



Effect of different soil washing solutions on bioavailability of residual arsenic in soils and soil properties



Jinwoo Im^a, Kyung Yang^a, Eun Hea Jho^{b,*}, Kyoungphile Nam^a

^a Department of Civil and Environmental Engineering, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 151-742, Republic of Korea

^b Department of Environmental Science, Hankuk University of Foreign Studies, 81 Oedae-ro, Mohyeon-myeon, Cheoin-gu, Yongin-si, Gyeonggi-do 449-791, Republic of Korea

HIGHLIGHTS

- Acidic and near-neutral washing solutions reduced the soil As level by 32–62%.
- The $(\text{NH}_4)_2\text{SO}_4^-$, $\text{NH}_4\text{H}_2\text{PO}_4^-$, $(\text{NH}_4)_2\text{C}_2\text{O}_4^-$ -extractable As were greatly reduced.
- But seed germination, growth, and enzyme activities were lower in the washed soils.
- Greater toxic effects are largely due to changes in soil properties after washing.
- Proper post-treatment techniques need to be selected for reuses of the washed soils.

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ABSTRACT

The effect of soil washing used for arsenic (As)-contaminated soil remediation on soil properties and bioavailability of residual As in soil is receiving increasing attention due to increasing interest in conserving soil qualities after remediation. This study investigates the effect of different washing solutions on bioavailability of residual As in soils and soil properties after soil washing. Regardless of washing solutions, the sequential extraction revealed that the residual As concentrations and the amount of readily labile As in soils were reduced after soil washing. However, the bioassay tests showed that the washed soils exhibited ecotoxicological effects – lower seed germination, shoot growth, and enzyme activities – and this could largely be attributed to the acidic pH and/or excessive nutrient contents of the washed soils depending on washing solutions. Overall, this study showed that treated soils having lower levels of contaminants could still exhibit toxic effects due to changes in soil properties, which highly depended on washing solutions. This study also emphasizes that data on the As concentrations, the soil properties, and the ecotoxicological effects are necessary to properly manage the washed soils for reuses. The results of this study can, thus, be utilized to select proper post-treatment techniques for the washed soils.

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1. Introduction

Various remedial techniques such as stabilization/solidification (Miretzky and Cirelli, 2010), soil washing (Tsang and Hartley, 2014), phytoremediation, specifically, phytoextraction using hyperaccumulator species, (Mandal et al., 2014), and electrokinetic remediation (Kim et al., 2013) can be applied for remediation of arsenic (As)-contaminated soils. In Korea, however, soil washing is often considered as a remedial technique for As-contaminated sites (e.g., a former Janghang smelting site, Dalchen mine), as it removes As from soils in a relatively short time compared to other

remedial techniques rather than just reducing bioavailability of As in soils via immobilization (Han and Shin, 2008; Moon et al., 2011). Since the treated soils are largely reused at the site and the current Korean regulation evaluates remedial efficiency based on the total As concentrations in soils, As removal techniques (e.g., soil washing) may be preferred over As immobilization techniques.

During soil washing, contaminated soils are mixed with a washing solution, and agitated to remove contaminants. The washing treatment process or the washing solution used in washing treatment is likely to affect soil properties that are related to soil qualities (Ko et al., 2005; Yi et al., 2012). For example, soil properties such as soil texture, water holding capacity, organic matter (OM) content, and total nitrogen concentration were changed after soil washing (Yi et al., 2012). Such changes in soil properties may change soil permeability and shear strength, which will

* Corresponding author.

E-mail addresses: dlawlsdn@snu.ac.kr (J. Im), deliyg22@snu.ac.kr (K. Yang), ehjho@hufs.ac.kr (E.H. Jho), kpnam@snu.ac.kr (K. Nam).

consequently affect long-term soil functions (Zihms et al., 2013). Changes in soil properties could also mean possible changes in bioavailability of As in soils as bioavailability of metals or metalloids is highly dependent on soil properties (e.g., pH, OM content, redox potential) (Masscheleyn et al., 1991; Huang et al., 2006).

Soil washing of contaminated soils usually involves strong acids as washing solutions. For example, a number of patented washing solutions targeting As removal from soils in Korea involve HCl or H_3PO_4 – 1 M hydrochloric acid (HCl) (Choi et al., 2007), 0.5 M phosphoric acid (H_3PO_4) (Cheong and Kang, 2012), and 2% sodium (Na) dithionite in 0.01 M HCl (Kim et al., 2012). When strong acid-based washing solutions are used for As removal from soils, acids containing oxyanions (e.g., H_3PO_4 or H_2SO_4) are more effective than acids without oxyanions (e.g., HCl). This is because of competitive oxyanions (PO_4^{3-} or SO_4^{2-}) that may decrease the re-adsorption of anionic species of As on reactive surfaces of soil (Goh and Lim, 2005; Ko et al., 2005; Lee et al., 2007). However, with increasing awareness of post-remediation soil qualities, mild washing solutions may be preferred over strong acid-based washing solutions, because strong acids could adversely affect soils by dissolving soil components during washing treatment (Ko et al., 2005). Thus, washing solutions prepared with phosphate containing salts that have neutral or near-neutral pH can be used as alternatives to minimize detrimental effects (e.g., losses of soil OM and mineral substances (e.g., Si, Ca, Mg, Fe, Al), decreases in soil pH) of soil washing solutions (Alam et al., 2001; Tokunaga and Hakuta, 2002; Zeng et al., 2008).

The performance of washing solutions has largely been assessed on As extractability, and what happens to the residual As in soils or soil properties has not been extensively studied. Since there is a growing interest in minimizing negative effects of soil washing on soil qualities, the changes in soil properties and bioavailability of the residual As in soils after soil washing need to be studied. Therefore, this study investigates how bioavailability of the residual As in soils and soil properties of the washed soils are affected by soil washing with different solutions. Selected patented washing solutions for As-contaminated soils in Korea and the neutral phosphate salt-based washing solution (hereafter referred to as neutral phosphate solution), which was developed in order to avoid problems arising from soil acidity after soil washing with strong acids, were used for comparison. Specifically, the effect of using different washing solutions on changes in bioavailability of the residual As in soils and soil properties was determined by using chemical extraction-based methods (e.g., sequential extraction method) and bioassay test-based methods (e.g., seed germination and growth, microbial enzyme activities).

2. Materials and methods

2.1. Soil sampling

Historically As-contaminated soil samples were taken from the vicinity of a former smelter in Janghang, Korea. The soil samples were air-dried at room temperature, and sieved through a 2 mm sieve. The soil pH was 6.1, and the OM content determined by the Walkley–Black procedure (Walkley and Black, 1934) and the iron oxide concentration determined by the bicarbonate-citrate-dithionite (DBC) method (Mehra and Jackson, 1958) were 2.1% and 8240 mg kg⁻¹, respectively. The concentrations of available phosphorus and the total nitrogen were determined by using the Bray's method (Bray and Kurtz, 1945) and the micro-Kjeldahl method (Bremner, 1960), respectively. The detection limits for the Bray's and the micro-Kjeldahl methods were 0.01 mg L⁻¹ and 0.1 mg L⁻¹, respectively. The soil texture was silt loam consisting of 28% sand, 54% silt, and 18% clay. The background concentrations

of Pb, Cd, Cu, Zn, and As determined following the *aqua regia* extraction method with slight modifications (International Standard Organization, 1995), complying with Korea Ministry of Environment (Korea Ministry of Environment, 2009), were 97, 0.5, 52, 89, and 59 mg kg⁻¹, respectively. The As concentration (59 mg kg⁻¹) exceeded the cleanup level (25 mg kg⁻¹ for As) of the Korean Soil Environment Conservation Act.

2.2. Soil washing procedures

The soil washing condition and procedure using a neutral phosphate solution were developed in our previous study (Im et al., 2014). Briefly, 24 h soaking of As-contaminated soils in a 0.5 M phosphate solution (pH 6.0), prepared from ammonium dihydrogen phosphate ($\text{NH}_4\text{H}_2\text{PO}_4$) and diammonium hydrogen phosphate ($(\text{NH}_4)_2\text{HPO}_4$), at soil-to-solution (S/L) ratio of 1:5 was followed by 1 h washing at 200 rpm. Since conventional washing procedures involve a single-step soil washing treatment, a 1 h washing period was used with the three selected patented washing solutions for As removal from soils in Korea. The selected washing solutions are 1 M HCl (Choi et al., 2007), 0.5 M H_3PO_4 (Cheong and Kang, 2012), and 2% Na dithionite in 0.01 M HCl (Kim et al., 2012). All the washing experiments were carried out in triplicate using 1 L jars with 120 g soils and 600 mL washing solutions. After 1 h soil washing at 200 rpm, the washed soils were rinsed twice with deionized water. The washing and rinsing solutions can be treated either by neutralization and precipitation or by sorption on to adsorbents such as granular ferric oxide (Choi et al., 2007; Cheong and Kang, 2012; Yang et al., 2013). Also, As in the solutions can be removed by adding oxidants (e.g., hydrogen peroxide) and coagulants (Kim et al., 2012).

2.3. Extraction and analyses of As

For As analysis in soils, soil slurries in the washing experiments were left to settle in order to separate soils from washing solutions. The residual As in the separated and then dried soils (3 g) were extracted following the method described in Section 2.1. Chemical forms of As (i.e., fractionation of As) in soils were assessed using the sequential extraction procedure of Wenzel et al. (2001), which divides As in soils into five fractions, with a slight modification – (1) ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$; 0.05 M)-extractable As (i.e., non-specifically sorbed As); (2) $\text{NH}_4\text{H}_2\text{PO}_4$ (0.05 M)-extractable As (i.e., specifically sorbed As); (3) ammonium oxalate ($(\text{NH}_4)_2\text{C}_2\text{O}_4$; 0.2 M)-extractable As (i.e., As associated with amorphous hydrous oxides of Fe and Al); (4) $(\text{NH}_4)_2\text{C}_2\text{O}_4$ (0.2 M)/ascorbic acid (0.1 M)-extractable As (i.e., As associated with crystalline hydrous oxides of Fe and Al); and (5) residual As fraction extracted with the USEPA method 3052, which digest siliceous and organically based matrices such as soils and sediments in a mixture of nitric acid, hydrofluoric acid, HCl, and hydrogen peroxide using microwave heating (US Environmental Protection Agency, 1996). The As concentrations in the extracts were analyzed by using inductively coupled plasma-optical emission spectrometry (ICP-OES; ICP-730ES, Varian, USA). The method detection limit for As was 0.072 mg kg⁻¹. Statistical analysis was conducted using Excel 14.0 (Microsoft Corporation, USA) and the statistical significance of differences in As concentrations was determined by using the Student's *t*-test at 5% significance level.

2.4. Bioassay tests using plants and soil microbial enzymes

Seed germination and growth tests were carried out with ten seeds of Indian mustard (*Brassica juncea* L.) sown in each petri dish (diameter = 90 mm) containing dried soil equivalent to 30 g. The tests were carried out in triplicate under controlled environment

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