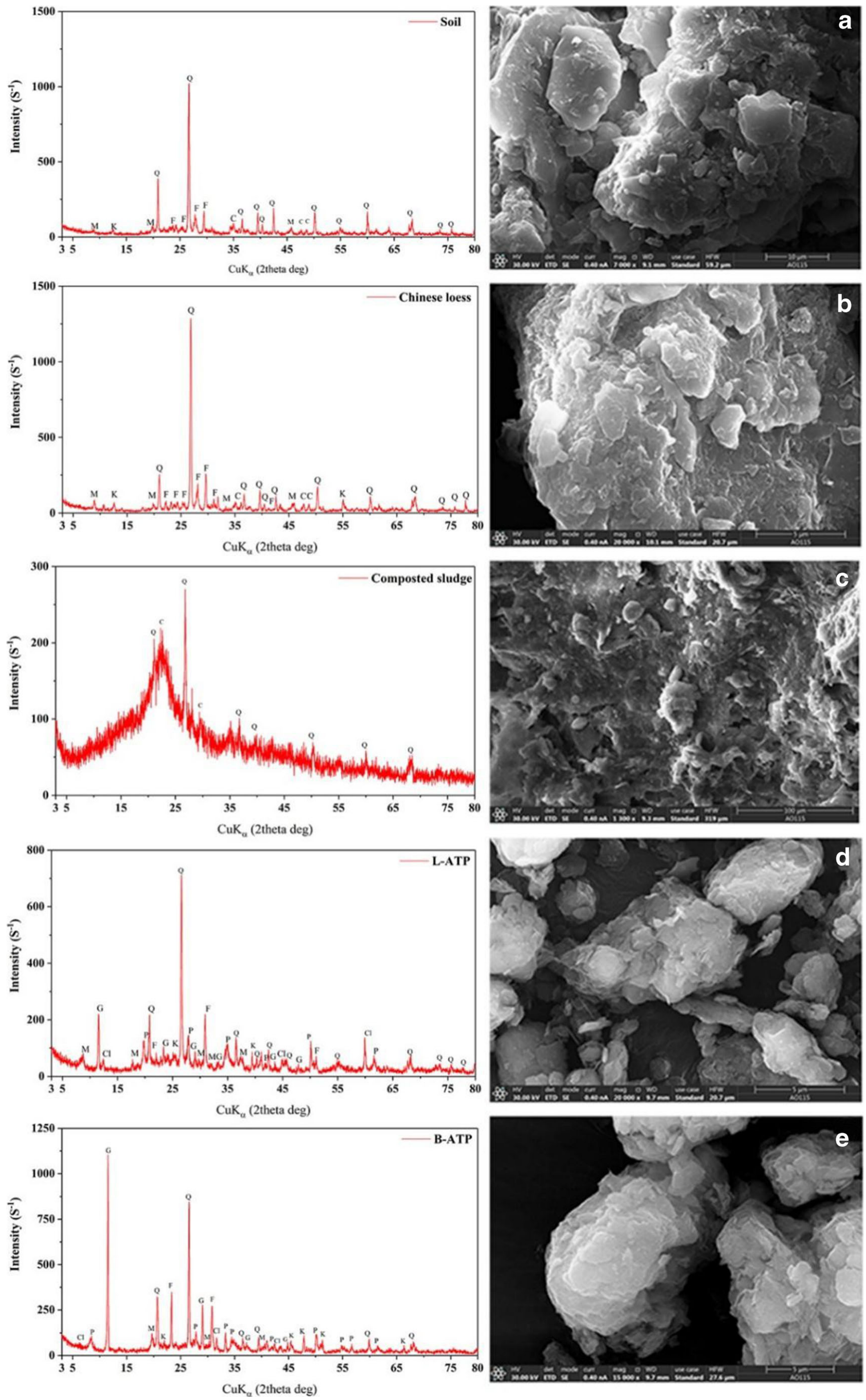
relationships and interactions among polymetals will be explored in the future study.

3.4 Possible mechanisms of heavy metal(loid)s immobilization in soil

3.4.1 Microscope analysis of soil, Chinese loess, composted sludge, and ATPs

The mineral crystalline phase of soil, Chinese loess, composted sludge, and ATPs was analyzed by X-ray diffraction (XRD) (Fig. 5). The mineral composition of soil and Chinese loess was mainly calcite, feldspar, kaolinite, muscovite, and quartz, while the ATPs also contained clinochlore, gypsum, and palygorskite. The XRD pattern of the composted sludge displayed a wide absorption peak from 20 to 30° belonged to amorphous carbon, indicating the amorphous structure of the material (Liu et al. 2020). The characteristic peaks of calcite and quartz also were also observed in material and well crystallized. The remediation mechanisms of clay minerals such as palygorskite, diatomite, sepiolite, vermiculite, and bentonite mainly include liming, precipitation, and sorption effects (Xu et al. 2017). Soil Cd was immobilized to the surface of clay

Fig. 5 XRD and SEM images of soil (**a**), Chinese loess (**b**), composted sludge (**c**), L-ATP (**d**), and B-ATP (**e**). Minerals are denoted with peaks labeled: C, calcite; Cl, clinochlore; F, feldspar; G, gypsum; K, kaolinite; M, muscovite; P, palygorskite and Q, quartz


Soil
Intensity (S⁻¹)
CuKα (2theta deg)
a
Chinese loess
b
Composted sludge
c
L-ATP
d
B-ATP
e

minerals through interlayer ion exchange and adsorption, whereas soil As can form precipitates with Cd and Mg in clay minerals (He et al. 2020). The active silanol groups (-OH) of attapulgite and composted sludge clay were conducive to enhance the adsorption of heavy metals forming heavy metal ligands (Fang et al. 2016; Xu et al. 2019). The calcite in minerals can immobilize Cd and Pb by forming $CdCO_3$ and $PbCO_3$ (Yin and Zhu 2016). The Al, Fe, and Mn oxides and oxyhydroxides in minerals and composted sludge are found to immobilize As and Pb by strong specific adsorption in soil (Fang et al. 2016; Palansooriya et al. 2020). However, the behavior mechanisms of heavy metal(loid) in amended soil merits further detailed study.

The morphologies of soil, Chinese loess, composted sludge, and ATPs were screened by SEM images (Fig. 5). The results reveal that the morphologies of soil, Chinese loess, composted sludge, and ATPs had irregular structures. Chinese loess had a rough surface and a porous structure with a specific surface area of 24.1m^2 g^{-1} (Wang et al. 2015). SEM imaging of composted sludge illustrated that the composted sludge surface was uneven and had many porous structures. Hazrati et al. (2020) found that the surface area of sewage sludge was 3.04 m^2 g^{-1}. Many rod-like single crystals were attached to the surface of ATPs with some pores forming on its surface, which was conducive to increasing the specific surface area of ATPs. Xu et al. (2019) reported that the specific surface area and pore size of attapulgite clay were 143m^2 g^{-1} and 71.0 nm, respectively. The huge specific surface area and pore size of materials can provide more sites for adsorption, which is conducive to the capture of heavy metals.

3.5 Variation in physicochemical properties of amended soil

The application of amendments to the soil affected the physicochemical properties of the soil. Soil physicochemical properties are important factors affecting the availability of heavy metal(loid)s in soil. Table 2 lists the variation of pH, EC, $CaCO_3$, and OM in unamended and amended soil after 180-day incubation. Specifically, compared to CK, soil pH significantly decreased ($p < 0.05$) after application of composted sludge and ATPs when the ratio of amendments was > 5 % and soil EC significantly elevated ($p < 0.05$) with increasing amendment ratio. Soil $CaCO_3$ and OM did not significantly change after application of Chinese loess and ATPs compared to the unamended soil, but soil OM significantly increased ($p < 0.05$) with increasing composted sludge ratio. The concentrations of As, Cd, Pb, Cu, and Zn in H_2O and $CaCl_2$ extracts presented positive correlation with soil OM, but negative correlation with soil pH (except for H_2O-As) (Table S2). Soil $CaCO_3$ gradually decreased with increasing composted sludge ratio. The degradation of organic matter in the soil can lead to a decrease in soil pH, resulting in the release of heavy metal(loid)s bounded by carbonate. Therefore, we believe that the increased concentrations of heavy metal(loid)s extracted by H_2O and $CaCl_2$ in the composted sludge treatment group were mainly from the release of heavy metal(loid)s and sludge itself. This also supported the increasing Cd concentration extracted by H_2O and $CaCl_2$ in ATPs treatments group. Interestingly, we found that soil pH exhibited significant positive correlation with most heavy metal(loid) concentrations in DTPA and HCl extracts, while soil EC, $CaCO_3$, and OM exhibited significant negative correlation with most heavy metal(loid) contents in DTPA and HCl extracts (Table S2). However, the soil pH showed a negative correlation with the bioavailability of heavy metals in soil amended with minerals in literature (Liang et al. 2014; Zang et al. 2019; He et al. 2020). It was impossible to decrease the bioavailability of heavy metals by regulating soil pH in alkaline soils (Liang et al. 2019). Therefore, the soil EC, $CaCO_3$, and OM play major roles in immobilizing availability metals in alkaline soil.

4 Conclusions

This study demonstrated the immobilization effects of As, Cd, Pb, Cu, and Zn by four raw amendments in agricultural soil. The remediation effectiveness of Chinese loess, composted sludge, and ATPs was relevant to the amendment properties, extractants, and metal species. Chinese loess showed higher removal rate than composted sludge and ATPs. DTPA can better predict the availability of As, Cd, Pb, Cu, and Zn in the soil than H_2O, $CaCl_2$, and HCl. Also, the seed germination test showed that the application of Chinese loess and composted sludge can pronouncedly elevate seed germination rates compared to ATPs. Additionally, the ratio of Chinese loess, ATPs, and composted sludge use as a passivating agent should be maintained between 5 and 10% for optimal seed germination. Moreover, Soil EC, $CaCO_3$, and OM played major roles in immobilizing availability metals in alkaline soil. That is noteworthy when the salinity of the soil is elevated to the detriment of heavy metal(loid) immobilization in soil. Further study will be conducted to explore the optimal combined application rate of Chinese loess with combination composted sludge to simultaneously immobilize heavy metal(loid)s in the soil.

This manuscript has not been published and is not under consideration for publication elsewhere.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11368-021-02933-y.

Availability of data and material The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

Author contribution Shengli Wang and Zhongren Nan conceived and designed the research; Yi Wu, Xiang Ning, Meng Yang, and Mengbo Liu were involved in the pretreatment of the samples; Yi Wu collected the data and analyzed the data; Yi Wu and Fei Zang wrote and reviewed the manuscript. All authors have read the manuscript, agree the work is ready for submission to a journal, and accept responsibility for the manuscript's contents.

Funding This work was supported by the National Key Research and Development Program of China (2018YFC1802905).

Declarations

Ethics approval and consent to participate Not applicable

Consent for publication Not applicable

Competing interests The authors declare no competing interests.

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