Removal of Pb by EDTA-washing in the presence of hydrophobic organic contaminants or anionic surfactant

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Abstract

Heavy metals and organic contaminants often coexist in contaminated soils, of which the remediation requires a combined or sequential use of surfactant and chelant in chemical-enhanced soil washing. This study investigated the Pb removal by EDTA-washing in the presence of the coexisting hydrophobic organic contaminants (marine diesel fuel, MDF) or anionic surfactant (sodium dodecyl sulfate, SDS). The results of batch experiments indicated a negative impact of MDF on Pb removal, whereas a positive or negative influence of SDS depending on the molar ratio of [EDTA]:[Pb]. The surface of MDF-contaminated soil was partially covered by free-phase MDF limiting the interaction between EDTA and sorbed Pb, especially at [EDTA]:[Pb] = 1:1. Despite the ability of SDS itself for extracting Pb to some extent, probably through electrostatic interaction and dissolution of soil organic matter (SOM), the addition of SDS into EDTA solution only slightly enhanced Pb removal at [EDTA]:[Pb] = 1:1 but inhibited at [EDTA]:[Pb] = 2:1. This may be attributed to the SOM dissolution and the potential formation of ternary surface complexes, respectively. Along the same line of reasoning, it was not surprising that the optimal sequence for Pb removal was EDTA- followed by SDS-washing rather than the reverse sequence or simultaneous EDTA- and SDS-washing.
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1. Introduction

Lead (Pb), one of the most extensively encountered heavy metals in the environment, usually displays high adsorption affinity on soils. Heavy metals are generally known to be sorbed on soils through ion exchange, surface precipitation, as well as inner-sphere surface complexation; the latter two can be referred to as specific adsorption [1]. Inner-sphere surface complexation of heavy metal ions, which is more relevant to Pb sorption in field settings than ion exchange and surface precipitation, involves proton exchange with hydroxyl groups on soil minerals (e.g. Al, Fe and Mn oxides and clay minerals) or functional groups (mainly carboxylic and phenolic) of soil organic matter (SOM) to form partial covalent bonds [2,3].

Chelating agents, such as ethylenediaminetetraacetic acid (EDTA), can form stable and soluble complexes with heavy metals and thus substantially increase heavy metal removal from contaminated soils. In view of high stability constants of EDTA with heavy metals, EDTA-washing has been extensively studied for remediation of contaminated soils [4–8]. Based on the risk assessment report on EDTA approved by the Technical Meetings of European Union member state representatives [9], EDTA was reported to have low aquatic toxicity and no bioaccumulation in living organisms through the food chain. The concern about EDTA application mainly arises from the enhanced mobility of heavy metals, eutrophication and nutrient deficiency that the use of EDTA should be discouraged in those sites that use large volumes of EDTA in the absence of effective biodegradation or high effluent dilution. In addition, although EDTA-washing was demonstrated to be widely applicable and

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economically feasible with acceptable environmental risk in a number of studies on the recovery of added EDTA and heavy metals from washing effluents [10–14], most of these previous studies, however, primarily focused on soils contaminated with heavy metals only. Instead, according to statistical data of Compensation, and Liability Act Information System by USEPA in 2003 [15], 67 % sites on the National Priority List were contaminated by both heavy metals and organic contaminants. Similarly, most of the contaminated sites in Hong Kong, especially former shipyards, gas stations and vehicle repair workshops, were often found to be contaminated by heavy metals and petroleum hydrocarbons together. For instance, the site investigation report of eleven shipyards in North Tsing Yi indicated that Pb and hydrocarbons of C_{15}–C_{25}, which correspond to the major fractions of marine diesel fuel (MDF), were the most commonly found contaminants. Compared with the understanding of the strong correlation between SOM and heavy metals [16–18], the interaction between coexisting organic contaminants and heavy metals in soils, such as MDF and Pb in former shipyards, has often been overlooked. Few recent studies [19,20] found that phenanthrene sorption was significantly higher and less reversible in heavy metal-contaminated soils. Such enhancement of PAH sorption in the presence of heavy metals was ascribed to the greater adsorption of dissolved organic matter (DOM) on soils [19], or to cation-π bonding between PAHs and exchangeable cations on mineral surfaces [21]. The potential influence of coexisting organic contaminants on heavy metal adsorption and extraction, on the other hand, should be investigated.

To remediate soils contaminated with both heavy metals and hydrophobic organics, more than one chemical agent may be needed [22,23]. Regarding the extraction of organic contaminants from soils, surfactants (e.g. anionic sodium dodecyl sulfate, SDS) that can increase the mobility and solubility of hydrophobic organics have been widely proven useful [24,25]. Thus, a sequential or concurrent use of EDTA- and SDS-washing appears to be a plausible method. It has been suggested that surfactants alone could extract heavy metals from soils directly by ion exchange and electrostatic attraction, and indirectly by facilitating the dissolution of SOM on which metals were adsorbed [26–28]. Nevertheless, it remains unclear about the contribution of SDS to heavy metal removal in the presence of EDTA of concentrations corresponding to different molar ratios of EDTA to heavy metal. More importantly, different sequences of SDS and EDTA addition in soil washing may undergo different predominant mechanisms and in turn extraction effectiveness.

In our previous study [29] that focused on the physicochemical interactions among Pb, MDF in contaminated soils and the added EDTA and SDS in washing solution, some preliminary findings underlined the importance of studying the inherent mechanisms of Pb removal by EDTA-washing in the presence of hydrophobic organic contaminants or anionic surfactant. Therefore, the objectives of this study are to scrutinize the following issues: (1) how MDF in solution or in soils influenced the Pb removal efficiency by EDTA; and (2) in which form the Pb existed in the contaminated soils and how these forms influenced the Pb removal by EDTA in the presence of 5000–25,000 mg/l SDS in solution.

2. Materials and methods

2.1. Contaminants and chemicals

In this study, MDF (British Petroleum Corp., UK) and Pb (AR grade $Pb(NO_3)_2$, Fisher Scientific Corp.) were selected as the representative hydrophobic organic contaminant and heavy metal, respectively, based on the environmental investigation of former shipyards in Hong Kong. SDS ($C_{12}H_{25}SO_4$, GPR grade, Fisher Scientific Corp.) and EDTA (tetra-sodium salt, Invitrogen, AR grade) were used as chemical agents in soil washing. All chemicals were used without further purification.

2.2. Soil contamination

Completely decomposed granite, one of the most commonly found soils in Hong Kong, was used for artificial contamination in this study. The soil was air-dried at room temperature, and pre-sieved with a US standard No.10 sieve (9 mesh) to remove coarse components and stones. Table 1 summarizes the soil properties along with the measurement methods and instruments. This soil was suitable for the soil washing treatment [29] because of its high sand content (~92%) and relatively low amount of clay and silt (~7.2%) and organic matter content (~0.023% as total organic carbon, TOC). Background levels of heavy metals such as Pb, Zn, Cr and Cu were negligible compared with subsequent contamination levels and would not significantly interfere with subsequent experiments.

Artificial contamination of the soil was employed to investigate the factors of interest with minimal interference from other variations that could arise in the case of field-contaminated soils, although it should be noted that the extraction effectiveness would be somewhat higher than that of field soils where contamination aging of decades takes place. To prepare Pb-contaminated soils, air-dried soil was mixed with 200 mL of $Pb(NO_3)_2$ solution (8 g in 200 mL) in a 250-polyester bottle, shaken end-over-end at 60 rpm for 6 h. The suspension was then separated using a Beckman Auegra™ 6 Centrifuge at 3000 rpm for 10 min. The supernatant fraction was collected and filtered through a G50 glass fiber filter for measurement of Pb concentration; the soil was air-dried at room temperature for 7 d. To investigate the influence of coexisting MDF, an additional step of MDF-contamination was applied. The Pb-contaminated soil was mixed with commercial MDF (Table 1) at 1 g mL^{-1} that had been dissolved in minimum amount of acetone solution (GPR grade, Fisher Scientific Corp.), shaken end-over-end at 60 rpm. The mixture was then ventilated in a fume hood for 8 d to stabilize the MDF contamination, which was adequate to evaporate all added acetone and the majority of light components of MDF in preliminary experiments.

2.3. Soil washing

During soil washing, 1 g of contaminated soil and 25 mL of EDTA- and/or SDS-washing solution (pH 7 which was the optimal pH indicated by previous study [8], adjusted by HNO$_3$ or NaOH) were completely mixed at a speed of 60 rpm for 2 h,

Table 1
Properties of uncontaminated soils and MDF

Soil properties	Value	Method	Measurement instrument
pH	6.2	Method 9045 in USEPA SW-846	350 corning pH/ion analyzer
Surface area	6.09 m^2 g^{-1}	BET method with N$_2$ as adsorbate	Coulter SA3100 surface area and pore size analyzer
Cation exchange capacity	10.9 meq/100 g	Method 9081 in USEPA SW-846	Z-8200 polarized Zeeman atomic absorption spectrophotometer
Total carbon	0.079%		5000A Shimadzu total organic carbon analyzer
Inorganic carbon	0.056%		
Organic carbon	0.023%		
Particle size distribution		ASTM soil classification	Endecotts ISO3310-1 laboratory test sieves
Silt and clay (<75 μm)	7.24%		
Fine sand (<425 μm)	34.3%		
Medium sand (<2 mm)	58.4%		
Heavy metal content		USEPA SW-846	Z-8200 polarized Zeeman atomic absorption spectrophotometer
Pb	<1.0 mg kg^{-1}	Method 7420	
Zn	<0.1 mg kg^{-1}	Method 7950	
Cu	1.4 mg kg^{-1}	Method 7210	
Cr	<0.6 mg kg^{-1}	Method 7190	

MDF properties (http://www.bp.com)	Unit	Value
Maximum viscosity at 40 °C	mm^2/s	11.0
Maximum density at 15 °C	kg/m^3	900
Maximum water content	%(v/v)	0.25
Minimum flash point	°C	62
Maximum sulfur content	% (m/m)	1.5
Maximum carbon residue (microcarbon)	% (m/m)	0.20
Maximum sediment content	% (m/m)	0.02
Maximum total existent sediment	% (m/m)	0.10
Maximum ash content	% (m/m)	0.01

as preliminary experiments showed that the majority of sorbed Pb was released from soil in the first 2 h. The suspension was then centrifuged using a Beckman Auegra^{TM} 6 Centrifuge at 3000 rpm (1610 × g) for 10 min and the supernatant fraction was decanted and filtered with a G50 glass fiber filter for further analyses. In this study, control experiment of EDTA-washing on 1376 ± 34 mg kg^{-1} Pb-contaminated soil at molar ratios of EDTA to Pb ([EDTA]:[Pb]) of 1:1 and 2:1 was referred to as benchmark to evaluate the influence of MDF in soils, SDS alone, and SDS together with EDTA on the removal of Pb in Exp. 1–3. The Pb removal effectiveness of three sequences of EDTA and SDS addition, in the absence of MDF, were compared in Exp.

4: EDTA followed by SDS addition, SDS followed by EDTA addition, and simultaneous addition of EDTA and SDS. The experimental parameters and conditions are listed in Table 2. All experiments were carried out at least in duplicate.

Two supplementary experiments (Table 2) were performed to study the potential influences of coexisting MDF. Exp. S1 was conducted in a way similar to Exp. 1 except that MDF was present in solution rather than adsorbed on soil, because preliminary experiments had shown that a portion of MDF in soils was released into the solution during EDTA-washing. To investigate the influence of MDF on Pb sorption, Exp. S2 was carried out on soil with simultaneous Pb- and MDF-contamination, in which

Table 2
Parameters of batch experiments for soil washing

Index		Contaminants		Washing solution	
		[Pb] (mg kg^{-1})	[MDF]	EDTA ([EDTA]:[Pb])	[SDS] (mg L^{-1})
Control[a]	EDTA alone	1376 ± 34	Nil	1:1; 2:1	Nil
Exp. 1	MDF in soils	1376 ± 34	1562 ± 23; 4740 ± 85 (mg/kg soils)	1:1; 2:1	Nil
Exp. 2	SDS alone	1376 ± 34; 844 ± 22	Nil	Nil	5,000; 20,000
Exp. 3	SDS & EDTA	1376 ± 34	Nil	1:1; 2:1	5,000; 10,000; 15,000; 20,000; 25,000
Exp. 4[b]	Sequence of EDTA & SDS addition	1376 ± 34	Nil	1:1; 2:1	5,000
Exp. S1 and S2	MDF in solution during Pb sorption or EDTA-washing	1376 ± 34	1.8; 4.5 (mg/25 mL solution)	1:1; 2:1	Nil

[a] Control experiment for the Exp. 1–3 and S1.

[b] Three sequences: EDTA followed by SDS, SDS followed by EDTA, and simultaneous addition of EDTA and SDS.

1.8 mg (completely dissolved) or 4.5 mg (partially dissolved) of MDF was present in $Pb(NO_3)_2$ solution during contamination process. The soil was then washed by EDTA solution as described above.

2.4. Metal speciation of Pb-contaminated soil

The distribution of sorbed heavy metal and its binding strength in Pb-contaminated soil can be investigated with selective sequential extraction of (1 g, run in duplicate) [30–32]. It should be noted that the fractions were operationally defined to provide qualitative information. The sequential extraction procedures developed by Tessier et al. [33] were adopted with minor modifications: 1 M $MgCl_2$ solution in the first extraction (exchangeable fraction) and microwave digestion with HNO_3 (70%), HF (48%), and HCl (37%) in the last extraction (residual fraction). The mixture was centrifuged at 3500 rpm ($2191 \times g$) for 10 min after each extraction step. The supernatant fraction was decanted for Pb concentration measurement and the soil was rinsed with 10 mL of deionized water before next extraction step.

2.5. Chemical analyses

The concentration of Pb in solution was measured using a Z-8200 Polarized Zeeman atomic absorption spectrophotometer based on the standard method 7402 in USPEA SW-846. MDF in soils was firstly extracted by ISCO SFX ^{1M} 220 supercritical fluid extractor (SFE), and then analyzed by gas chromatography with a flame ionization detector (GC/FID). The operating conditions of SFE were based on the USEPA SW-846 Method 3560 as follows: 1 g of soil sample was extracted in the dynamic mode for 30 min with supercritical CO_2 at 5000 PSI, 80 °C and a gas flow rate of 1 mL min^{-1}; the extracted MDF was collected in a vial containing 10.0 mL of *n*-hexane (HPLC grade, Fisher Scientific Corp.); and the temperature of the restrictor was maintained at 80 °C during the extraction procedure. The measurement of MDF with GC/FID was conducted using a Hewlett Packed 5890 Series II capillary GC equipped with FID under the following operating conditions: 1 μL MDF extract in hexane was injected into a 30 m AT_5 fused silica capillary column with 0.25 mm inter-diameter (Alltech, USA); the flow rate of the helium carrier gas was 1.0 mL/min; and the temperature in the column was initially held at 45 °C for 5 min and then increased from 45 to 225 °C at a rate of 12 °C/min, and thereafter held at 225 °C for 12 min. The dissolved amount of SOM was measured by UV absorption intensity at a wavelength of 254 nm using Milton Roy UV/visible Spectrometric 3000 [34,35].

3. Results and discussion

3.1. Influence of MDF in soils on Pb removal by EDTA

Exp. 1 was conducted to investigate the influence of coexisting MDF in soils on Pb removal by EDTA-washing. As shown in Fig. 1(a), for the 4740 mg kg^{-1} MDF-contaminated soil, 25 and 8% decrease in Pb removal efficiency was observed at [EDTA]:[Pb] = 1:1 and 2:1, respectively. The Pb removal
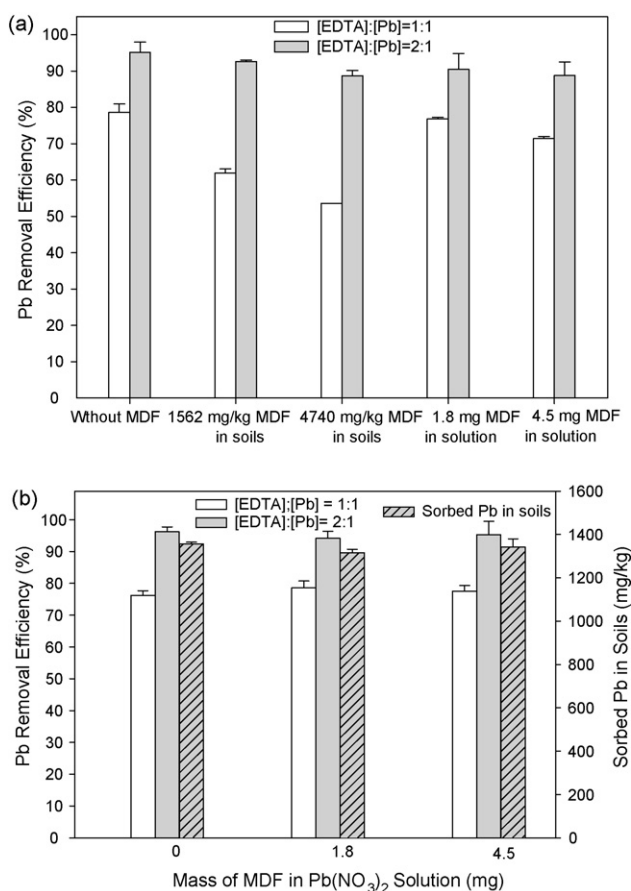

Fig. 1. Influence of MDF on Pb Removal by EDTA: (a) MDF in contaminated soils (Exp. 1) and MDF in solution (Exp. S1); and (b) MDF in $Pb(NO_3)_2$ solution for the preparation of Pb-contaminated soils (Exp. S2).

by EDTA-washing was apparently hindered in the presence of MDF, which may: (1) alter the chelating capacity of EDTA; (2) influence the amount and affinity of sorbed Pb; or (3) exist as free phase that adhere on the soil surface due to hydrophobic forces. Supplementary experiments were therefore carried out to study these effects independently.

The results of Exp. S1 (Fig. 1(a)) indicated that a larger amount of MDF in solution led to a greater decrease in Pb removal, the reason for which remains ambiguous. The magnitude of such decrease was, however, less appreciable (<8%). Since only 11% of MDF (equivalent to 0.417 mg MDF) was released from soils by EDTA-washing in preliminary experiment, the corresponding impact should be even less than that of Exp. S1. Besides, the amount of Pb sorbed onto soils was not influenced statistically in Exp. S2 (Fig. 1(b)), indicative of negligible interaction between Pb and MDF during sorption. This was corroborated by nearly the same Pb removal efficiencies (Fig. 1(b)) at a given [EDTA]:[Pb] ratio.

To identify whether free-phase MDF was present on the soil surface, the MDF-contaminated soil was scanned using an optical microscope. The images (not shown) at a magnification of 200 times illustrated the presence of some red spots that was reflective of free-phase MDF on the soil surface (because the commercial MDF is a red, non-aqueous liquid). Although the surface coverage of MDF could not be quantified, it was

obvious that a part of soil surface, as well as sorbed Pb, was physically isolated from EDTA-washing solution by free-phase MDF coverage, which probably resulted in the above-mentioned decrease of Pb removal efficiency. It is noteworthy that the influence of MDF in contaminated soils was more noticeable at lower EDTA concentration, i.e., smaller [EDTA]:[Pb] ratio in Fig. 1(a). This was because a more concentrated EDTA solution could extract more MDF from the soils (11 and 21% removal of 4,281 mg kg^{-1} MDF at [EDTA]:[Pb] = 1:1 and 2:1, respectively), probably through EDTA-promoted dissolution of soil minerals and organic matter [17,36].

3.2. Pb removal by SDS alone

The capacity of SDS itself for Pb removal is presented in Fig. 2. Using 5000 and 20,000 mg/L SDS solution (critical micelle concentration of SDS in the absence of soils is 2418 mg/L, beyond which substantial amount of micelles form in solution), 18 and 28% of Pb was extracted from 844 mg kg^{-1} Pb-contaminated soil (while 23 and 30% of Pb from 1376 mg kg^{-1}), respectively. Because no Pb was extracted by washing with deionized water, it was evident that SDS is able to extract some sorbed Pb, to which the extent depends on SDS concentration. In addition, a higher Pb removal efficiency was attained in soils with higher initial Pb concentration, where a larger portion of sorbed Pb would be weakly bound [30].

SDS itself could extract some sorbed Pb via ion exchange, electrostatic attraction [26,27] and SOM dissolution [16,17,35]. Upon SDS dissociation in water, sodium cation (Na$^+$) may replace some exchangeable Pb on soil surfaces and dodecyl sulfate anion (DS$^-$) has negative hydrophilic head that attracts Pb cations. The above mechanisms are only applicable for weakly bound Pb, whereas SOM dissolution driven by hydrophobic partitioning into SDS micelles can extract Pb that is strongly bound to SOM. Fig. 3 is expressed in terms of UV absorption intensity because the concentration of DOM is linearly correlated to the UV absorption [37], provided that fractionation of different molecular weight of humic substances (i.e., higher sorption affinity for high-molecular-weight fraction) is insignificant [38,39]. The extent of SOM dissolution induced by SDS
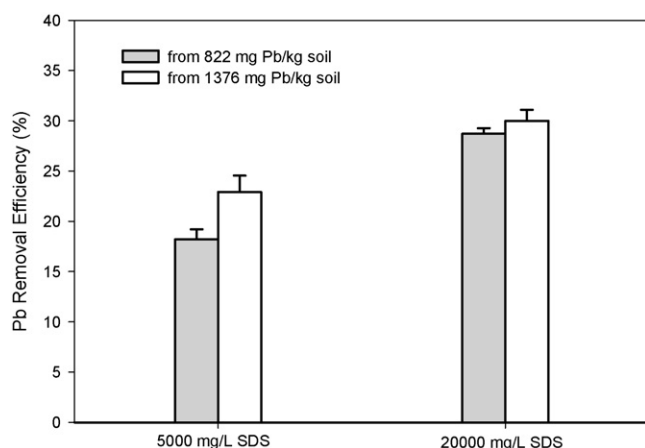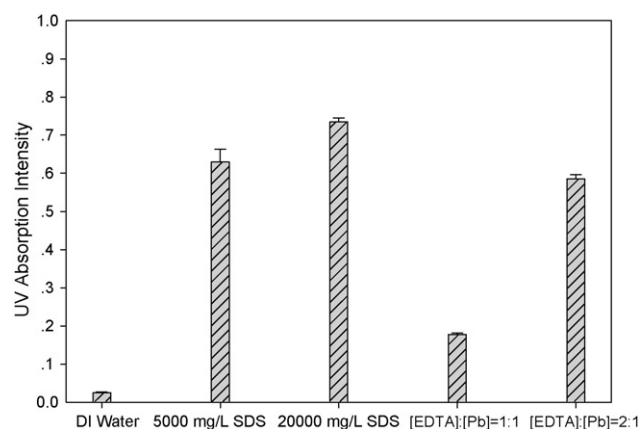

Fig. 3. SOM dissolution (expressed as UV absorption intensity) by SDS and EDTA solution alone. Experimental condition: 25 mL washing solution/g soils, pH 7.0, shaken at 60 rpm for 2 h.

solution was significantly greater than that by EDTA solution and deionized water; therefore, SOM-bound Pb was probably extracted along with SOM dissolution induced by SDS.

3.3. Pb removal by EDTA in the presence of SDS

Because of a relatively low Pb removal efficiency of SDS, EDTA addition appears to be necessary for efficient Pb removal. Thus, the influence of the presence of SDS in EDTA-washing solution was investigated in Exp. 3, of which the results are shown in Fig. 4. In order to eliminate misleading statistic deviation, an one-way analysis of variance (ANOVA) was used to test whether SDS concentration ranging from 0 to 25,000 mg/L in EDTA solution indeed influenced the Pb removal at [EDTA]:[Pb] = 1:1 or 2:1. Based on the ANOVA statistic analyses, Pb removal in the presence of SDS was about 5% larger at [EDTA]:[Pb] = 1:1 but nearly 10% less at [EDTA]:[Pb] = 2:1. The trend was more or less independent of SDS concentration in the studied range. These findings are seemingly paradoxical as SDS itself could enhance Pb removal as shown in the earlier experiments.
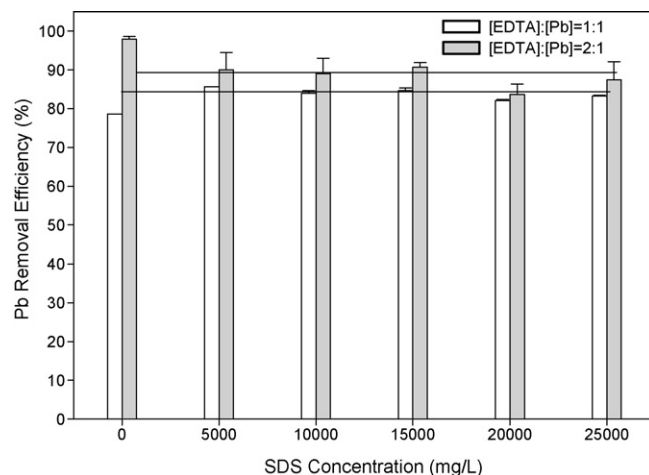

Fig. 4. Influence of SDS at various concentrations on Pb removal by EDTA (Exp. 3).

Fig. 2. Pb Removal by SDS and EDTA alone (Exp. 2).

The contrasting trends are unlikely to arise from weakly bound Pb that can be largely extracted by either EDTA or SDS, but probably from strongly bound (e.g., on SOM and Fe/Al/Mn oxides) and entrapped (e.g., in micropores of aggregates) Pb, which can play important roles in two ways. Because SOM has a high affinity for metals and it is a binding agent for aggregation of soil components, greater SOM dissolution induced by SDS compared with EDTA (Fig. 3) would enhance Pb extraction, as a result of the release of SOM-bound Pb [16,17,35]. On the other hand, ternary surface complexes of Pb, DS$^-$, and oxide surface may form and inhibit Pb extraction by EDTA. It was found that the residual (i.e., nonextractable) Pb on goethite, kaolinite, and montmorillonite was substantially increased in the presence of sulfate, which was attributed to the formation of ternary surface complexes [40,41]. Recent FTIR and EXAFS spectroscopic studies confirmed that, depending on solution pH and ligand concentration, type A (ligand-metal-oxide) or type B (metal-ligand-oxide) ternary surface complexes of sulfate or organic ligands (e.g., humic acid) can be formed [42–45]. The formation of ternary surface complexes of DS$^-$ was plausible in this study, of which direct evidence requires spectroscopic study in future.

Both SOM dissolution and ternary surface complexation would take place in the presence of SDS, however, it appears that the former was dominant at [EDTA]:[Pb] = 1:1, whereas the latter was influential at [EDTA]:[Pb] = 2:1. The speciation of Pb in soil (based on sequential extraction results) can reflect increasing binding strength and help explain the difference of Pb extraction [30,31,35,39]. The Pb speciation was operationally defined as: exchangeable fraction (59.4%), bound to carbonates (18.5%), bound to Fe/Mn oxides (19.8%), bound to NOM (1.25%), and residual fraction (3.4%). Therefore, it seems that 78.6% Pb removal at [EDTA]:[Pb] = 1:1 without SDS (Fig. 4) mainly corresponds to weakly bound Pb, i.e, exchangeable and bound to carbonates. Although a small part of strongly bound Pb on Fe/Mn oxides could be EDTA-extractable at [EDTA]:[Pb] = 1:1 [46,47], ternary surface complexes of DS$^-$ and Pb that may form on oxide surface would not significantly hinder Pb removal. Nevertheless, most Pb (97.9%) could be extracted at [EDTA]:[Pb] = 2:1 without SDS (Fig. 4), indicating substantial extraction of strongly bound Pb on oxides and SOM in the presence of excessive EDTA [8,35]. Under such circumstance, the possible formation of ternary surface complexes could significantly suppress the EDTA extraction of Pb.

3.4. SDS and EDTA addition sequence on Pb removal

The above results indicated that the sequence of SDS and EDTA addition is likely to influence the overall Pb removal efficiency. As shown in Fig. 5, EDTA-washing followed by SDS-washing achieved the highest Pb removal efficiency, whereas the reverse sequence attained the lowest. Simultaneous use of EDTA and SDS was also found to be unfavorable. EDTA-washing at first can extract the majority of Pb from soil, reducing the potential amount of ternary surface complexes in subsequent SDS-washing. Comparing the individual contributions of EDTA and SDS to Pb removal, EDTA concentration was most signif-
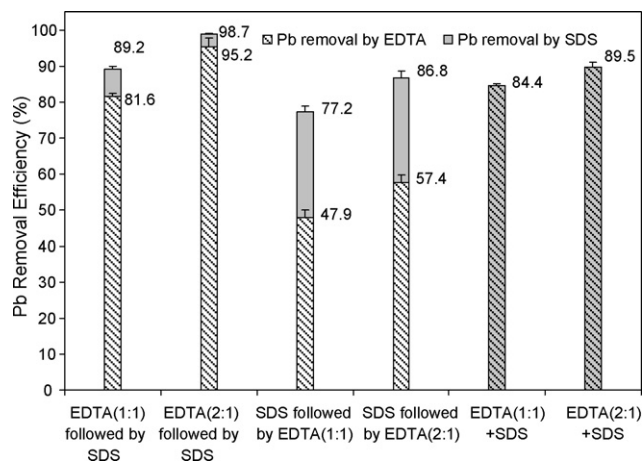

Fig. 5. Pb removal efficiencies attained by different sequences of EDTA and SDS addition (Exp. 4).

icant and the presence of SDS was slightly more beneficial at lower EDTA concentration. In line with the results of previous experiments, EDTA- followed by SDS-washing was, therefore, the optimal sequence for Pb removal.

4. Conclusions

Soils have prevalently been contaminated by both heavy metals and organics, whereas previous studies have focused on only one type of contaminant and one type of chemical agent accordingly. The influences of coexisting MDF and SDS (used for MDF removal) on Pb removal by EDTA-washing were therefore investigated in this study. Results indicated that the presence of MDF in contaminated soils partially covered the soil surface and thereby reducing Pb removal, especially at [EDTA]:[Pb] = 1:1. On the other hand, although SDS alone was found to enhance Pb removal and more concentrated SDS resulted in higher Pb removal, the presence of SDS slightly increased but inhibited Pb removal at [EDTA]: [Pb] = 1:1 and 2:1, respectively. Different mechanisms associated with SDS, including electrostatic interaction, SOM dissolution, and formation of ternary surface complexes, would be dominant depending on the EDTA concentration as well as the sequence of EDTA and SDS addition. Accordingly, the sequence of EDTA- followed by SDS-washing was demonstrated to attain the highest Pb removal.

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