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Research article

Feasibility of four wastes to remove heavy metals from contaminated soils

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ABSTRACT

Soil washing is one of the permanent techniques to remove heavy metals, and washing agent is a key influence factor for this technique, but there is still lack of high-efficiency, eco-friendly, and inexpensive agents. In this study, four wastes including pineapple peel (PP), soybean straw (SS), broad bean straw (BBS) and tea residue (TR) were employed to remove cadmium (Cd), lead (Pb) and zinc (Zn) in contaminated soils. The Fourier transform infrared spectroscopy (FTIR) analysis indicated that hydroxyl, carboxyl, amine, carbonyl and amide groups were involved in the interaction with metal ions by complexation or ion exchange. We then investigated the influences of various conditions including washing solution concentration, pH, and washing time. The metal removal efficiencies with these agents increased as the concentration augmented from 5 to 80 $g\,L^{-1}$, decreased or presented an asymmetric V-shaped curve with increasing pH from 2.5 to 7.5, and fit intraparticle diffusion or Elovich model with washing time increasing. PP has the highest removals for Cd (90.1%), Pb (18.6%), and Zn (15.2%) in soil A, and 85.8, 24.8, and 69.4% in soil B, respectively. The relatively high metal removal was mainly attributed to effective removal of the exchangeable and acid soluble fractions. Moreover, single washing not only lowered the potential ecological risk of the heavy metals, but moderated the effects on soil chemical properties. Therefore, PP was a feasible washing agent to remediate soils contaminated by heavy metals.

1. Introduction

Soil contamination with heavy metals has become a major environment concern around the world for the last decades, due to mining, smelting and waste water irrigating (Jez and Lestan, 2016; Liu and Chen, 2013). Among these heavy metals, cadmium (Cd), lead (Pb), zinc (Zn) are non-biodegradable and persistent in the soil (Adrees et al., 2015; Wu et al., 2010). Moreover, they pose serious threats to plant, animal and human health because of their high toxicity (Adrees et al., 2015). Consequently, it is imperative to remediate those soils contaminated by Cd, Pb and Zn.

Soil washing, which is cost-effective and time-efficient (Mao et al., 2015), is one of the few permanent techniques for removing metal contaminants from soils (Dermont et al., 2008). Therefore, this technology has been widely utilized in heavy metal removal (Hu et al., 2014; Kulikowska et al., 2015b). And soil washing efficiency is closely associated to the washing agents. Recently, acids, chelators and surfactants have been used to extract heavy metals from contaminated soils (Dermont et al., 2008). Among these washing agents, chelators including ethylenediaminetetra-acetic acid (EDTA) and low-molecular-weight organic acids (LMWOAs) are used frequently (Hu et al., 2014; Pérez-Esteban et al., 2013; Wang et al., 2016). In particular, EDTA, as a synthetic chelating agent, is effective (Jelusic et al., 2013). However, it is low-toxicity and easily retained in soil (Jez and Lestan, 2016), thus its excessive dose may affect soil quality and pollute groundwater (Wu et al., 2010). Moreover, metal-EDTA complexes may have toxic effects (Zupanc et al., 2014). LMWOAs such as citric and tartaric acids were alternates to synthetic chelating agents (Pérez-Esteban et al., 2013). Although these agents are degradable and have less destruction for remediation of soils (Wen et al., 2009), they may cause relatively much soil nutrient loss (Liu and Lin, 2013). Consequently, there is an urgent need to search for other high-efficiency, eco-friendly, and inexpensive reagents.

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Wastes from agro-industrial production may be promising alternatives to remove heavy metals in soil for they contain varieties of functional groups that can bind with metal cations (Nguyen et al., 2013). Furthermore, some studies reported that their application can maintain or increase soil organic carbon and diminish soil nutrient loss (Liu and Chen, 2013; Medina et al., 2015). Only a few of wastes including wine-processing waste sludge, however, have been proven to be high removal for Cd from contaminated soils (Liu and Chen, 2013; Liu and Lin, 2013). Specifically, relatively little attention has been devoted to studying the effects of pineapple peel (PP), soybean straw (SS), broad bean straw (BBS) and tea residue (TR) on soil heavy metal removal via washing. It would thus be of interest to study whether these four materials will be feasible for metal removal.

The aims of this study were to: (1) assess the ability of the four materials for heavy metal removal under different conditions including concentration, pH and time; (2) evaluate changes in the metal fraction distribution and potential ecological risk from metals; (3) discuss the effects of washing on the soil chemical properties.

2. Materials and methods

2.1. Soil sampling and characterization

Two soils (0–20 cm) contaminated by Cd, Pb and Zn were collected from a farmland near an abandoned mine in Hanyuan, Sichuan (102°38′E, 29°24′N) and used in the study. The soil samples (A and B) were air-dried, ground and then passed through a 2-mm nylon mesh to obtain homogenous soils.

The determination methods of conventional soil properties were presented in Appendix A, Supplementary data. The both soils are classified as moisture soils with a sandy loam texture. The following characteristics were obtained from the original soils A and B: sand (81.20 and 83.40%), silt (12.87 and 6.44%) and clay (5.93 and 10.16%), pH 6.82 ± 0.15 and 7.07 ± 0.21, and CEC 7.42 ± 0.22 and 6.23 ± 0.13 cmol (+) kg^{-1}, respectively. Total Cd, Pb, and Zn concentrations were 13.42 ± 0.53, 52.12 ± 1.16, 92.91 ± 2.35 mg kg^{-1}, and 34.93 ± 0.75, 231.73 ± 5.90, 658.06 ± 16.72 mg kg^{-1}, respectively.

2.2. Preparation of soil washing solutions

The four materials were acquired from Chengdu, Sichuan. They were air-dried, ground, passed through 2-mm mesh, and then added into plastic bottles with distilled water. The bottles were continuously shaken on a shaker (140 rpm, 12 h) at 25 °C. After shaking finished, the suspensions were filtered to collect their supernatant. As described by the method above, a series of solutions with different concentrations were prepared for the washing experiment.

2.3. Effects of washing solution concentration, pH, and washing time on metal removal

The 100-mL acid-rinsed plastic bottles containing 2 g of soils were prepared for batch washing experiments, followed by the addition of 20-mL solutions. The same experiments were conducted by distilled water as controls. And next, they were continuously shaken on a shaker (200 rpm) at 25 °C for a certain period of time. Subsequently, the suspensions were centrifuged at 4000 rpm for 5 min and filtrated through a 42 Whatman filter paper. The concentrations of Cd, Pb and Zn were measured by AAS. All chemical reagents used in all experiments were analytically pure. And each treatment was performed in triplicate. The batch washing experiments were performed as following parameters:

A Washing solution concentrations: 0, 5.0, 20.0, 35.0, 50.0, 65.0, and 80.0 g L^{-1} (soil: liquid ratio of 1: 10, washing time of 2 h, solution pH of 2.5). The concentration with relatively high removal will be used to analyze the effects of the other factors on metal removal efficiency.
B Washing solution pH values: 2.5, 3.5, 4.5, 5.5, 6.5, and 7.5 (soil: liquid ratio of 1: 10, washing time of 2 h). The pH values were adjusted with dilute HNO_3 or NaOH. The pH with relatively high removal will be used in the further batch washing experiments.
C Washing time: 0.25, 0.5, 1.0, 2.0, 4.0, and 8.0 h (soil: liquid ratio of 1: 10).

2.4. FTIR analysis

The solutions of the four materials before and after washing in soils A and B (65 g L^{-1}, pH 2.5, solid/liquid of 1:10 and washing time 4 h) were dried at 60 °C in an oven, ground with spectroscopic grade KBr, and subsequently pressed them into disks. The FTIR spectra, used to analyze the chemical bonds reacting with metals, were measured in the range of 4000–400 cm^{-1} wavenumber with 32 scans and a resolution of 4 cm^{-1} by a PerkinElmer spectrometer (Spectrum Two, PerkinElmer Inc., USA).

2.5. Fraction distribution and potential ecological risks of heavy metals in soils

To assess the distribution and potential ecological risks of metals in contaminated soils before and after washing, the modified BCR 3-step sequential extraction procedure was applied (Nemati et al., 2011). Soil metals were separated into operational defined four fractions: exchangeable and acid soluble, reducible, oxidizable and residual.

In addition, the potential ecological risk of each individual metal (E_r^i) was calculated with the following Eq. (1) (Kulikowska et al., 2015a):

$$E_r^i = T_r^i \times C_D^i \times \Omega \Big/ C_R^i \qquad (1)$$

where T_r^i is the toxic-response factor of given metals (Cd = 30, Pb = 5, Zn = 1) according to the approach of Hakanson (1980). C_D^i is the soil metal concentration. C_R^i is the threshold value of metals in soil (Cd, 0.45 mg kg^{-1}; Pb, 80 mg kg^{-1}; Zn, 250 mg kg^{-1} (Wang et al., 2016). Ω is the modified index of metal concentration calculated as A δ + B (A is percentage of the exchangeable and acid soluble fractions (F1) for a given metal, B = 1 - A, δ is the toxic index of the F1 fraction), The δ values differ with relation to the percentage of the F1 fraction of metal: 1.0 ($1\% \le F1 \le 10\%$); 1.2 ($11\% \le F1 \le 30\%$); 1.4 ($31\% \le F1 \le 50\%$) and 1.6 ($F1 > 50\%$) (Perin et al., 1985; Zhu et al., 2012). Each heavy metal in the soils can be classified as potentially low risk, moderate risk, considerable risk, high risk, and very high risk when an E_r^i value of <40, 40–80, 80–160, 160–320, and >320, respectively (Hakanson, 1980).

2.6. Statistical analysis

All data statistical analyses were performed using SPSS version 19.0 (SPSS Inc., Chicago, Illinois). One-way analysis of variance (ANOVA) was used to compare whether the metal removal under different experimental conditions was significantly different. It was considered significant if a *P* value of <0.05.