



# Removal of toxic metals from vanadium-contaminated soils using a washing method: Reagent selection and parameter optimization

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## HIGHLIGHTS

- Soil washing technology was applied to treat V-contaminated soil.
- The V-forms were analyzed using the BCR three-step sequential extraction procedure.
- Citric acid, tartaric acid, oxalic acid, and Na<sub>2</sub>EDTA exhibited V-high removal rate.
- Concentration of reagents, liquid-to-solid ratio, and washing times were optimized.
- V removal mechanism of different washing reagents was discussed.

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## ABSTRACT

Vanadium (V) contamination in soils is an increasing worldwide concern facing human health and environmental conservation. The fractionation of a metal influences its mobility and biological toxicity. We analyzed the fractionations of V and several other metals using the BCR three-step sequential extraction procedure. Among methods for removing metal contamination, soil washing is an effective permanent treatment. We conducted experiments to select the proper reagents and to optimize extraction conditions. Citric acid, tartaric acid, oxalic acid, and Na<sub>2</sub>EDTA all exhibited high removal rates of the extractable state of V. With a liquid-to-solid ratio of 10, washing with 0.4 mol/L citric acid, 0.4 mol/L tartaric acid, 0.4 mol/L oxalic acid, and 0.12 mol/L Na<sub>2</sub>EDTA led to removal rates of 91%, 88%, 88%, and 61%, respectively. The effect of multiple washing on removal rate was also explored. According to the changes observed in metal fractionations, differences in removal rates among reagents is likely associated with their pK<sub>a</sub> value, pH in solution, and chemical structure. We concluded that treating with appropriate washing reagents under optimal conditions can greatly enhance the remediation of vanadium-contaminated soils.

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## 1. Introduction

Vanadium (V) is an important metal resource used in the production of steel, batteries, and various chemicals. However, mining and smelting of V have caused serious environmental pollution in many countries, such as Russia, China and South Africa (Jiao and Teng, 2008). There have been many studies about V-

contamination, especially in China (Yang et al., 2017), such as Sichuan province (Teng et al., 2006), Hunan province (Xiao et al., 2015). V contamination not only inhibits plant growth (Hidalgo et al., 1988), but also endangers human health via the food chain, increasing the risk of cancer (Ghosh et al., 2015). Methods of removing metal contamination from soil can be divided into physical, chemical, and biological approaches. Although the large-scale application of chemical extraction processes presents some disadvantages, such as high costs of reagents, it can be a targeted approach to treat certain metal compounds and the fine-grained soils. The extracted metals may be easily recovered by a wide

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variety of methods (Dermont et al., 2008). Therefore, chemical remediation is still one of research hotspots.

Soil washing is one useful remediation method among chemical approaches, and has been previously applied to the removal of chromium (Cr) (Li et al., 2015a, b), lead (Pb) (Torres et al., 2012), zinc (Zn) (Udovic and Lestan, 2009), copper (Cu) (Gusiati and Klimiuk, 2012), arsenic (As) (Liao et al., 2016), and cadmium (Cd) (Li et al., 2013). Selection of the proper reagents is the key to effective soil washing. There are several types of soil washing reagents: inorganic acids, such as HCl (Udovic and Lestan, 2012); low-molecular-weight organic acids (LMWOS), such as citric acid (Wang and Mulligan, 2013; Chen et al., 2016), tartaric acid (Wang et al., 2015) and oxalic acid (Qiu et al., 2010b); chelates, such as EDTA (Zhang et al., 2010; Pocięcha et al., 2011; Voglar and Lestan, 2014) and Na<sub>2</sub>EDTA (Qiu et al., 2010a); surfactants (Mao et al., 2015; Chen et al., 2016); and alkalis (Kim, 2013). Soil washing with inorganic acids or basic solutions presents great disadvantages. The very low or high pH levels promote dissolution of metals, including some non-contaminant elements. The use of biodegradable washing reagents has been an excellent option on soil washing, which include: biodegradable chelating agents (such as EDDS and MGDA) (Hauser et al., 2005; Arwidsson et al., 2010a); biosurfactants (Mulligan and Wang, 2006; Dahrazma and Mulligan, 2007); the organic metabolites from Fungi (Arwidsson et al., 2010b); DOC solutions (Lee and Chen, 2010). Organic acids are widely used for soil washing due to their biodegradability (Di Palma and Mecozzi, 2007) and minimal effects on soil properties (Makino et al., 2008). To improve the efficiency of removal, efforts have been made to improve the washing process. Aside from reagent concentrations (Wang et al., 2015), the liquid-to-solid ratio (Zhang et al., 2013) and multiple washing (Makino et al., 2006) also have important effects on contaminant removal. However, few studies have investigated the use of soil washing to treat V-contaminated soil.

The fractionations of metals are related to their mobility and bioavailability in the soil, which have important effects on soil remediation. The Community Bureau of Reference (BCR) procedure is a useful tool for predicting short- and long-term mobility of trace elements, even under complex environmental conditions (Pueyo et al., 2008). Although, the sequential chemical extraction procedures can't determine specific chemical forms or the speciation of metals, it can match the target compound with the metal mobility in contaminated soil (Hlavay et al., 2004; Bacon and Davidson, 2008). This study analyzed the general process when metals were extracted from solid into liquid under the view point of washing technology. Therefore, we adopted BCR procedure in this study.

This work assessed a soil washing method for the treatment of V-contaminated soil. Selection of reagents is the basis of the washing method, as they determine the remediation time and effects. We investigated the removal rates of toxic metals using LMWOS and chelates. The effects of reagent concentration, liquid-to-solid ratio, and multiple washing on metal removal were also investigated in detail. Associated with a change in the fractionation of metals, we analyzed the removal mechanism of V when different reagents were employed, laying a foundation for choosing the proper washing reagents for V-contaminated soil.

## 2. Materials and methods

### 2.1. Soil samples

We collected samples from the surface (0–20 cm) soil of a V-polluted area in Hunan Province, China. There was some V slag in the sample. Soil samples were collected from several different sites, pretreated by air-drying, crushing, and sieving (2 mm mesh), and then sealed in bag and kept in the shade. The basic physicochemical

properties of these samples are presented in Table 1. We used a handheld XRF to test the total concentrations of toxic metals in each soil sample 3 times. All of the reagents used in this study, including citric acid, tartaric acid, oxalic acid, acetic acid, sodium acetate, ethylenediaminetetraacetic acid (EDTA), disodium ethylenediaminetetraacetate (Na<sub>2</sub>EDTA), used most frequently in soil washing according to the references: are analytical grade and were purchased from Sinopharm, Co., Ltd., China.

### 2.2. Washing experiments

We systemically investigated the influence of reagent concentration, liquid-to-solid ratio, and multiple washing on the removal of toxic metals. In a typical washing experiment, soil samples were placed into individual centrifuge tubes that had been air-dried after soaking in 10% nitric acid. Next, various reagents were added to the tubes, which were shaken at 250 rpm in a water bath at 25 °C (SHA-BA Shaking table, Jintan Science Analysis Instrument Co., Ltd., China). After 24 h, the tubes were centrifuged for 20 min at 3000 g, the supernatant from each was filtered through a 0.45 μm membrane, and the filter was stored in a 10 mL centrifuge tube. Five concentrations were tested for each of four selected reagents (citric acid, tartaric acid, and oxalic acid: 0.05, 0.1, 0.2, 0.4, and 0.8 mol/L, respectively; Na<sub>2</sub>EDTA: 0.04, 0.08, 0.1, 0.12, and 0.16 mol/L, respectively). We also investigated five liquid-to-solid ratios (1.5, 2, 5, 10, and 15). Based on several liquid-to-solid ratios, sample soil and 30 mL reagent at the optimal concentration were placed in individual 50 mL centrifuge tubes. Equal volumes of reagents were used in all of the tubes to avoid errors due to non-uniform stirring. Then we washed soil sample five times, each time discarding the supernatant and retaining the sediment.

### 2.3. Analytical methods

We used the BCR three-step sequential extraction procedure (Rauret et al., 1999) to analyze the toxic metal fractionations, which included the acid-extractable state (F1), reducible state (F2), oxidizable state (F3), and residual state (F4). Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES; ThermoFisher, Waltham, MA, USA) was used to determine toxic metal concentrations. Each experiment was replicated three times. The toxic metal leaching rate was calculated as follows:

$$R = \frac{c \cdot V}{m \cdot M} \times 100\%$$

where R is the removal rate of the extractable state for a given toxic metal, in%; c is the concentration of toxic metal in the leachate, ppm; V is the volume of soil leachate, mL; m is the mass of soil in the tube, g; and M is the mass concentration of the extractable state for a given toxic metal in the soil, mg/kg.

## 3. Results and discussion

### 3.1. Soil analysis

The total toxic metal contents of each soil sample are presented in Table 1. Compared to the regulatory limits published in *Standard for environmental quality of agricultural land in China (third revision)*, four toxic metals (V, Cu, Zn, and Cr) exceeded the limits. Total V and Cr were 10 and 4 times greater than the standards, respectively, indicating that the soil sample for this study was seriously polluted with V and Cr. In two Polish fields, it was found for vanadium speciation in soils with different vanadium content: from 5 to 250 mg/kg (Poledniok and Buhl, 2003). In Sichuan Province, China,

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