RESEARCH ARTICLE

Efficiency and comprehensive risk assessment of soil Pb and Cd by washing technique with three biodegradable eluents

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Abstract

Soil washing with environmentally friendly eluents is a rapid remediation technique for farmland polluted by heavy metals. In this study, polyepoxysuccinic acid (PESA), ethylenediamine tetra (methylene phosphonic acid) sodium (EDTMPS), and phosphonyl carboxylic acid copolymer (POCA) were applied to remedy paddy and arid soils polluted by Pb and Cd. At the same time, ethylenediaminetetraacetic acid (EDTA) was used as a control eluent. PESA showed comparable removal of soil Pb and Cd (over 80.0%) with EDTA, and EDTMPS and POCA removed two heavy metals by 35.2–50.3%. For labile fractions, PESA significantly removed Pb by 93.5–96.7% and Cd by 84.9–90.3% in two soils. EDTMPS and POCA removed Pb by 75.5–85.8% in two soils, while they only removed Cd by 11.7–42.2% in paddy soil, and 76.3–81.7% in arid soil. The risks of total heavy metal concentrations were reduced from the high risk to low risk in paddy soil, and to considerable risk in arid soil, while only dropped to considerable or even had no change by EDTMPS and POCA leaching. The risks of the two soils reduced from high to low or considerable level after PESA washing based on labile fraction change, and to considerable or high level after EDTMPS and POCA leaching, respectively. Therefore, PESA is an ecological benefit eluent for remediating the farmland polluted by heavy metals, and the risk assessment based on labile fraction more easily identifies the dynamic change of heavy metal during the washing process.

Keywords Soil washing · Heavy metal · Polyepoxysuccinic acid · Risk assessment · Modified RI

Introduction

Soil pollution by heavy metal is a severe environmental issue throughout the world due to various anthropogenic activities (Ghosh and Singh 2005; Zhang et al. 2010; Karak et al. 2017; Contessi et al. 2021). As non-essential elements for biological functions, lead (Pb) and cadmium (Cd) are two heavy metals of particular concern with respect to environmental quality and human health for their high toxicity and carcinogenic properties (Ashraf et al. 2019). Specifically, they have become major soil pollutants in Asian, African, and American countries owing to mining, battery manufacturing, smelting, and sewage sludge in recent decades (Ferg and Rust 2007; Karak et al. 2017; Frank et al. 2019). Therefore, there is an urgent need to seek some practices to remediate the polluted soils (Feng et al. 2020).

At present, soil leaching or washing technique is considered to be a fast practice of removing heavy metal from polluted soil compared with the conventional remediation ones such as replacement (Kobayashi et al. 2008), electrodialysis (Rosestolato et al. 2015), stabilization (Contessi et al. 2021), and phytoremediation (Zhang et al. 2010). This practice was usually conducted to remedy the polluted soil with higher metal concentrations in industrial and mining sites in the past years; in contrast, it is difficult to be applied in farmland with lower metal concentrations in the field due to some disadvantages such as destruction to soil fertility (Cao et al. 2017a, b; Feng et al. 2020) and expensive cost. However, the remediation technique for farmland at large scale mainly depends on selecting the effective, green, and inexpensive eluents (Gong

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et al. 2018) when application of portable wastewater treatment system drastically reduced the time and cost of the remediating project. Among various eluents, ethylenediaminetetraacetic acid (EDTA) has been frequently applied in soil washing for its efficiency, availability, and low cost (Lestan et al. 2008). However, it is poorly biodegradable and might have negative side-effects on soil microorganisms and plants (Grcman et al. 2001; Jez and Lestan 2016). Up to now, only limited green washing materials such as plant extractants and biodegradable agents have been reported in the literatures at large scale (Cao et al. 2017a, b; Piccolo et al. 2019; Thinh et al. 2021). Accordingly, it is imperative to seek new environmentally friendly and low-cost eluents (Wang et al. 2018; Piccolo et al. 2019).

Polycarboxylic acids (e.g., polyepoxysuccinic acid, PESA), phosphonic acid (e.g., ethylenediamine tetra (methylene phosphonic acid) sodium, EDTMPS), and copolymer (e.g., phosphonyl carboxylic acid copolymer, POCA) are three water treatment agents widely used in chelating metal ions (Chaussemier et al. 2015; Han et al. 2019). Most of them contain carboxylic acid and hydroxyl groups that pose high capacity to complex metal ions (Marco et al. 2004). Moreover, they are photodegradable and have no biotoxicity (Lesueur et al. 2005). Nevertheless, it is unclear whether the three reagents own high removal efficiencies for the soils polluted by Pb and Cd from battery manufacturing.

It is important to evaluate the potential ecological risk of an eluent except for its high efficiency during the soil heavy metal removal (Gusiatin and Kulikowska 2014; Gong et al. 2018). The risk assessment would be an aid to investigate whether some eluent is feasible for soil washing of farmland (Zhu et al. 2012). To date, many risk assessment indices, such as contamination factor, enrichment factor, geo-accumulation index, single pollution index, Nemerow pollution index, and potential ecological risk index (RI), have been used to estimate the change in soil ecological risk before and after heavy metal washing (Kowalska et al. 2018). However, most of them are calculated according to total metal concentrations, and rarely addressed the diversity of toxic effect among different fractions of the heavy metals (Liang et al. 2017; Ji et al. 2019). It is well-known that labile fractions of heavy metals played more important roles than total metal concentrations in reflecting ecological risk of heavy metal to soil ecosystem (Boesten 1993; Adriano 2001; Zhong et al. 2021). Especially, the labile fraction might be increased from stable component transformation during the washing process (Beiyuan et al. 2016; Wang et al. 2018). Although several indices have been employed to assess the risk of labile or mobile heavy metal, e.g., risk assessment code, it was described as the ratio of labile fractions to their total concentration; thus, it is only suitable for risk assessment of polluted soil in no disturbing condition (Liang et al. 2017), and unsuitable for change in fraction and total concentrations of heavy metal.

Among these indices, RI can be a guideline for risk assessment, which has been widely applied in the assessment of soil heavy metal pollution (Ntakirutimana et al. 2013; Maanan et al. 2015). However, simple RI is difficult to reveal the status changes of different fraction concentrations and toxicity of some heavy metal, and even the disparity between its total concentration and background value. Therefore, it is necessary to modify the calculation equation of RI to classify risk levels of labile fraction of heavy metal more accurately (Zhu et al. 2012; Gusiatin and Kulikowska 2014).

We hypothesized that the three reagents would be promising washing eluents with high efficiencies and low ecological risk to remove soil Pb and Cd. The objectives of this study were (1) to investigate the effects of solution concentrations, and pH of these eluents and duration on removal efficiencies of heavy metals with EDTA acting as control eluent, and (2) to estimate the potential ecological risk of total and available Pb and Cd in soils before and after washing based on the original and modified RI.

Materials and methods

Study area and soil samples

The heavy metal polluted soils were derived from paddy field and arid land nearby a lead-acid battery factory in Chengdu, China. The factory is responsible for lead battery manufacturing including oxide and grid processing, plate processing, and battery assembly according to the investigation. The Pb and Cd pollution was mainly derived from industrial exhaust produced during casting process, and discharging the waste mixtures and toxic wastewater directly into soil. Soil samples were collected from the 0–20-cm layer, air-dried, sieved through a 2-mm nylon screen, and homogenized for further processing.

Washing eluents

Ethylenediaminetetraacetic acid (EDTA) was purchased from XiLONG SCIENTIFIC Co., Ltd. (Chengdu, China). PESA, EDTMPS, and POCA were purchased from Changzhou Runyang Chemicals Co., Ltd. (Jiangsu, China). POCA is synthesized from phosphorous acid, acrylic acid, and 2-acrylamide-2-methylpropanesulfonic acid. Their purities are 40, 40, and 35%, and their structures are shown in Fig. S1.

Chemical analysis

Soil pH was measured in water using a soil-to-water ratio of 1:5 with a pH electrode (pHSJ–3F, Shanghai INESA Scientific Instrument Co., Ltd., China) following a 2-h extraction (Vogel 1994). Soil texture was determined by the pipette

method (Gee and Bauder 1986). Soil organic matter was analyzed by the Walkley-Black titration method (Walkley and Black 1934). Electrical conductivity was conducted. Total concentrations of heavy metals including Pb, Cd, As, Cr, Cu, and Zn were determined after digestion in a 1:2:2 (v:v:v) mixture of HNO_3−HCl−$HClO_4$ (Ministry of Land and Resources of the People's Republic of China 2016). During digestion, the reference material (GBW07405) was used as quality control. The recovery rates of metals from standard samples were approximately 95–105%. Pb, Cd, Cr, Cu, and Zn concentrations were measured by the inductively coupled plasma optical emission spectrometry (ICP-OES; PerkinElmer Optima 8000, USA), and As was analyzed by the atomic fluorescence spectrometry (AFS) (AFS-3000, Haiguang instrument, China).

Textures of two soils are loamy clay with clay, silt, and sand contents of 29, 36, and 35% for paddy soil, and 23, 34, and 43% for arid soil. Soil pH was 5.98 for paddy soil, and 7.12 for arid soil. Soil Pb, Cd, As, Cr, Cu, and Zn concentrations were 1630.88, 2.51, 14.96, 40.23, 22.90, and 48.54 mg kg^{-1} in paddy soil, and 2218.28, 5.81, 20.75, 65.00, 24.67, and 50.25 mg kg^{-1} in arid soil (Table 1). Among these heavy metals, Pb and Cd concentrations exceeded the threshold limits in agricultural soils according to *Soil environment quality-Risk control standard for soil contamination of agricultural land* (GB 15618-2018) (Table S1).

Washing experiments

Washing experiments with different eluents including PESA, EDTMPS, POCA, and control eluent EDTA were conducted as a function of solution concentrations, pH, and duration. Soil sample (2.5 g) was mixed with 25 mL eluent solution in a 50-mL centrifuge tube. Tubes were shaken at room temperature (20 °C), then suspension was separated by centrifuge at 4000 r/min for 5min, and filtered. Each experiment was run in triplicate. In a concentration-dependent washing process, the four eluents were prepared at concentrations of 0.001, 0.005, 0.02, 0.05, 0.08, 0.1, and 0.2 mol L^{-1}, at the solution pH value of 4.0, and duration of 120 min. In a pH-dependent washing process, the solution pH was set at 4.0, 4.5, 5.0, 6.0, 7.0, 7.5, and 8.0 by adding diluted HNO_3 and/or NaOH, the corresponding eluent concentration at 0.08 mol L^{-1}, and duration at 120 min. In a duration-dependent washing process, the duration varied from 10, 30, 60, 120, 180, to 240 min under the eluent concentration at 0.08 mol L^{-1} and solution pH at 4.0. The total Pb and Cd removal efficiencies (%) were calculated using the below equation:

$$R_T = \frac{C_{solution} \times V}{m \times C_T} \times 100\% \qquad (1)$$

where R_T is the removal efficiency of total heavy metal, $C_{solution}$ is the concentration of metal in washing solution (mg L^{-1}), V is the volume of washing solution (L), m is the soil weight (kg), and C_T is the total concentration of heavy metals in polluted soil (mg kg^{-1}).

Labile fractions of soil heavy metals

Based on single factor washing experiments, soils were collected after washed with the eluents of 0.08 mol L^{-1}, solution pH of 4.0, and duration of 120 min to extract heavy metal concentrations of labile fractions. The labile fractions of heavy metals before and after washing were determined by 0.1 mol L^{-1} HCl, which has been proven fine correlation with biological absorption data (Sutherland and Tack 2008; Leleyter et al. 2012; Xu et al. 2016), and calcium chloride (0.01 mol L^{-1})-triethanolamine (0.1 mol L^{-1})-diethylenetriamine pentaacetic acid (0.005 mol L^{-1}) (DTPA) (GB/T 23739-2009, China), standard available heavy metal extraction methods, respectively. Briefly, air-dried soil (2 g) in a 50-mL tube, add 20 mL of extraction reagent, agitate 2 h at 180 rpm at room temperature (20 °C), then centrifuge for 10 min at 3000 r/min, the suspension was filtered through 0.45-μm filters, and analyzed by ICP-OES (PerkinElmer Optima 8000, USA). Each experiment was run in triplicate. The calculation followed the equation:

$$ML = C_L \times V/m \qquad (2)$$

where ML is the extraction concentration of labile fractions (mg kg^{-1}), C_L is the heavy metal concentration in the extract (mg L^{-1}), V is the volume of the corresponding extraction eluent (L), and m is the weight of soil sample for extraction (kg).

FTIR analysis

The four eluents before and after the soil washing process at concentration of 0.08 mol L^{-1}, pH of 4.0 and duration of 120 min were dried at 60 °C in an oven, ground with KBr (spectroscopic grade), and were identified by FTIR spectrometry (Spectrum Two, PerkinElmer Inc., USA). The possible functional groups involved the washing process were confirmed in the range of 4000–400 cm^{-1} wavenumber.

Ecological risk assessment

In this study, we evaluated ecological risk for both total and labile fraction concentrations of soil Pb and Cd.

Table 1 Soil chemical characteristics and heavy metal concentrations in soils before and after washing

Soil type	Treatment	Sand/Silt/Clay (%)	pH	SOM (g kg⁻¹)	CEC (cmol kg⁻¹)	Ca (g kg⁻¹)	Fe (g kg⁻¹)	Pb_T (mg kg⁻¹)	Pb_HCl (mg kg⁻¹)	Pb_DTPA (mg kg⁻¹)	Cd_T (mg kg⁻¹)	Cd_HCl (mg kg⁻¹)	Cd_DTPA (mg kg⁻¹)
Paddy soil	Unpolluted	/	6.47	/	14.79	/	/	66.65±0.53	18.87±0.31	14.81±0.93	0.28±0.002	0.07±0.001	0.06±0.001
	Polluted	29/36/35	5.98	29.19±0.20	17.03	3.48±0.23	29.75±0.10	1630.88±2.46	920.31±0.42	882.79±6.78	2.51±0.04	0.64±0.02	0.59±0.004
	EDTA		6.37	/	13.85	/	/	195.65±1.66	48.91±0.55	46.25±1.43	0.45±0.006	0.07±0.006	0.10±0.0005
	PESA		6.24	/	17.68	/	/	169.24±1.34	30.58±0.58	28.68±1.78	0.38±0.006	0.08±0.003	0.09±0.0004
	EDTMPS		6.25	/	12.77	/	/	391.25±5.65	137.59±6.87	134.38±2.12	1.01±0.05	0.36±0.005	0.36±0.003
	POCA		6.32	/	9.35	/	/	880.46±7.52	216.11±1.44	214.99±7.53	2.05±0.002	0.54±0.004	0.53±0.009
Arid soil	Unpolluted	/	7.22	/	20.02	/	/	74.50±0.58	13.44±0.12	11.73±0.08	0.23±0.001	0.07±0.001	0.06±0.001
	Polluted	23/34/43	7.12	26.67±0.12	22.53	13.64±1.15	28.87±0.74	2218.28±1.53	1389.35±37.2	1272.54±10.37	5.81±0.02	2.41±0.02	2.14±0.002
	EDTA		7.25	/	16.89	/	/	177.44±0.47	23.74±0.69	24.86±0.56	1.55±0.003	0.20±0.0008	0.28±0.002
	PESA		7.15	/	16.59	/	/	643.46±1.75	81.43±0.14	81.59±0.43	1.25±0.01	0.24±0.007	0.27±0.007
	EDTMPS		7.27	/	10.47	/	/	687.77±5.43	204.71±0.89	179.94±0.58	3.68±0.005	0.53±0.007	0.41±0.005
	POCA		7.34	/	10.04	/	/	1109.46±17.25	312.10±7.35	240.14±16.57	3.77±0.005	0.45±0.008	0.42±0.003

EDTA, ethylenediaminetetraacetic acid; *PESA*, polyepoxysuccinic acid; *EDTMPS*, ethylenediamine tetra (methylene phosphonic acid); *POCA*, phosphonyl carboxylic acid copolymer; *SOM*, soil organic matter; *CEC*, cation exchange capacity; *Pb_T*, *Cd_T*, total concentration of Pb and Cd; *Pb_HCl*, *Pb_DTPA*, *Cd_HCl*, *Cd_DTPA*, labile fractions of Pb and Cd extracted by HCl and DTPA, respectively.

Ecological risk assessment for total heavy metal

The potential ecological risk index proposed by Håkanson (1980) was commonly used as a diagnostic tool for heavy metal pollution evaluation. We applied it to evaluate the risk degree of total concentration (RI_Total), and it is calculated as the following equation:

$$RI\_Total = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \frac{C_i}{C_{GB}^i} \quad (3)$$

where C_i is the total concentration of metal i; C_{GB}^i is the geochemical background value of metal i, and in this study, it being referred to as the threshold limits in agricultural soils according to *Soil environment quality-Risk control standard for soil contamination of agricultural land* (GB 15618-2018) (Table S1); T_r^i is the toxic response factor for a given metal i (Pb=5, Cd=30) (Håkanson 1980); n is the numbers of heavy metals; and E_r^i is potential ecological risk of an individual metal i based on its total concentration.

Based on Håkanson's report, the risk classification was based on the calculation of seven metals including Pb, Cd, Hg, As, Cr, Zn, and Ni, and one organic pollutant (polychlorinated biphenyls) (Table S2). However, in our study, only Pb and Cd were involved; thus, the RI classification thresholds were modified. In Hankanson's work, the "Low risk" of RI threshold is 150, corresponding to the value (133) of the sum of Er when the ratio of C_i to C_{GB} is 1.0. Therefore, we proposed a replacement threshold of 40 as "Low risk" for only considering Pb and Cd with toxic response factor of 5 and 30. The original and modified RI thresholds are given in Table S3.

Ecological risk assessment for labile fractions of heavy metals

a.   The removal rate of labile fraction

The removal rates of heavy metals for labile fractions (R_L) directly reflect the risk reduction based on the concentrations in before and after washing soils. It can be defined as follows:

$$R_L = \frac{ML_{before} - ML_{after}}{ML_{before}} \times 100\% \quad (4)$$

where R_L is the removal rate of labile fraction and ML_{before} and ML_{after} are the labile fraction concentration in before and after washed soil, respectively.

b.   Potential ecological risk index for labile fractions (RI_Labile)

The labile fractions of heavy metals were enshrined in C_i value to highlight the importance of available heavy metals.

We proposed the threshold limits of labile fractions (C_{TL}) of Pb and Cd for the first time, and based on the concentrations of labile fractions in unpolluted soil and the threshold limits of Pb and Cd in GB 15618-2018, it is calculated as:

$$C_{TL} = \frac{\text{Lablie\_HCl} + \text{Labile\_DTPA}}{2 \times \text{UnC}_T} \times C_{GB}^i \quad (5)$$

where C_{TL} is the threshold limits of labile fractions, Labile_HCl and Labile_DTPA are heavy metal concentrations in unpolluted soil extracted by HCl and DTPA, respectively, UnC_T is total concentration in unpolluted soil, and C_{GB}^i is the geochemical background value of heavy metal i referred to as the threshold limits in GB 15618-2018. According this equation, the threshold limits of labile fractions of Pb and Cd were obtained that 25 and 0.09 mg kg^{-1} for paddy soil, and 20 and 0.08 mg kg^{-1} for arid soil.

Secondly, we use the extracted concentrations of labile fractions (C_L^i) and obtained threshold limits (C_{TL}^i) as enshrined in the Ci and C_{GB}^i values. Thus, the risk degree of labile fraction (RI_Labile) is calculated using the following equation:

$$RI\_Labile = \sum_{i=1}^{n} L_r^i = \sum_{i=1}^{n} T_r^i \frac{C_L^i}{C_{TL}^i} \quad (6)$$

where C_L^i is the average concentration of labile fractions extracted by HCl and DTPA, C_{TL}^i is the threshold limit of labile fraction metal i obtained by Eq. (5), T_r^i is the toxic response factor for a given metal i (Pb=5, Cd=30) (Håkanson 1980), L_r^i is the potential ecological risk of individual metal i based on its labile fraction, and n is the numbers of heavy metals.

Statistical analysis

The analysis of variance (ANOVA, one way) was applied on the mean±standard deviation of triplicates independent experiments, and mean differences among different treatments were compared by Fisher's least significant difference test at $P < 0.05$ in SPSS version 19.0 (SPSS Inc, Chicago, USA). Figures were created in Origin version 9.1 (OriginLab Corporation, Northampton, USA).

Results

FTIR analysis of washing eluents before and after the soil washing process

The functional groups involved in the washing process for four eluents were identified by FTIR and are shown in Fig. 1. In the EDTA washing process, new adsorption peaks arose at around 3400–3200 cm^{-1} (–OH), 2300 cm^{-1} (C=O), 1750
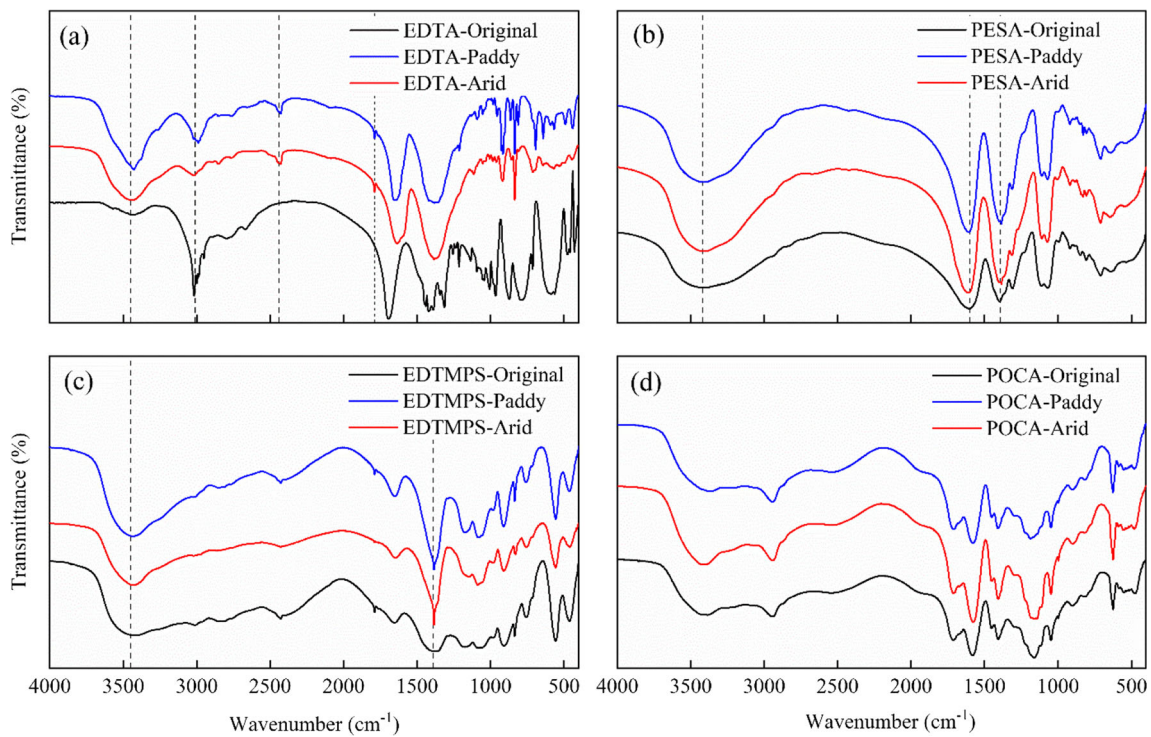
Fig. 1 FTIR spectra of EDTA, PESA, EDTMPS, and POCA before and after washing process for paddy soil and arid soil in spectral wavenumber range from 4000 to 400 cm⁻¹. EDTA, ethylenediaminetetraacetic acid (**a**); PESA, polyepoxysuccinic acid (**b**); EDTMPS, ethylenediamine tetra (methylene phosphonic acid) sodium (**c**); POCA, phosphonyl carboxylic acid copolymer (**d**)

Fig. 2 Soil Pb and Cd removal under different concentrations of EDTA, PESA, EDTMPS, and POCA under solution pH of 4.0, and time of 120 min. EDTA, ethylenediaminetetraacetic acid; PESA, polyepoxysuccinic acid; EDTMPS, ethylenediamine tetra (methylene phosphonic acid) sodium; POCA, phosphonyl carboxylic acid copolymer (**a**, Pb in paddy soil; **b**, Pb in arid soil; **c**, Cd in paddy soil; **d**, Cd in arid soil)
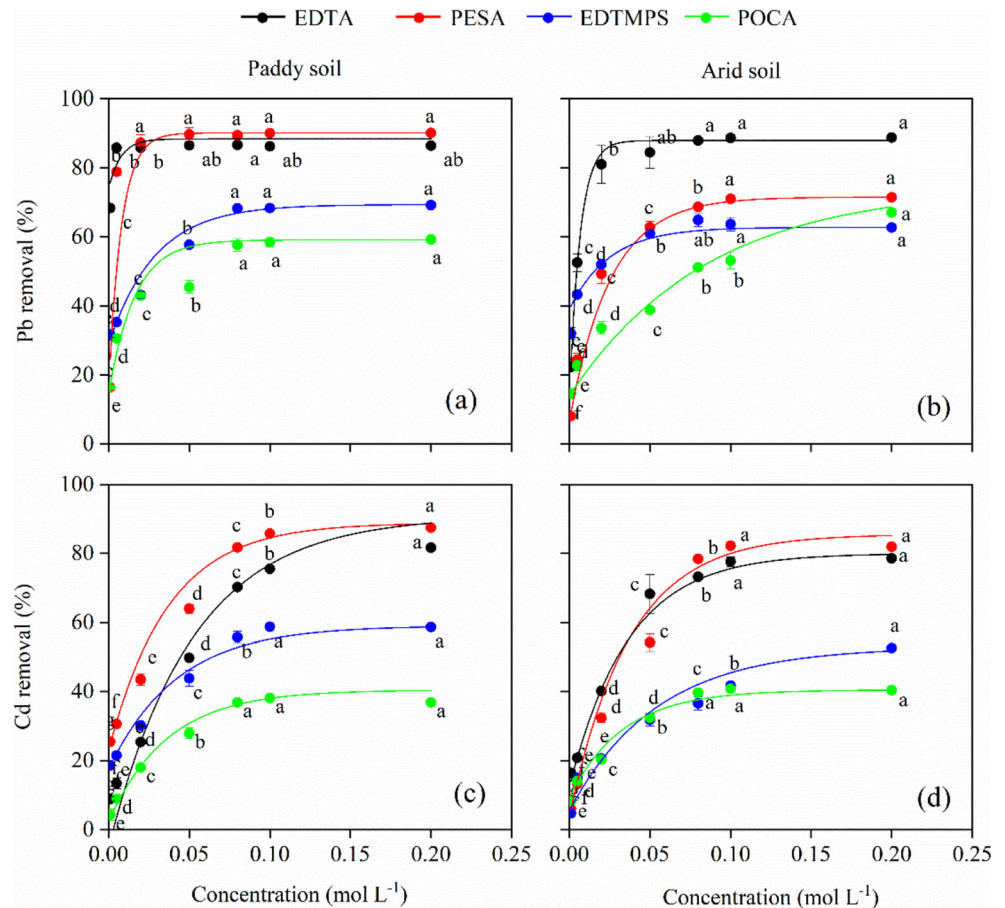
cm^{-1} (C=O) (Fig. 1a); the peaks of untreated EDTA at 1700 cm^{-1} (C=O) (Fig. 1a), 960 cm^{-1} (C–O), 870 cm^{-1} (C–C/C–H), and 720 cm^{-1} (C–C/C–H) slightly shifted to right location; and peaks around 3000 cm^{-1} (C–H), 1400–1300 cm^{-1} (–COOH), and 600–400 cm^{-1} (C–C/C-H) significantly decreased even disappeared (Figs. 1a and S2a). Compared with EDTA, no obvious peak variation was found in PESA, EDTMPS, and POCA washing processes. Briefly, the broad peaks at 3600–3400 cm^{-1} (–OH), 1400 cm^{-1} (C–H), and 1200–1000 cm^{-1} (C–O–C) became narrow (Figs. 1b and S2b), and peaks at 1700–1600 cm^{-1} (C=O) increased remarkably in PESA washing (Fig. 1b). For EDTMPS, only peaks at 1400–1300 cm^{-1} (–COOH) obviously increased (Fig. 1c), while there was nearly no variation observed on POCA (Fig. 1d).

Effect of a single factor on removal efficiencies of heavy metals

Concentration

Removal efficiencies of soil Pb and Cd sharply increased with higher eluent concentrations firstly and remained stable when the concentrations exceeded 0.08–0.1 mol L^{-1} (Fig. 2, Table S4). The Pb removal under washing by PESA was similar to EDTA, with the highest removal efficiency around 88.4% at its concentration > 0.05 mol L^{-1}, and higher than the other two eluents in paddy soil (Fig. 2a), while the efficiencies of the three eluents were lower than that of EDTA in arid soil (Fig. 2b). The highest Cd removal efficiencies of both EDTA and PESA washing in two soils were around 80.0%, and those of EDTMPS and POCA only owned 35.2–50.3% (Fig. 2c and d).

pH

Soil Pb and Cd removal efficiencies showed similar patterns when washed by EDTA, PESA, and EDTMPS in two soils, and decreased with increasing solution pH from 4.0 to 8.0 (Fig. 3, Table S5). The Pb removal of PESA in arid soil indicated a remarkable decreasing trend with higher pH, and its highest removal efficiency was 73.3% at pH 4.0, then decreased to lower than 20.0% at pH 7.0–8.0 (Fig. 3d). However, the Pb and Cd removal of POCA showed different trends compared with the other three eluents. The eluent obtained higher Pb removal at solution pH around 6.0, and lower Pb removal under more acid or alkaline condition in two soils (Fig. 3a and b). While for soil Cd removal, it owned higher values at solution pH of 7.5–8.0 (Fig. 3c and d).

Fig. 3 Soil Pb and Cd removal across different levels of solution pH values at the conditions of 0.08 mol L^{-1} EDTA, PESA, EDTMPS, and POCA, and duration of 120 min. EDTA, ethylenediaminetetraacetic acid; PESA, polyepoxysuccinic acid; EDTMPS, ethylenediamine tetra (methylene phosphonic acid) sodium; POCA, phosphonyl carboxylic acid copolymer (**a**, Pb in paddy soil; **b**, Pb in arid soil; **c**, Cd in paddy soil; **d**, Cd in arid soil)
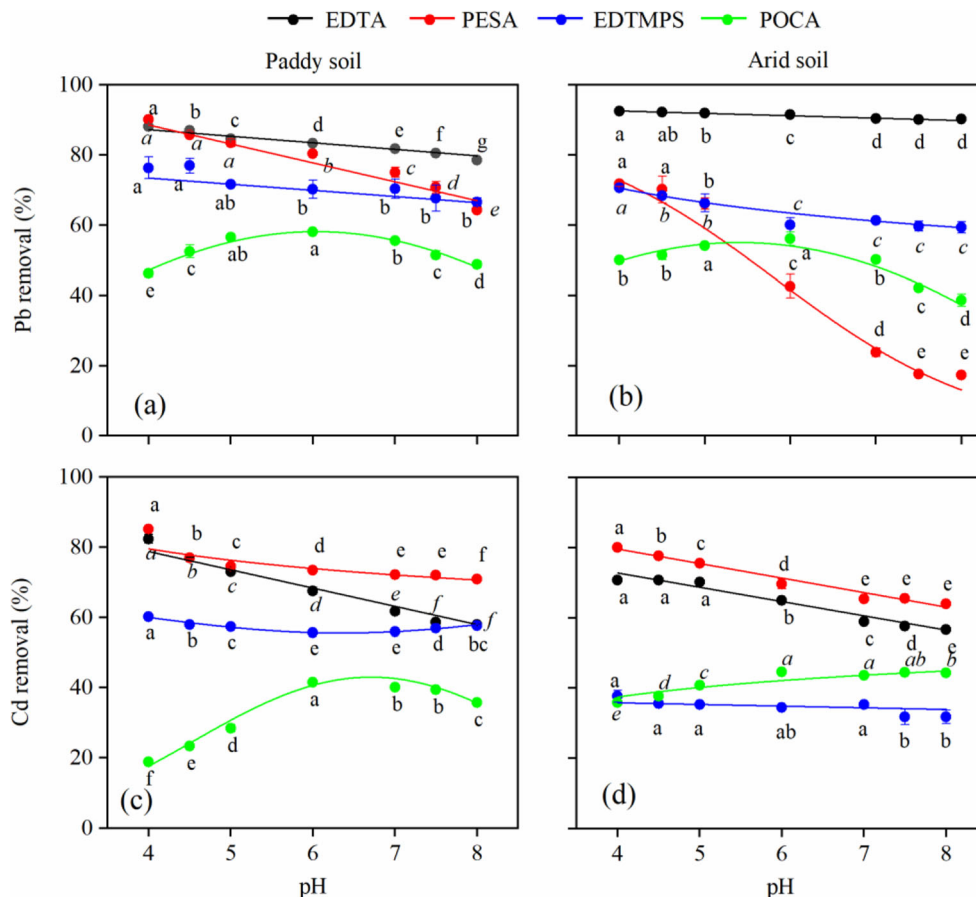
Fig. 4 Soil Pb and Cd removal during washing duration under the condition of 0.08 mol L^{-1} EDTA, PESA, EDTMPS, and POCA, and pH of 4.0. EDTA, ethylenediaminetetraacetic acid; PESA, polyepoxysuccinic acid; EDTMPS, ethylenediamine tetra (methylene phosphonic acid) sodium; POCA, phosphonyl carboxylic acid copolymer (**a**, Pb in paddy soil; **b**, Pb in arid soil; **c**, Cd in paddy soil; **d**, Cd in arid soil)
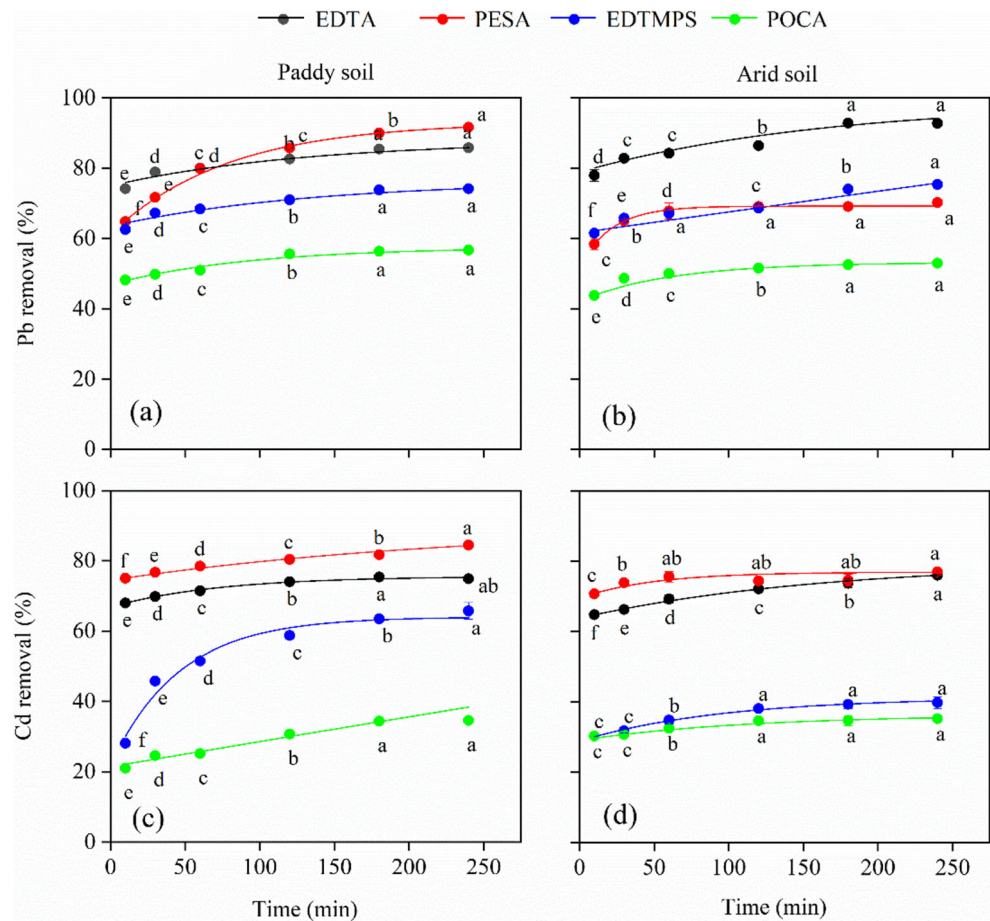
Duration

As shown in Fig. 4, high removal efficiencies of soil Pb from 60.0 to 80.0% occurred at shortest time (10 min) for EDTA, PESA, and EDTMPS washing the two soils (Fig. 4a and b), and slightly increased to 85.0–95.0% with duration extending to 180 min, and then arrived at plateau. Cd significantly was removed by EDTA and PESA with duration in two soils (Fig. 4c and d). The POCA showed the weakest capacity to remove soil Pb and Cd during the whole duration.

Risk assessment

Removal rates of labile fractions of soil Pb and Cd

The labile fractions of soil Pb and Cd extracted by HCl and DTPA were 920.31 and 882.79 mg kg^{-1} for Pb_HCl and _DTPA, 0.64 and 0.59 mg kg^{-1} for Cd_HCl and _DTPA in paddy soil, 1389.35 and 1272.54 mg kg^{-1} for Pb_HCl and _DTPA, and 2.41 and 2.14 mg kg^{-1} for Cd_HCl and _DTPA in arid soil, respectively (Table 1). The Pb leaching amounts by HCl and DTPA were removed by 93.5–98.3% by EDTA and PESA in two soils, and a lower reduction 13.4–

23.1% in those fractions was observed in EDTMPS and POCA treatments (Fig. 5a, b, e, and f). Similarly, greater decrease amounts around 83.6–91.7% in HCl and DTPA extracted Cd were observed after EDTA and PESA washing (Fig. 5c, d, g, and h), while those decreased less than 50.0% by EDTMPS leaching (Fig. 5c), and only reduced by 11.7–13.9% after POCA leaching in paddy soil (Fig. 5d).

Potential ecological risk index

Paddy soil had high values of *RI_Total* (270) and *RI_Labile* (385), and arid soil owned much higher values of *RI_Total* (673) and *RI_Labile* (1187) before soil washed (Fig. 6). In contrast, the two risk index values in unpolluted soil were lower than the value (40) of low risk. After washed, the *RI_Total* and *RI_Labile* values in polluted paddy soil reduced to 37 and 35 by PESA, to 44 and 37 by EDTA, while lower RI value reduction was observed by EDTAPS with *RI_Total* of 95, and *RI_Labile* of 147, and by POCA with *RI_Total* of 197, and *RI_Labile* of 220 (Fig. 6a). In arid soil, the values of *RI_Total* and _Labile dropped to 152 and 115 by PESA, to 162 and 96 by EDTA, to 396 and 224 by EDTMPS, and 423 and 232 by POCA, respectively (Fig. 6b).
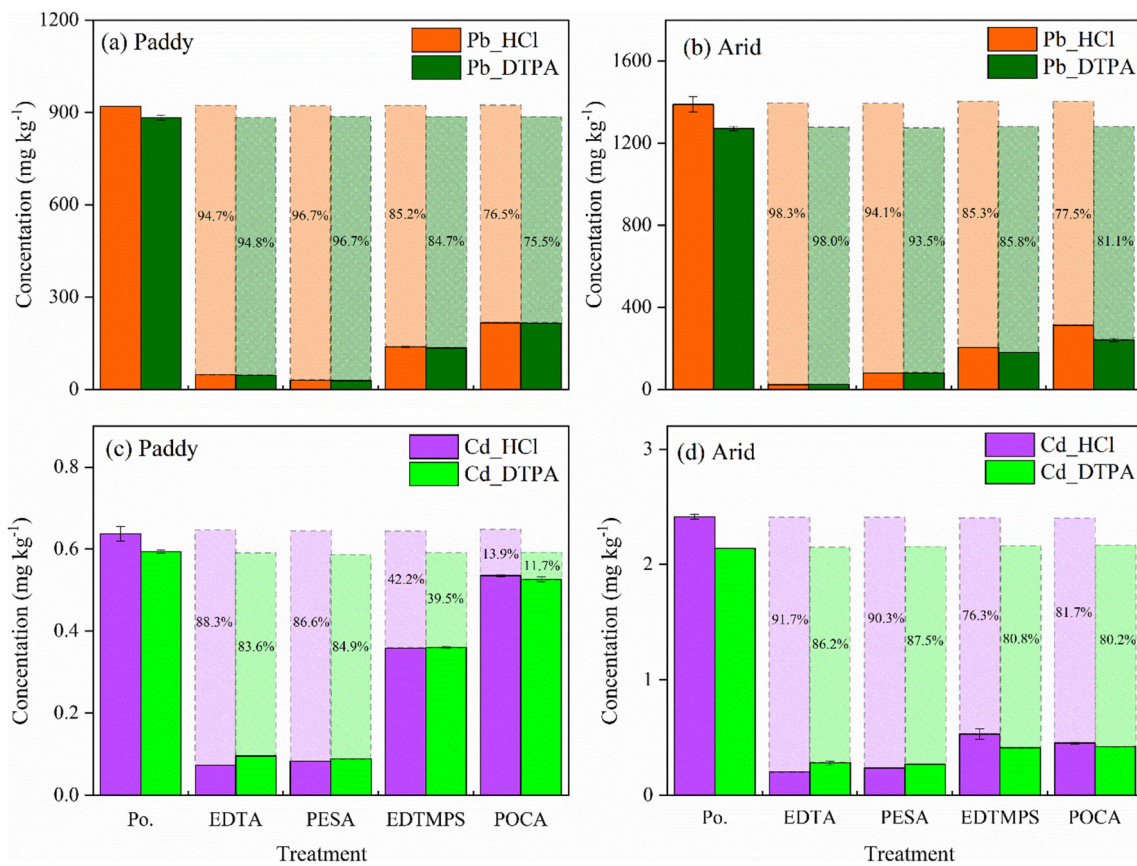
Fig. 5 The concentrations and removal efficiencies of heavy metal labile fractions in soils before and after washing with eluents. Pb_HCl, Pb concentration extracted by 0.1M HCl; Pb_DTPA, Pb concentration extracted by DTPA; Cd_HCl, Cd concentration extracted by 0.1 M HCl; Cd_DTPA, Cd concentration extracted by DTPA; Po., polluted soil; EDTA, ethylenediaminetetraacetic acid; PESA, polyepoxysuccinic acid; EDTMPS, ethylenediamine tetra (methylene phosphonic acid) sodium; POCA, phosphonyl carboxylic acid copolymer. The dark color indicates the concentration of labile fraction. The numbers in light color column represent the removal rates extracted by HCl and DTPA (**a**, Pb in paddy soil; **b**, Pb in arid soil; **c**, Cd in paddy soil; **d**, Cd in arid soil)

Discussion

Effects of single factor on heavy metal removal

Concentration

The eluents with different functional groups remove heavy metals through ion exchange, chelating, and electrostatic adsorption (Feng et al. 2020; Wang et al. 2020). In this study, PESA and EDTA had comparable higher capacity to remove Pb and Cd than the other two eluents in two soils, but they worked in different functional groups with various ways.

EDTA is a common chelator with hydroxyl, carboxyl, and amine groups. The increased peaks of –OH from hydroxyl groups indicated it may interact with Pb^{2+} and Cd^{2+} by coordination or ions exchange (Pladzyk et al. 2011). Similarly, the new peaks around 2300 cm^{-1} and 1750 cm^{-1} suggested that C=O complexed with metal ions (Alikhani and Manceron 2015). Additionally, these functional groups produced many ligands. The ligands not only can bind metal ions that weakly

adsorbed on the surface of soil colloid, but also increase dissolution of soil colloid to release heavy metal ions. These processes can be expressed as the below equations (Sparks 1989):

$$R–COOH \cdot H_2O \rightarrow R–COO– + H_3O^+$$
$$R–NH_2 \cdot H_2O \rightarrow R–NH_3 + +OH^-$$
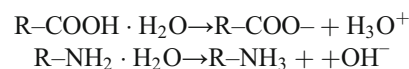
Differently, PESA, an oligomer, is composed of deprotonated carboxylic acid groups attached to carbon backbone (Pesonen et al. 2005). It has carboxyl, hydroxyl groups, and ether oxygen bonds working in several mechanisms. First, its carboxyl groups (–COOH) ionize hydrogen ions and carboxyl polyions in solution. On the one hand, the Pb^{2+} and Cd^{2+} adsorbed by soil colloids were easily replaced by hydrogen ions through cation exchange; on the other hand, the hydrogen ions enhance dissolution of metal ions, thus cooperate with carboxyl polyions to bind heavy metal ions in chelation and electrostatic adsorption (Piccolo et al. 2019). These behaviors were proved by increasing peaks of –OH and C=O of PESA after washing two soils (Fig. 1b). Second, PESA can act

Fig. 6 Potential ecological risk index of heavy metals in soils before and after washing with eluents. *RI_Total*, potential ecological risk index based on total concentration of heavy metal; *RI_Labile*, potential ecological risk index based on labile fraction of heavy metal; Un., unpolluted soil; Po., polluted soil; EDTA, ethylenediaminetetraacetic acid; PESA, polyepoxysuccinic acid; EDTMPS, ethylenediamine tetra (methylene phosphonic acid) sodium; POCA, phosphonyl carboxylic acid copolymer; classification level: <40, low risk (LR), 40–80, moderate risk (MR); 80–160, considerable risk (CR); 160–320, high risk (HR); and >320, very high risk (VHR) (**a**, paddy soil; **b**, arid soil)
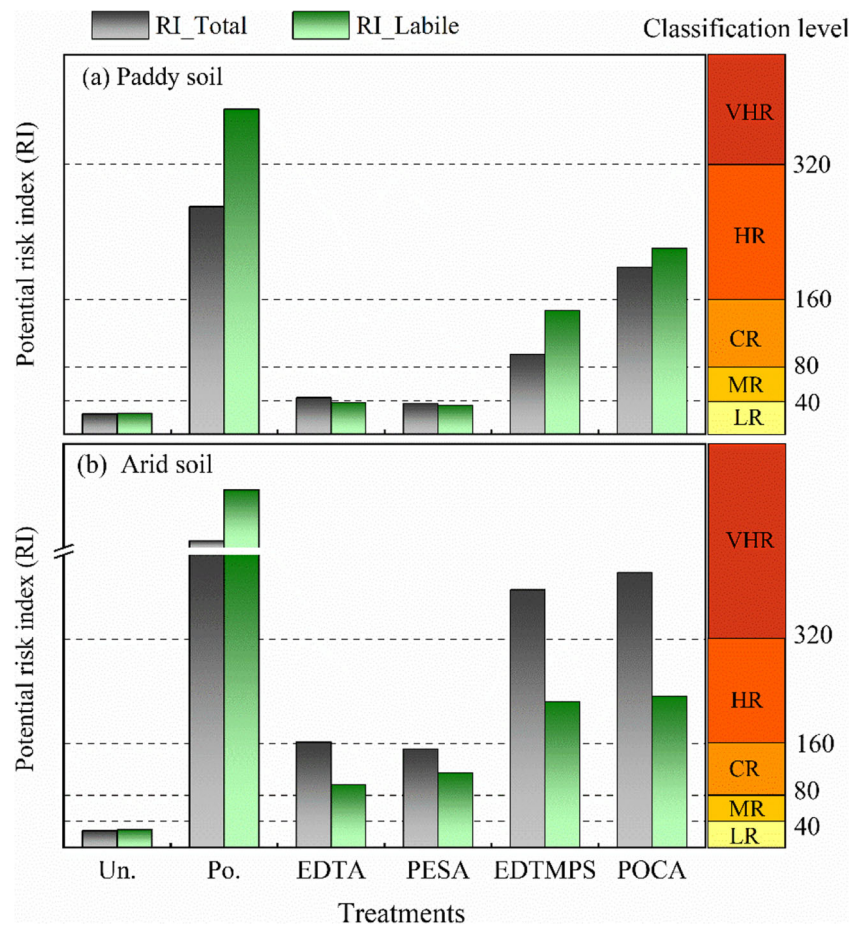
as a dispersing agent through electrostatic repulsion. PESA had strong electronegativity derived from the dissolution and ionization (Dong et al. 2021). It can chelate metal ions with hydroxy oxygen, ether oxygens, and metal-coordination carboxylate oxygens forming stable coordinated structure (Pesonen et al. 2005; Dong et al. 2021). We speculated that some negative charges of PESA (COO-) promoted it strongly adsorbed on soil colloid surface forming micro-aggregates, and left negative charges made these aggregates with the same electronegativity difficult to clump and precipitate (Chen et al. 2019). Thus, it broadened PESA molecular reaction contact area with soil particles, and promoted removal efficiencies of Pb and Cd in this study. Third, the significant difference with EDTA is PESA with ether oxygen atom backbone (C–O–C), which became narrow after washing two soils (Fig. S2b). It is more flexible than backbones only consisting of carbon atoms, which exerts prominent complexation capacity (Pesonen et al. 2005). For EDTMPS and POCA, the two eluents with carboxyl, hydroxyl, amide, and phosphine carboxyl groups theoretically have promising capacity to combine heavy metal ions (Chaussemier et al. 2015), while there were no such results observed in this study. The reason for limited capacity of EDTMPS could be that such nitrogen-containing reagents firstly favor of chelating trivalent irons (Pesonen

et al. 2005). For POCA, it might be because the copolymers have chelated heavy metal ions, but these chelate products with long chain easily generate chain entanglement, agglomeration, and precipitation.

It should be noted that no further significant change was observed for metal leaching by increasing eluent concentrations exceeding 0.1 mol L^{-1}. In the EDTA washing process, the limited removal capacity was probably because co-existing cations including alkaline-earth cations such as Ca^{2+}, Mg^{2+}, $Fe^{2+/3+}$, and Mn^{2+} occupied reaction sites of EDTA for the non-selective nature of this reagent (Gómeza et al. 2016), and it was also proved by the significant variation of peak range from 1000 to 400 cm^{-1} after EDTA washing measured by FTIR in this study (Fig. S2a), because the characteristic peaks of metal reaction with eluents generally occur in this spectral region (Tsang and Hartley 2014; Wei et al. 2018). Although the other three eluents also showed limited removal of soil Pb and Cd at higher eluent concentrations (Fig. 2b, c, and d), only slight variation of peaks was observed in that spectral region (Fig. S2b, c, and d). This may be because these eluents chelate some specific ions, e.g., Ca^{2+} or Mg^{2+} instead of all cations. Especially for PESA, it was reported to favor complexation with Ca^{2+} via hydroxyl oxygen atoms (Tamura et al. 1998; Pesonen et al. 2005). The less almost

20.0% removal efficiency of Pb in PESA washing than in EDTA washing for arid soil (pH=7.12), which had 3.9 times higher Ca concentration than in paddy soil, could be evidence of this speculation.

pH

The effects of pH on removal efficiencies of heavy metals are controlled by the amount of H$^+$ ions in the eluents (Dermont et al. 2008). High removal efficiencies of heavy metals at acidic condition have been documented in previous reports (Kulikowska et al. 2015; Feng et al. 2020; Xia et al. 2019), for H$^+$ ions may act in several mechanisms. First, abundant H$^+$ ions displace the heavy metal ions weakly adsorbed on soil colloid surface by physical forces including Van der Waals force, and this reaction process is important at close distance between liquid and solid phases. Second, these protons can bond with soil (hy)oxides, and weaken the binding force between soil (hy)oxides and heavy metal ions, then the oxides are dissolved via a surface-controlled reaction in two steps to release heavy metals (Stumm 1990):

$$\equiv MOH + H^+ \leftrightharpoons \equiv M-OH_2^+$$
$$\equiv M-OH_2^+ + L^- \leftrightharpoons \equiv M-L + H_2O$$
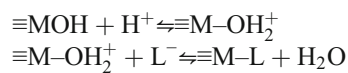
where $\equiv M-OH_2^+$ is metal-proton coordination compound and L is the organic ligands.

Third, H$^+$ ions with smaller hydrated radius and high flexibility can easily enter into interlayer space of soil minerals, which cause partial or total space collapse. This is followed by detachment of heavy metals fixed between interlayers of silicate into solution (Xia et al. 2019). However, under alkaline medium conditions, the desorption of heavy metal ions may be hindered by OH$^-$ as $\equiv MOH + OH^- \leftrightharpoons \equiv M-O^- + H_2O$, limiting the abilities of eluent functional groups (Im et al. 2015). This is evidenced by the removal efficiencies of Pb and Cd sharply decreased with higher pH in the PESA washing process. Moreover, the metal ions generally become inert ions, and losing their active; therefore, most of them precipitate in higher pH (Dijkstra et al. 2006).

In this study, the same trends were found in soils washed by EDTA, PESA, and EDTMPS, except for POCA in pH-dependent washing experiments. In the case of soil washed by POCA, lower removal efficiencies of Pb and Cd were found under acidic condition, a similar phenomenon was observed by Feng et al. (2020) using polyacrylic acid to wash soil polluted by Pb, Cd, and Zn. POCA also contains acrylic acid and thus might perform in the same mechanism, that is, the electrostatic field strength around POCA probably is weakened by the abundant H$^+$ ions, and the electrostatic adsorption capacity of POCA is inhibited.

Duration

Over 60.0% removal of soil Pb and Cd was observed in 10 min for the two soil by EDTA and PESA washing. A two-step process depending on time mainly documented in previous studies seemed not to occur in our study (Bermond and Ghestem 2001; Feng et al. 2020; Wang et al. 2020). This indicated that the first fast step already happened before 10 min, and labile fractions including water-soluble, exchangeable, and carbonate of Pb and Cd were the most fractions of total amount. These fractions are efficiently washed by EDTA and PESA in the short term, and implied that these fractions posed high environment risk in original soils. The labile fractions are sensitive to change of environmental conditions, and should be considered their potential environment risk during the washing process (Wang et al. 2019).

Changes of potential ecological risk of heavy metals by soil washing

The ecological risk for soil quality and sustainability in agro-ecosystems is a key index to evaluate the feasibility of some eluent (Feng et al. 2020), and should be investigated accurately (Kelepertzis 2014). Although the three eluents, especially PESA, had significant removal efficiencies in single factor experiments, their ecological risk for application should be assessed comprehensively.

According to *RI_Total* and *_Labile* values of heavy metals, paddy soil had a high risk and even very high risk, and arid soil owned a very high risk (Fig. 6). The values of risk degrees of total and labile fractions were 11–42-fold higher in polluted soils than in unpolluted soils (lower than 40), indicating that the paddy and arid soils might be threatened by Pb and Cd pollution. The ecological risk of total heavy metal concentrations reduced from high risk to low risk by PESA washing, to moderate risk by EDTA washing in paddy soils, and those risks were considerable or high risk when soil washed by EDTMPS and POCA (Fig. 6a). In arid soil, the risk degree of total heavy metal was decreased from very high risk to high risk after EDTA washing, or to considerable risk after PESA washing (Fig. 6b). In original polluted soil, the labile fractions were significantly removed by the eluents, and posing a less risk after washing. EDTA and PESA effectively removed not only the labile fractions but also the relatively resistant fractions since the removal amounts were obviously higher than the labile fraction concentrations of soil Pb and Cd (Table S6). They reduced the very high risk to low risk in paddy soil (Fig. 6a), and to considerable risk in arid soil (Fig. 6b). While EDTMPS and POCA might remove most labile fractions and a little of the other fractions, thus, soil still remained at high or considerable risk. Therefore, these eluents significantly reduced the risk of the polluted soil, particularly, PESA

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