



KH₂PO₄-aided soil washing for removing arsenic from water-stable soil aggregates collected in southern China

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ABSTRACT

Removal of arsenic (As) from soil aggregates with particle sizes of > 2.0, 2.0-0.25, 0.25-0.053, < 0.053 mm by soil washing of KH₂PO₄ and the kinetics of As releasing from soil aggregates were investigated. Effects of KH₂PO₄ concentration, ratio of liquid/soil and washing duration on the removal were fully explored. The results showed that the high As removal was obtained in > 2 mm aggregates (48.56%) and < 0.053 mm aggregates (42.88%) under the optimum condition (KH₂PO₄ concentration of 0.1 mol/L, and liquid/soil ratio (10 mL/g) for 360 min). 62.82% of As was extracted from aggregates with size less than 0.25 mm. Only 11.88% was contributed by the large aggregates (> 2.0mm). Using KH₂PO₄ washing, it was also found that extracted As is mainly in form of either specifically sorbed As or As associated with oxides of Fe and Al. Elovich model can describe the removal process of As more precisely than Two-constant kinetic models. The optimum washing conditions and removal process is also applied to bulk soil. This technique in this study is reliable, cost-effective and offers a great potential for practical application in soil remediation.

Keywords: Arsenic (As), Potassium dihydrogen phosphate (KH₂PO₄), Soil aggregates, Soil washing

1. Introduction

The arsenic (As) contamination of soil has become an important environmental issue throughout the world [1]. Contaminated soil by As can result in massive and persistent environmental damage, giving risk to human health [2, 3]. As such, research on the remediation of the As contaminated soil has become a hot research topic. Many techniques such as phytoextraction [4], biosurfactant [5, 6], chemical immobilization or stabilization [7, 8], and soil washing [9, 10], had been used to remediate the As-contaminated soil. It has been widely recognized that soil washing is one of the most promising techniques for removing the heavy metals from the contaminated soil. Moreover, this method is still applicable when there is multiple As concentrations from region to region [11].

Many studies had been conducted in recent years on the remediation of As contaminated soil by soil washing technique. Previous studies mainly focused on the feasibility of different kinds of washing solutions, the As removal efficiency from contaminated soil, and the changes on soil toxicity after washing [12-14]. Phosphate (PO₄³⁻) has been used for As removal from soil due to its capability to replace arsenic or arsenate (AsO₄³⁻). Alam et

al. [15] found that up to 40% of arsenic can be extracted from a forest soil by 0.9 mol/L of KH₂PO₄ and less than 0.4% of arsenic was extracted by other potassium-containing solutions such as KNO₃, K₂SO₄ and KCl. The As removal by 0.5 mol/L of KH₂PO₄ (74.03%) was higher than that by 0.5 mol/L citric acid (53.87%) or 0.1 mol/L EDTA (62.98%) [16]. The ability of As removal between 0.2 mol/L H₃PO₄ and KH₂PO₄ was similar from soil [13]. Importantly, KH₂PO₄ has limited interaction with soil components such as Ca, Mg and Si. Therefore, KH₂PO₄ is selected as an extraction reagent in present study due to its efficiency and environmentally friendly.

It is known that chemical forms of As could not only affect its removal efficiency, but also reflected the ecological risks of soils after the treatment [12, 17]. The different chemical forms of As can result in different ecological risks. The different As chemical forms targeted by phosphate had been reported. Jho et al. [13] found that phosphate-aided soil washing can only remove the non-specifically sorbed and specifically sorbed As. While Alam et al. [15] reported that phosphate solution could extract the As bounded with Fe and Al. However, Chen et al. [16] found that phosphate-aided soil washing could reduce all the chemical forms of As.

Soil aggregates were the basic unit of soil structure [18]. Heavy



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metals can be adsorbed and remained in soil aggregates in different ways [19]. As such, soil washing efficiency can be greatly affected by the particle size of the soil aggregates. The larger the particle size of soil aggregates were, the more the removal of heavy metals was during soil washing [9, 20-22]. However, the investigation of desorption of As from soil aggregates of contaminated soil by soil washing is still in its infancy. The changes of As forms in soil aggregates after soil washing should be paid attention to and the resultant risks should be carefully assessed.

Therefore, in this study, soil aggregates with different particle sizes and bulk soil were used to investigate the As removal by KH_2PO_4 -aided soil washing method. The influence of the washing conditions (KH_2PO_4 concentration, liquid/soil ratio and washing duration) on the removal of As from soil aggregates and bulk soil were comprehensively studied. The kinetic process of As removal were analyzed and the changes of As chemical forms were also compared among soil aggregates. The pH value of all the KH_2PO_4 solutions used in present study was adjusted the same as that of the bulk soil (4.60) to minimize the damage of leaching on soil properties. The results would be helpful to accumulate the data for the As removal from soil aggregates and to supply useful references for developing the remediation technique which was cost-effective and applied easily.

2. Materials and Methods

2.1. Soil Sample Preparation

Surface soil samples (0-20 cm) were collected from a farmland close to a smelter in Hunan Province, southern China. Soil samples were maintained their structure and transported back to the laboratory within 2 d of collection. Part of the samples was air-dried at room temperature and was sieved through a 2-mm polyethylene sieve as the bulk soil (S0). The rest fresh samples were kept in the refrigerator at 0-4°C before separation into soil aggregates with different sizes.

2.2. Separation of Soil Aggregates

Fraction of soil aggregates was obtained by wet-sieving procedure [23]. In specific, the fresh soil was immersed in water on a set of three nested sieves (2.0, 0.25, and 0.053 mm) and shaken vertically 3 cm for 50 times during a period of 2 min. The aggregates retained in each sieve, large-aggregates (> 2.0 mm) (S1), macro-aggregates (2.0-0.25 mm) (S2), micro-aggregates (0.25-0.053 mm) (S3) and silt + clay fractions (< 0.053 mm) (S4) were collected after centrifugation.

2.3. Soil Washing Treatment Procedure

2.3.1. Effect of KH_2PO_4 concentration

1.0 g of soil was carefully added in a 50 mL centrifugal tube. 20 mL of KH_2PO_4 solution with different concentrations (0, 0.01, 0.05, 0.1, 0.5 and 1 mol/L) was slowly added to achieve a liquid/soil ratio of 20, and then the tubes were shaken at $25 \pm 1^\circ\text{C}$ for 24 h. The suspension was centrifuged for 10 min at 4000 rpm. The supernatant was collected through a $0.45 \mu\text{m}$ membrane. The filtrate was analyzed for As concentration.

2.3.2. Effect of liquid/soil ratio

The liquid/soil ratios of 5, 10, 15, 20 and 30 were obtained by adding 5, 10, 15, 20 or 30 mL of 0.1 mol/L KH_2PO_4 solution to a 50 mL centrifugal tube with 1.0 g of soil or soil aggregates. Obtained solutions were shaken at $25 \pm 1^\circ\text{C}$ for 24 h and then centrifuged for 10 min at 4000 rpm. The supernatant was filtered through a $0.45 \mu\text{m}$ membrane. The filtrate was analyzed for As concentration.

2.3.3. Effect of washing time

The tubes added with 1.0 g of bulk soil or soil aggregates and then 10 mL of 0.1 mol/L KH_2PO_4 solution were shaken at $25 \pm 1^\circ\text{C}$. The samples were collected at 20, 40, 60, 120, 240, 360, 720, 1440 min, and then centrifuged at 4000 rpm for 10 min. The supernatant was filtered through a $0.45 \mu\text{m}$ membrane. The filtrate was analyzed for As concentration.

2.4. As Removal from Soil Aggregates under the Optimum Washing Conditions

Soil slurries were prepared under the optimum washing conditions (washing period, concentration of KH_2PO_4 and liquid/soil ratio) based on the results from 2.1-2.3. The slurries were shaken at $25 \pm 1^\circ\text{C}$ for optimum washing period and then centrifuged at 4000 rpm for 10 min. The supernatant was filtered through a $0.45 \mu\text{m}$ membrane. The filtrate was analyzed for As concentration.

2.5. As Fractionations in Soil Aggregates after Soil Washing

10 mL KH_2PO_4 (in 0.1 mol/L) solution was added into 50 mL centrifugal tubes contained 1.0 g soil or soil aggregate sample. The suspension was shaken for 360 min at $25 \pm 1^\circ\text{C}$ and then centrifuged at 4000 r/min for 10 min. The supernatant was discarded and the precipitates were rinsed twice using deionized water and then dried for As extraction.

2.6. Chemical Analysis

The pH of soil and aggregates was measured using a pH electrode (pHs-3C, Shanghai Jing Ke Test Equipment Institute, China) (soil:water = 1:2.5). The cation exchange capacity (CEC) was determined in BaCl_2 [24], organic matter (OM) content was determined by a volumetric method of potassium dichromate heating. Free iron oxide was measured by the method described by Lu et al. [25]. Total phosphorus (TP) of soil samples was measured according to the method introduced by Bao et al. [26].

The As fractions in soil aggregates and bulk soil before and after washing were extracted following the method described by Wenzel et al. [27]. As in soils were divided into five fractions (1) non-specifically sorbed As (F1); (2) specifically sorbed As (F2); (3) As associated with amorphous hydrous oxides of Fe and Al (F3); (4) As associated with crystalline hydrous oxides of Fe and Al (F4) and (5) residual As fraction (F5).

Total arsenic content was detected using the atomic fluorescence spectrometry (AFS-9700A, Haiguang Instrumental Corp., China) after aqua regia extraction (0.2 g soil aggregates and 10 mL (1 + 1) aqua regia digested for 2 h in boiling water) in which the conditions were determined according to the Soil quality-Analysis of total Mercury, arsenic and lead contents - Atomic Fluorescence Spectrometry - Part 2: Analysis of total arsenic contents in soils

constituted by the State Environmental Protection Administration of China(GB-T 22105.3-2008) All above treatments had triplicates.

2.7. Data Analysis

The experimental results were subjected to a one-way analysis of variance and least significance difference using SPSS 16.0 statistical software at 95% confidence, SigmaPlot 10.0 was used to generate the graphs.

3. Results and Discussion

3.1. Effect of KH_2PO_4 Concentration on As Removal

The optimum KH_2PO_4 concentration was 0.1 mol/L for both soil aggregates and bulk soil (Fig. 1). It was shown that As removal from all the aggregates increased dramatically with KH_2PO_4 concentration increasing from 0 to 0.1 mol/L. The As removal increased at a slow pace when the KH_2PO_4 concentration was higher than 0.1 mol/L. When the KH_2PO_4 solution was 0.1 mol/L, As removals were 50.1%, 38.2%, 45.1% and 47.0% for S1, S2, S3 and S4, respectively. For comparison, the As removal was only 11.4%, 4.5%, 11.3%, 11.5% for S1, S2, S3 and S4 when no KH_2PO_4 was added in the recipe. Using 0.5 mol/L KH_2PO_4 only increased As removal by another 5-8% and about 74.07-87.72% of the As removed using 1 mol/L KH_2PO_4 could be removed using 0.1 mol/L KH_2PO_4 in different size aggregates. The similar trend of the As removal from bulk soil was found. The As removal increased rapidly when the concentration of KH_2PO_4 was lower than 0.1 mol/L and no significant increase was observed after that. These results are consistent with the one reported by Zou *et al.* [28] in which of As removal is more efficient for a higher concentration of Na2EDTA. However, the linear relationship between As removal and $\text{NH}_4\text{H}_2\text{PO}_4$ had been reported too [12].

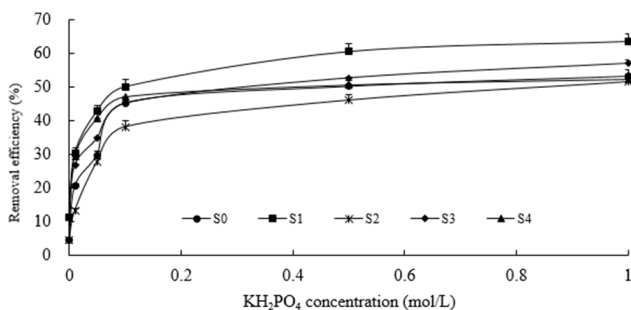


Fig. 1. Effect of KH_2PO_4 concentration on As removal by soil washing treatment. S0: bulk soil, S1: large-aggregates (> 2.0 mm), S2: macro-aggregates (2.0-0.25 mm), S3: micro-aggregates (0.25-0.053 mm), S4: silt + clay fractions (< 0.053 mm).

3.2. Effect of Liquid/soil Ratio on As Removal

The liquid/soil ratio could significantly affect the efficiency of heavy metals removal from soils [28]. Based on the experiment, the optimum liquid/soil ratio should be 10 mL/g for bulk soil, S1, S3 and S4, and 15 mL/g for S2 (Fig. 2). The As removal from soil aggregates, when the liquid/soil ratios were 10, 15 and 20

mL/g, was significantly higher than that when the liquid/soil ratios were 5, 25 and 30 mL/g ($p < 0.05$). The As removal from S2 in the treatment of 15 mL/g was higher than that of 10 and 20 mL/g. However, the As removal from S3 and S4 did not vary significantly among these three concentrations. The As removal from S1 decreased when the liquid/soil ratio increased from 10 to 20 mL/g. Since the higher the liquid/soil ratio was in soil washing, the more leachate needed to be dealt with [29], the optimum liquid/soil ratio was selected to be 10 mL/g in present study. It was also found that the As removal for S2, S3 and S4 decreased with the increase of liquid/soil ratio when the liquid/soil ratio was higher than 20 mL/g. The reason might be that acid-based KH_2PO_4 washing solutions dissolved soil fractions including metallic fractions (e.g., hydrous oxides of Fe and Al). Thus, more soil fractions for re-adsorption of the extracted As may be available [16]. In addition, high liquid/soil ratio increased the contact area between the extracted As and soil, which led to the re-adsorption of As [30]. The highest As removal from bulk soil was also obtained when the liquid/soil ratios were 10, 15 and 20 mL/g. These results were different with those from the study of Yin *et al.* [31] in which lower liquid/soil ratio removed more Cu, Zn, Ni from soils by water was found. The difference might be from that the heavy metals dissolved in water could not re-adsorbed on the surfaces of soil solids during soil washing under conditions of acidic and circum-neutral [30].

The As removal was different among soil aggregates with the changes of L/S ratios. This was similar with the study of Lee and Oa [21]. The different compositions and properties among soil aggregates might be related to these kinds of results. Lee and Oa [21] had reported that in the samples collected from the slope area, the Pb removal was different from each other with the change of L/S ratios. The optimum L/S ratios were 2 mL/g (> 2.8 mm), 3 mL/g (2.8-0.075 mm), and 4 mL/g (< 0.075 mm). Compared to those from slope area, Pb removal efficiency from > 2.8 mm soil aggregates collected from the land area was not affected by the L/S ratios. The optimum L/S ratios for 2.8-0.075 and < 0.075 mm soil aggregates were 4 mL/g.

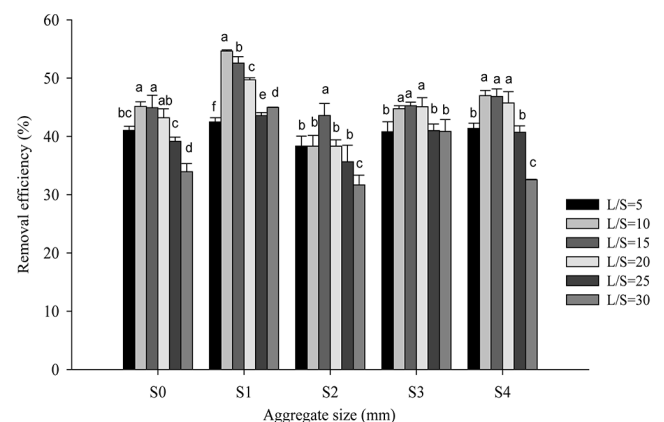


Fig. 2. Effect of liquid/soil ratio on As removal by soil washing treatment. Different small letters in the same aggregate indicated significant difference among the different treatments at 0.05 level. S0: bulk soil, S1: large-aggregates (> 2.0 mm), S2: macro-aggregates (2.0-0.25 mm), S3: micro-aggregates (0.25-0.053 mm), S4: silt + clay fractions (< 0.053 mm).

3.3. Effect of Washing Duration on As Removal

The effect of washing duration on As removal from soil aggregates was the same as bulk soil (Fig. 3). The removal of As increased significantly with the increase of washing time from 0 to 360 min ($p < 0.05$), after which only 3.7-9.1% of the As was removed. A longer washing period meant a higher operating cost [32]. Hence, 360 min washing could be chosen as the optimum washing duration to remediate As-contaminated soil. This result was similar with that of Zeng et al. [13].

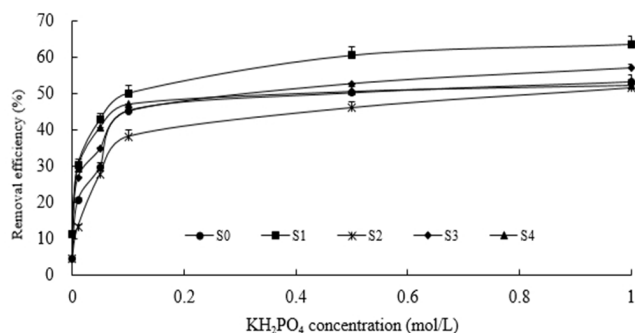


Fig. 3. Kinetics of As removed from soil aggregates. S0: bulk soil, S1: large-aggregates (> 2.0 mm), S2: macro-aggregates (2.0-0.25 mm), S3: micro-aggregates (0.25-0.053 mm), S4: silt + clay fractions (< 0.053 mm).

In the present study, the As removal among the soil aggregates was similar with the washing time extended. This was different from some previous studies. Liao et al. [22] had found that the As removal in > 2 mm aggregates slowly increased with washing duration, which decreased in those of < 0.1 mm aggregates when using the mixed solution (a mixture of 1.0% rhamnolipids in 2.0 mol/L HCl) for soil washing. Meanwhile, the Pb removal from 2.8-0.075 and < 0.075 mm aggregates were almost not affected by the washing duration was also reported [21]. The different results were probably attributed to the difference of extraction parameters, target heavy metals, the combined forms of heavy metals and properties of studied soils which were highly related to the release of heavy metals.

The kinetic data of As removed from soil aggregates were analyzed by kinetic models. Desorption kinetic analysis was conducted by using Elovich model and two-constant model, which were general equations used to describe the characteristics of the desorption mechanism in soil [33]. Elovich model (Eq. (1)) and two-constant model (Eq. (2)) equations were as follows:

$$q_t = 1/\beta_s \ln \alpha_s \beta_s + 1/\beta_s \ln t \quad (1)$$

$$q_t = a t^b \quad (2)$$

Where q_t was the As removal content at time t (in mg/kg); t was the washing time (min), α_s and a were initial As removal rate (mg/kg/min), β_s and b were As removal constant [$1/(\text{mg/kg})$].

The parameters of kinetics equations were presented in Table 1. The result indicated that the mechanism of As removal from soil aggregates was the same as bulk soil. Compared to those of two-constant model, coefficients of determination (R^2) obtained by Elovich model (0.86-0.96) were high for all the soil aggregates and bulk soil. Thus, Elovich model was better to describe the process of As removal. This was similar with the study of Zeng et al. [13]. The removal of As from soil aggregates and bulk soil might be the reactions composed of a series of complex processes [34].

3.4. As Removal from Soil Aggregates under Optimum Conditions

Fig.4 compared the As removal from different soil aggregates at the optimum conditions (0.1 mol/L of KH_2PO_4 , liquid/soil ratio 10 mL/g and washing for 360 min). The highest As removal was obtained in S1 (48.56%) resulting from the initial low contamination, which was followed by S4 (42.88%). The As removal from S2 was difficult (31.38%). The result was similar with that of Liao et al. [22].

In present study, the high As removal not only came from S1 (> 2 mm) but also S4 (< 0.053 mm). The content of Fe oxides and organic matter was low in coarse aggregates, thus heavy metals

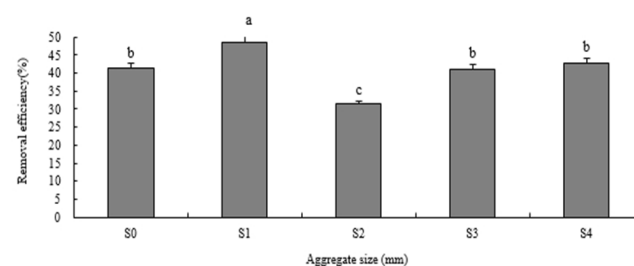


Fig. 4. As removal from soil aggregates with 0.1 mol/L KH_2PO_4 . Different small letters indicated significant difference among the different soil aggregates at 0.05 level. S0: bulk soil, S1: large-aggregates (> 2.0 mm), S2: macro-aggregates (2.0-0.25 mm), S3: micro-aggregates (0.25-0.053 mm), S4: silt + clay fractions (< 0.053 mm).

Table 1. Parameters of Removal Kinetics Equations of As from Soil Aggregates and Bulk Soil

Kinetic models	Parameter	S0	Aggregate size/mm				
			S1	S2	S3	S4	
Elovich	R^2	0.90	0.91	0.96	0.92	0.86	
	α_s /(mg/kg/min)	2.50	4.02	2.00	3.06	6.17	
	β_s /(L/mg/kg)	0.08	0.06	0.11	0.07	0.05	
two-constant	R^2	0.80	0.81	0.89	0.83	0.77	
	a /(mg/kg/min)	11.38	17.67	8.28	13.28	24.78	
	b /(L/mg/kg)	0.25	0.24	0.25	0.25	0.22	

R^2 : coefficients of determination. S0: bulk soil, S1: large-aggregates (> 2.0 mm), S2: macro-aggregates (2.0-0.25 mm), S3: micro-aggregates (0.25-0.053 mm), S4: silt + clay fractions (< 0.053 mm).

Table 2. The Properties of the Soil and Soil Aggregates

Size (mm)	pH	OM (g/kg)	TP/ (g/kg)	Free iron oxide (g/kg)	CEC (c mol/kg)	As (mg/kg)	Mass (%)
S0	4.60	29.15 ± 1.40c	0.87 ± 0.05b	1.96 ± 0.08b	5.70 ± 0.03c	142.25 ± 6.01d	-
S1	4.40	34.98 ± 1.06b	0.86 ± 0.01b	1.76 ± 0.02c	4.86 ± 0.03d	178.71 ± 4.02b	8.70
S2	4.55	21.48 ± 1.00e	0.83 ± 0.02b	1.97 ± 0.02b	7.89 ± 0.03b	126.03 ± 3.80e	40.63
S3	4.58	26.64 ± 0.20d	1.28 ± 0.04a	1.82 ± 0.03c	5.09 ± 0.01e	165.47 ± 4.56c	33.03
S4	4.65	63.53 ± 1.61a	1.22 ± 0.05a	3.06 ± 0.06a	9.68 ± 0.03a	230.35 ± 6.95a	17.64

Different small letters in the same column indicated significant difference among the different soil aggregates ($p < 0.05$); S0: bulk soil, S1: large-aggregates (> 2.0 mm), S2: macro-aggregates (2.0-0.25 mm), S3: micro-aggregates (0.25-0.053 mm), S4: silt + clay fractions (< 0.053 mm).

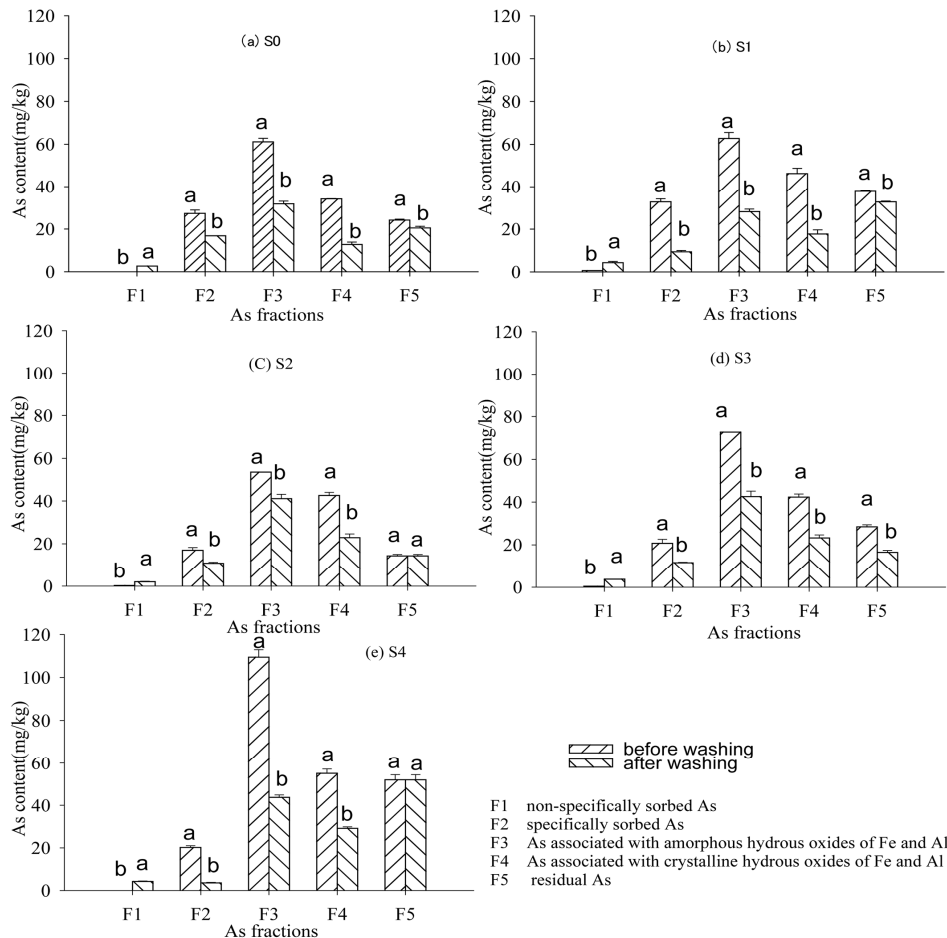


Fig. 5. Distribution of As fractions in soil aggregates before and after washing with KH_2PO_4 . Different small letters in the same aggregate indicated significant difference among the different treatments at 0.05 level. KH_2PO_4 concentration = 0.1 mol/L, washing time = 360 min, temperature = $25 \pm 1^\circ\text{C}$. S0: bulk soil, S1: large-aggregates (> 2.0 mm), S2: macro-aggregates (2.0-0.25 mm), S3: micro-aggregates (0.25-0.053 mm), S4: silt + clay fractions (< 0.053 mm).

were easier to be released (Table 2). Therefore, the coarse aggregates usually obtained high removal efficiency [22, 35]. Meanwhile, some studies showed that the fine aggregates also could achieved high heavy metals removal. Hansen *et al.* [20] found that copper removal was nearly 90% in the fine particles and less than 40% in coarse particles by 1 mol/L acid washing. Heavy metal removal from soil aggregates was affected not only by leaching parameters but also the combined form of heavy metals and physicochemical properties [22]. The different removal of As among soil aggregates

could be mainly attributed to the arsenic speciation, organic matter contents and initial As contents in soil aggregates, which were highly responsible for As desorption from soils. The more the As associated with oxides of Fe and Al was, the more the As removal was. Alam *et al.* [16] had reported that the As extracted from soil by KH_2PO_4 washing was all from the As associated with Fe and Al. In present study, 71.8%-88.1% of the As removed came from the As associated with oxides of Fe and Al among soil aggregates and bulk soil (Fig. 5). The removed As amounts was

in accordance with the amounts of As associated with oxides of Fe and Al in soil aggregates before soil washing. Therefore, arsenic speciation might affect the As removal in soil aggregates, which was similar to the result of Lee and Kao [36]. The higher the organic matter contents were, the more the As removal in soil aggregates was. Soil organic acids such as humic acids (HA), the most abundant fraction of decomposed OM, may compete with phosphate for sorption on soil constituents [37]. In turn, competition with arsenate affected As release from soils [38]. In present study, the organic matter contents in S4, S1 and S3 were 3.0, 1.6 and 1.2 times higher than S2, respectively (Table 2). Thus, the As removal from S4, S1 and S3 was also 2.5, 2.1 and 1.7 times higher than that from S2. Besides the organic matter, the high initial concentration in soil aggregates was also a factor that was likely to result in the greater As removal. Xu et al. [33] had found that the removal of heavy metals from medium pollution soil load (Pb, 230.94 mg/kg; Zn, 232.69 mg/kg) was 6.45%-12.35% higher than that in the slight pollution load soil (Pb, 89.77 mg/kg; Zn, 113.54 mg/kg). In present study, the As concentrations (Table 2) in S4, S1 and S3 were 82.8%, 41.8% and 31.3% higher than that in S2 and the removal efficiency were 136.7%, 154.8% and 131.2% higher than S2, respectively. The combined effects of these characteristics of soil aggregates influenced the As desorption and might result in different removal of As from soil aggregates.

According to the proportion of different particle size aggregates in bulk soil, the amounts of As removed from bulk soil and the contribution of each particle size aggregate to total removed As could be calculated. The results indicated that calculated amount of As removed from bulk soil (63.4 mg/kg) was the same as the results from the experiment (62.3 mg/kg). The results also indicated that the As removed mainly came from the < 0.25 mm aggregates (S3 and S4), which contributed 62.8% for the total As removed from the soil, among which S3 contributed 35.4%. Meanwhile, S1 contributed the least (11.88%) for the total As removed.

4. Conclusions

Laboratory batch experiments were conducted to study As removal with KH_2PO_4 from soil aggregates and bulk soil. The effect of KH_2PO_4 concentration, liquid/soil ratio and washing duration on As removal from soil aggregates and bulk soil were discussed. The results indicated that the As removal increased with KH_2PO_4 concentration and washing duration increasing. The As removal from soil aggregates decreased with the following sequence: S1 > S4 > S3 > S2 under the optimum condition (KH_2PO_4 concentration (0.1 mol/L), liquid/soil ratio (10 mL/g) and washing period (360 min)). The removal of As in soil was mainly from S3 and S4, which accounted for 62.82% of the total removed As, while S1 contributed the least (11.88%) for the total As removed. Soil washing reduced the specifically sorbed As and As associated with oxides of Fe and Al. Compared to the two-constant kinetic models, Elovich model was better to describe the removal process of As from soil aggregates. The similar optimum washing conditions and removal process were found in bulk soil.

Acknowledgments

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