COMPARISON OF SINGLE AND COMPOUND WASHING OF REMEDIATING PB CONTAMINATED SOIL OF NON-FERROUS SMELTERS

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(Received 13th Jul 2019; accepted 25th Nov 2019)

Abstract. Orthogonal and single factor experiments were used to investigate the remediation effect of single and compound washing on Pb contaminated soil from a typical non-ferrous metal smelter. The optimal washing conditions and speciation distribution of Pb were determined. The obtained removal efficiency of Pb from the soil is 68.35%, which could be remedied to less than "risk intervention values (Pb < 2500 mg·kg⁻¹) for soil contamination of the development land" in soil environmental quality standard China (GB36600-2018) when the compound ratio of HCl & Rhamnolipid is 1:1, the ratio of liquid-solid is 10:1 and the washing time is 1080 min. The removal efficiency of Pb are significantly higher when using compound eluents compared to single eluents, such as HCl, EDTA, oxalic acid and rhamnolipid. Moreover, compound eluents can effectively remove active Pb from soil, which greatly reduces their bioavailability and environmental risk.

Keywords: heavy metal pollution, soil washing, rhamnolipid, speciation distribution, kinetic analysis

Introduction

Global industrialization is accelerating at a continuous rate. The development of the nonferrous metal mining, smelting and processing industry in particular has led to the severe degradation of soils due to heavy metal pollution, and such contamination incidents have also attracted much attention (Park et al., 2002; Li et al., 2013; de Andrade Lima and Bernardez, 2017). Many experimental remediation technologies for heavy metal contaminated soils, such as soil exchange, solidification and stabilization, vitrification, and phytoremediation microbial remediation, have been developed (Dermont et al., 2008; Voglar and Leštan, 2011; Reza et al., 2015). However, they are difficult to implement in practical remediation projects of heavy metal contaminated soil due to their limitations, characteristics, secondary pollution, cost end effect on contaminated soil (Moutsatsou et al., 2006; Chen et al., 2017).

Soil contaminated by heavy metals in non-ferrous metal smelters is characterized by its high pollution intensity, complex forms, and is often accompanied by other heavy metals, which brings great challenges to its remediation (Bacon and Dinev, 2005; Burges et al., 2017). However, chemical washing has a promising application prospect as it is highly efficient, low cost, and has a short cycle (Abumaizar and Smith, 1999; de Andrade Lima and Bernardez,

2017). The type of eluent is the key to the repair effect, and the mechanism and removal capacity of different types of eluents vary. Common eluents include: inorganic eluents, artificial chelating agents, natural organic acid, biological surfactant and compound eluents (Dermont et al., 2008). For example, inorganic eluents can quickly and effectively remove heavy metals from the soil, and is low cost, but may damage the soil structure and biology. Artificial chelating agents can effectively remove heavy metals by self-chelating, but they are not easily biodegradable, and therefore cause secondary soil pollution. The costs of biological surfactants are often too high, which limits their engineering applications (Chen et al., 2017). Compound washing can not only improve the removal efficiency of pollutants, but also avoids secondary pollution and high cost associated with a single eluent through the synergistic solubilization of compound reagents.

In this paper, Pb contaminated soil from typical non-ferrous metal smelter was selected as the research object, and remedial target is to satisfy with the requirement of the "Risk control standard for soil contamination of development land" in soil environmental quality standard China (GB36600-2018). Hydrochloric Acid (HCl), Oxalic Acid, Ethylene Diamine Tetraacetic Acid (EDTA) and Rhamnolipid were selected as eluents, and oscillating washing experiments were carried out on the contaminated soil by single and compound washing. The optimal combination of eluents and conditions of washing were determined, and the washing kineties and washing mechanism were explored to establish the theoretical basis and technical support for the actual remediation project of heavy metal contaminated soil in non-ferrous metal smelters.

Materials and methods

Soil samples collection and preservation

Soil samples were collected from the surface layer (0-100 cm) of a typical non-ferrous metal smelter in Yunnan, China (103.329, 26.455). Remove the impurities, such as stones and plant roots, and air-dry naturally in the absence of light. After crushing, they are fully mixed and evenly stored in plastic bags. Subsequently, soil samples are screened according to the specific requirements of the experiment. Some of the soil samples were screened with 100 meshes for the determination of soil physical and chemical properties and heavy metals. Another part of the soil samples was screened with 20 meshes for washing experiments. The basic characteristics and test methods of the soil sample are shown in *Table 1* and *Fig. 1*.

Parameter	Detection method ^a	Value
Soil type	/	Red soil
Soil size (%)	LY/T 1225-1999	/
Clay (<0.002mm)	/	3.57
Silt (0.002~0.05mm)	/	57.41
Sand (0.05~2mm)	/	38.02
pH	NY/T 1377-2007	6.70
Organic matter content $(g \cdot kg^{-1})$	LY/T 1237-1999	61.2
CEC^{b} (cmol·kg ⁻¹)	LY/T 1243-1999	13.5
$TN (g kg^{-1})$	LY/T 1228-2015	0.95
$TP(g \cdot kg^{-1})$	LY/T 1232-2015	2.69
Total Pb (mg·kg ⁻¹)	GB/T 17141-1997	7500.00

Table 1. Basic characteristics of contaminat	ed soil
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^a China standard detection method for soil, ^b Cation exchange capacity



Figure 1. Soil samples collection and preservation

Experimental equipment

Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES-5100, Agilent Technologies), Thermostatic oscillator (THZ-82, Guohua electric appliance Co., Ltd., China), Ultraviolet spectrophotometer (Cary 4000, Agilent Technologies), electronic balance (CP-214, Ohaus instrument), High speed tabletop refrigerated centrifuge (LXJ-II B, Shanghai anting scientific instrument, China), Microwave digestion instrument (Mars-6, CEM Corporation) were used in the experiment. All experimental water is deionized water.

Method of washing experiment

The washing experiment conditions of single eluents are shown in *Table 2* according to the principle of an orthogonal experiment (Cui et al., 2007; Zhao et al., 2013; Zuo et al., 2016).

Levels	HCl (M)	Oxalic acid (M)	EDTA (M)	Rhamnolipid (%)	Liquid-solid ratio	Washing time (min)
1	0.1	0.1	0.01	0.25	5:1	60
2	0.5	0.2	0.05	0.5	8:1	180
3	1.0	0.5	0.1	1.0	10:1	480
4	2.0	1.0	0.2	2.0	15:1	900

Table 2. Orthogonal experiment conditions for single eluents

A single factor experiment was used for compound washing, and the basic specific experimental methods were carried out in the following order: (1) Determination of the optimum compound ratio: HCl was compounded with EDTA and Rhamnolipid (2%), respectively. The compound ratios were set as 3:1, 2:1, 1:1, 1:2, and 1:3. The liquid-solid ratio was 8:1, and the washing time was 480 min; (2) Determination of optimal liquid-solid ratio: The compound ratio of HCl & EDTA was 1:1, the compound ratio of HCl & Rhamnolipid was 1:2, and the washing time was 480 min. The liquid-solid ratio gradients were set to 3:1 5:1, 8:1, 10:1, 15:1, and 20:1; (3) Determination of the optimum washing time: The compound ratio of HCl & EDTA was selected for 1: 1, and liquid-solid ratio for 10: 1. The compound ratio of HCl & Rhamnolipid as 1: 2, and liquid-solid ratio for 15: 1. The washing times for the two compound eluents were set at 60, 180, 300, 480, 720, 1080, 1440, and 2160 min.

Soil samples of 3.00 g were weighed and packed into 100 mL centrifugal tubes. The eluents were added according to the experimental methods to oscillate for a given time at 210 RPM at room temperature, centrifuged at 5000 RPM for 6 min, and the supernatant was filtered with a 0.45 μ m filter membrane. The contents of Pb were determined by ICP-OES.

All data were averaged by three groups of parallel experiments.

Determination method

In order to determine the distribution of heavy metal speciation in the soil before and after washing, the continuous extraction method of modified BCR was selected for the determination of Pb speciation in the soil (Tessier et al., 1979; Ashraf et al., 2012): (1) *Acid extractable*: add 40 mL of 0.1 M CH₃COOH to 1.00 g of soil samples, shaken at 210 RPM for 16 h at $(22\pm5)^{\circ}$ C, and centrifuged at 3000 RPM for 20 min; (2) *Reducible*: 40 mL of 0.5 M NH₂OH HCl was added to the residue, and shaken at $(22\pm5)^{\circ}$ C for 16 h, centrifuged at 3000 RPM for 20 min; (3) *Oxidizable*: 10 mL of H₂O₂ was added to the residue at room temperature for 1 h, and 10 mL of H₂O₂ was added again at $(85\pm2)^{\circ}$ C and maintained for 1 h, evaporated to near dryness, then 50 mL of 1 M CH₃COONH₄ was added, shaken at $(22\pm5)^{\circ}$ C for 16 h, and centrifuged at 3000 RPM for 20 min; (4) *Residual*: residual residue of 0.10 g was weighted and 10 mL HNO₃, 1 mL HClO₄ and 1 mL HF were added. The residue was microwaved until clear and transparent.

The content of various forms of Pb was determined by ICP-OES.

Methods of data processing and analysis

In this study, the ratio of heavy metal content in washing solution to original heavy metal content in contaminated soil was used to calculate the removal rate of Pb; and the results of washing were analyzed by Origin 2017.

Method of washing kinetics

In order to better understand the process of Pb washing in soil, three different models, Elovich equation, Two-constant equation, First-order equation, were selected to analyze and evaluate the washing kinetics data by least squares regression.

Elovich equation (Eq.1) contains a series of reactions including solute diffusion and surface activation in the liquid phase. Two-constant equation (Eq.2) is applicable to the dynamic system process with relatively complex reaction. First-order equation (Eq.3) reflects the relationship between reaction equilibrium concentration and reaction rate. Their expressions are as follows:

$$S = A + Blnt \tag{Eq.1}$$

$$lnS = A + Blnt \tag{Eq.2}$$

$$lnS = lnS_{max} + Bt \tag{Eq.3}$$

where, $S (\text{mg} \cdot \text{kg}^{-1})$ was the washing amount of Pb, $S_{max} (\text{mg} \cdot \text{kg}^{-1})$ was the total washing amount at equilibrium, A and B were both constants, and t (min) was the washing time. The determination coefficient (R^2) is used to judge the significance and merits of the model. The larger R^2 indicating that the higher the fitting degree, the better the model.

Results and discussion

Effect of single eluent washing conditions

The experiment results are shown in *Table 3~Table 6*.

Table 3 shows that the concentration of HCl is the key factor affecting the removal rate of Pb. The higher the concentration, the higher the removal efficiency of Pb in soil, with the highest removal rate of 39.22%. However, there was no significant change in the removal rate of Pb in soil when HCl exceeds 1.0 mol·L⁻¹, and increasing HCl concentration leads to the destruction of soil structure and physic-chemical properties, and increases the cost of remediating the contaminated soil (Moutsatsou et al., 2006). The washing process of HCl is mainly the result of the interaction of H⁺ promoting the dissolution of solid heavy metals and Cl⁻ complexation promoting the formation of metal ion complexes.

No.	HCl (M)	Liquid-solid ratio	Washing time (min)	Pb Removal rate (%)
1	0.1	5:1	60	5.22
2	0.1	8:1	180	7.94
3	0.1	10:1	480	10.40
4	0.1	15:1	900	12.43
5	0.5	5:1	480	16.94
6	0.5	8:1	900	18.71
7	0.5	10:1	60	23.35
8	0.5	15:1	180	29.34
9	1.0	5:1	900	12.03
10	1.0	8:1	60	16.96
11	1.0	10:1	180	27.53
12	1.0	15:1	480	31.20
13	2.0	5:1	180	19.43
14	2.0	8:1	480	21.74
15	2.0	10:1	900	24.73
16	2.0	15:1	60	39.22
R(Pb)	0.173	0.146	0.041	—

Table 3. Orthogonal experiment and results of HCl washing

Oxalic acid is a kind of natural organic acid eluent, which has the characteristics of extensive existence and easy biodegradation. It can be seen from *Table 4* that the oxalic acid has little effect on removing Pb from soil, only about 1%, regardless of the concentration of oxalic acid. The main reason is that oxalic acid is one of the dicarboxylic acids, and has few reaction sites, which provides limited acidity and concentration of oxalate ion, and provides fewer reaction sites. In addition, oxalate ion reacts with Pb released from soil to produce insoluble compound C₂O₄Pb, whose standard solubility product constant is 4.8×10^{-10} (Wei et al., 2016), which further inhibits the migration of metal ions to the aqueous phase, resulting in a poor washing effect.

It can be seen from *Table 5* that EDTA has a strong extraction efficiency for Pb in soil, and removal effect increases with the increase of EDTA concentration, up to 43.41%. This is due to its strong complexing ability and easy formation of 1:1 soluble complex with metals. The equilibrium constant of Pb-EDTA complex is 19.0 (Zhang et al., 2010; Qiao et al., 2017), which can further promote the desorption of Pb from the surface of soil

particles and inhibit the re-adsorption of desorbed heavy metals. However, EDTA can easily cause secondary pollution to soil due to its own difficulty in degradation, and the instability of its chelates with heavy metals may bring potential environmental risks (Wei et al., 2016). Therefore, the optimal elution concentration of EDTA is $0.2 \text{ mol} \cdot \text{L}^{-1}$.

No.	Oxalic Acid (M)	Liquid-solid ratio	Washing time (h)	Pb Removal rate (%)
1	0.1	5:1	60	0.10
2	0.1	8:1	180	0.10
3	0.1	10:1	480	0.13
4	0.1	15:1	900	0.19
5	0.2	5:1	480	0.07
6	0.2	8:1	900	0.11
7	0.2	10:1	60	0.58
8	0.2	15:1	180	0.41
9	0.5	5:1	900	0.02
10	0.5	8:1	60	0.46
11	0.5	10:1	180	0.61
12	0.5	15:1	480	0.92
13	1.0	5:1	180	0.42
14	1.0	8:1	480	0.65
15	1.0	10:1	900	1.11
16	1.0	15:1	60	1.34
R(Pb)	0.007	0.006	0.002	

Table 4. Orthogonal experiment and results of Oxalic acid washing

Table 5. Orthogonal experiment and results of EDTA washing

No.	EDTA (M)	Liquid-solid ratio	Washing time (h)	Pb Removal rate (%)
1	0.05	5:1	60	8.81
2	0.05	8:1	180	10.59
3	0.05	10:1	480	13.36
4	0.05	15:1	900	10.02
5	0.1	5:1	480	24.29
6	0.1	8:1	900	18.34
7	0.1	10:1	60	29.50
8	0.1	15:1	180	26.82
9	0.2	5:1	900	36.84
10	0.2	8:1	60	40.17
11	0.2	10:1	180	31.50
12	0.2	15:1	480	36.46
13	0.5	5:1	180	29.88
14	0.5	8:1	480	41.57
15	0.5	10:1	900	43.41
16	0.5	15:1	60	37.94
R(Pb)	0.275	0.045	0.044	_

Rhamnolipid, an environmentally friendly anionic biological surfactant, can complexes with heavy metal ions by its own special hydroxyl and carboxyl structure, and can also reduce the surface tension of eluent and weaken the adhesion of heavy metal ions to soil (Nitschke et al., 2010). However, as shown in *Table 6*, rhamnolipid washing alone is less effective in removing Pb from contaminated soil, with a maximum efficiency of less than 2.0%. The reason is that rhamnolipid is a pH neutral substance and can only remove water-soluble heavy metal ions, which is consistent with the research results of Zhou et al. (Wen et al., 2009; Zhou et al., 2017). Therefore, it must be combined with other acid eluents in order to wash heavy metals in soil more efficiently and economically.

No.	Rhamnolipid (%)	Liquid-solid ratio	Washing time (h)	Pb Removal rate (%)
1	0.5	5:1	60	0.01
2	0.5	8:1	180	0.06
3	0.5	10:1	480	0.14
4	0.5	15:1	900	0.03
5	1.0	5:1	480	0.01
6	1.0	8:1	900	0.22
7	1.0	10:1	60	0.33
8	1.0	15:1	180	0.36
9	2.0	5:1	900	0.15
10	2.0	8:1	60	0.31
11	2.0	10:1	180	0.67
12	2.0	15:1	480	0.73
13	5.0	5:1	180	0.38
14	5.0	8:1	480	0.77
15	5.0	10:1	900	1.08
16	5.0	15:1	60	1.33
R(Pb)	0.008	0.005	0.001	

Table 6. Orthogonal experiment and results of Rhamnolipid washing

In addition, by comparing the magnitude of the Range (R) of orthogonal experiments, the influence degree of washing conditions for single eluents on the removal rate of Pb can be shown as follows: *Eluent Concentration* > *Liquid-Solid Ratio* > *Washing Time*.

In summary, there are great differences in the removal efficiency of heavy metals in soil by four kinds of single eluents. Among them, HCl and EDTA had better removal efficiency of Pb in soil, optimum washing conditions of HCl and EDTA were 1 mol·L⁻¹, 0.2 mol·L⁻¹, respectively; the liquid-solid ratios were 15:1 and 8:1; the washing time of both was 480 min; Oxalic acid is not suitable for remediation of contaminated soil in this site because of its poor washing effect; rhamnolipid needs to be combined with acid eluents in order to fully play its role.

Effect of compound eluent washing conditions

Compound eluents are optimized by compounding different types of single eluents, which mainly utilize the synergistic effect of different agents to further improve the removal efficiency of pollutants, and can avoid the secondary pollution and high cost of the single eluent. HCl was compounded with EDTA and rhamnolipid according to the orthogonal experimental results of single eluent, and the elution conditions were optimized by single factor experiments of compound washing.

Effect of the compound ratio of eluent on washing efficiency

It can be seen from *Fig.* 2 that with the increase of the compound ratio of HCl & EDTA, the removal rate of Pb first increase and then decrease. The removal rates of Pb are the highest (up to 60.36%) when the ratio of HCl & EDTA is 1:1. However, for HCl & Rhamnolipid, the higher the compound ratio, the better the removal efficiency of heavy metals, but when the compound ratio exceeds 1:2, the removal rate does not change much. In addition, the removal rate of Pb by compound eluents were better than that by single eluents.



Figure 2. Effect of compound ratio on Pb removal rate

For HCl & EDTA combined elution, the elution mechanism is mainly as follows: (1) The acid solubility of H^+ and the complexation of Cl^- provided by HCl; (2) EDTA can not only reduce the surface tension of the eluent, but also form a stable complex Pb-EDTA with a coordination ratio of 1:1 with Pb; (3) the synergistic solubilization among different agents. However, for HCl & Rhamnolipid, besides (1) and (3), rhamnolipid is an anionic biosurfactant, which can not only reduce the surface tension of the eluent, but also form stable complexes with the special hydroxyl and carboxyl structures of the eluent and the Pb released from the soil.

Effect of liquid-solid ratio and washing time on washing efficiency

It can be concluded from *Fig.* 3(a) that with the increase of liquid-solid ratio, the removal rate of Pb in the soil of the two kinds of compound eluents are generally increasing; however, rate of removal increases slowly when it reaches a certain liquid-solid ratio. The essence of the increase of liquid-solid ratio is the increase of the amount of eluent, which leads to the increasing removal rate of heavy metals. The optimal liquid-solid ratio of both HCl & EDTA and HCl & Rhamnolipid was 10:1, according to the removal rate of Pb and the cost of remediation.

Washing time is a key factor that determines whether the eluent can interact adequately with the heavy metal pollutants in soil. Appropriate washing time can not only shorten the period of soil remediation, but also save costs. As can be seen from *Fig.* 3(b), the longer the washing time, the higher the removal efficiency of the two kinds of compound

eluents. However, when washing time exceeds a certain limit, the removal efficiency of heavy metals does not exhibit much change. Therefore, the washing time of HCl & EDTA was 720 min, while that of HCl & Rhamnolipid was 1080 min.



Figure 3. Effect of liquid-solid ratio and washing time on washing efficiency

Therefore, considering various factors of soil remediation, such as washing cost, secondary pollution, etc., HCl & Rhamnolipid compound washing effect is better, the optimum mixed ratio is 1:1, the liquid-solid ratio is 15:1, the washing time is 1080 min, and the removal rate of Pb in soil is 68.35%, which can be repaired to meet "risk intervention values (Pb content < 2500mg·kg⁻¹) for soil contamination of the development land" in soil environmental quality standard China (GB36600-2018).

Kinetics of compound washing

In order to better understand the process of Pb washing in soil, three different models, Elovich equation, Two-constant equation, First-order equation, were selected to analyze and evaluate the washing kinetics data by least squares regression, and the fitting results are shown in *Table 7*.

Equation	Elovich equation S = A + Blnt	Elovich equationTwo-constant equation $S = A + Blnt$ $lnS = A + Blnt$	
Fitting result	y = 10.391x - 6.3177	y = 0.2009x + 2.7916	y = 0.0003x + 3.8352
R ²	$R^2 = 0.9339$	$R^2 = 0.9199$	$R^2 = 0.5468$

Table 7. Fitting results of three different models

By comparing R^2 of the three models, it can be seen that Elovich equation and Two-constant equation have a high fitting degree for the washing kinetics of heavy metal Pb in contaminated soil, while First-order equation is not suitable for describing the washing kinetics. Therefore, the washing desorption process of Pb in soil by HCl & Rhamnolipid is controlled by diffusion factors, and it is mainly a heterogeneous diffusion process.

Speciation distribution of Pb before and after washing

The risks of heavy metal pollution to soil is directly related to their total concentration as well as their speciation distribution. In this study, we further compare the speciation changes of Pb in the soil before and after compound washing. The results are shown in *Table 8*.

	Before washing		HCl & EDTA		HCl & Rhamnolipid	
Pb speciation	Content	Percentage	Content	Percentage	Content	Percentage
	(mg·kg ⁻¹)	(%)	(mg·kg ⁻¹)	(%)	(mg·kg ⁻¹)	(%)
Acid extractable	1426.50	19.02	350.35	12.47	240.70	10.14
Reducible	2058.00	27.44	233.75	8.32	141.24	5.95
Oxidizable	1192.50	15.90	272.52	9.70	190.14	8.01
Residual	2823.00	37.64	1952.88	69.51	1801.67	75.9
Total	7500.00	100.00	2809.50	100.00	2373.75	100

Table 8. Speciation distribution of Pb in soil before and after washing

Table 8 shows that Pb in contaminated soils of the smelter exists mainly in the form of Residual and Reducible, accounting for 37.64% and 27.44%, respectively, indicating that Pb forms a stable crystal structure with soil or firmly binds with clay minerals. After compound washing, the speciation distribution of Pb in soil changed greatly, and the concentration of all forms decreased significantly, indicating that the compound eluents of HCl & EDTA and HCl & Rhamnolipid could release Pb in various forms.

The Acid extractable is the most susceptible metal form to washing, and it is very sensitive to pH changes, and can be easily eluted when the pH is reduced. Therefore, the addition of HCl among the compound eluent greatly reduced the content of acid extractable Pb in the soil. Reducible mainly refers to heavy metals bound by Fe and Mn oxides in soil (Shen et al., 2007). The content of reducible Pb in soil also decreased significantly, which is mainly due to the high content of Fe and Mn in the soil polluted by this smelting site. A large amount of Fe and Mn are extracted at the same time in the process of composite washing, which leads to the decrease of Pb content in the form of Fe and Mn oxides in soil. The Oxidizable mainly refers to heavy metals bound by organic matter and sulfide in soil. The higher the content of organic matter in soil, the better the extraction of Oxidizable (i.e. organic matter bound) (Li et al., 1996). The content of organic matter in the soil used in this study is relatively high, so the content of Oxidizable Pb has also decreased significantly after complex washing. The properties of Residual are usually stable, and they usually exist in silicate, primary minerals and secondary minerals (Barona et al., 2001). The residual Pb in soil also decreases to some extent after complex washing, which may be due to the release of some residual Pb accompanied by silicate dispersion.

Conclusion

In this study, single and compound eluents were applied to washing remediate the soils with Pb contamination intensity of 7500 mg·kg⁻¹ in an abandoned non-ferrous metal smelter in Yunnan China. The results showed that when the compound ratio of HCl $(1 \text{ mol} \cdot \text{L}^{-1})$ & Rhamnolipid (2%) was 1:1, the liquid-solid ratio was 10:1, and the washing

time was 1080 min, the removal rate of Pb in the soil was 68.35%, which met "*Risk* screening value for soil contamination of the development land" (Pb < 2500 mg·kg⁻¹) in "soil environmental quality standard China (GB36600-2018)". The washing desorption process of Pb in soil conforms to Elovich model and is mainly a heterogeneous diffusion process. In addition, from the distribution of Pb forms in soil, the main forms of Pb in the waste smelter soil are acid extractable and reducible. The active Pb was effectively removed, which greatly reduced the biological toxicity and environmental risk of the soil.

The heavy metals in the contaminated soil are eventually transferred to the washing waste liquid, so the recovery and treatment of the washing waste liquid should be further studied in the future. In addition, this study mainly simulated the ectopic washing process of contaminated soil, and the amount of soil used in the experiment was less. Therefore, the soil column washing experiment could be conducted by simulating the in-situ washing process in the future.

Acknowledgements. This work was financially supported by the National Water Pollution Control and Treatment Science and Technology Major Project (2015ZX07406005), and Preparation Project of Education Material for Soil Environmental Protection Publicity of Ministry of Ecology and Environment of People's Republic of China (H201606).

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