



Release of Pb in soils washed with various extractants



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ABSTRACT

The suitability of an agent for washing heavy metal contaminated soils depends not only on the efficiency of heavy metal removal but also on the persistence of the washing effect, especially for agricultural soil. Redistribution of residual heavy metals in washed soil from inert pools to labile pools can reverse the initial washing effect. In order to study the redistribution of residual Pb in Pb-contaminated soils washed with citric acid (CA), ethylenediaminetetraacetic acid (EDTA), FeCl₃, and HCl solutions, washed soils were incubated under both flood and 70% field water capacity (FWC) conditions for 180 days. The Pb availability in CA-, EDTA-, FeCl₃-, and HCl-washed soils varied with incubation time. Diethylenetriaminepentaacetic acid (DTPA)-Pb generally increased with incubation time, except in CA-washed soil under flood incubation condition. Pb associated with the exchangeable/acid extractable fraction (F1) increased, whereas Pb in the easily reducible fraction (F2) decreased over time, for all washed soils under flood incubation condition; the changes in the amount of Pb associated with each fraction were much smaller under 70% FWC incubation. The transformation of Pb from F2 to F1 during flood incubation was largely caused by the mobilization of Fe and Mn oxides. The mobility factors (M_f) of Pb for samples under flood incubation increased with increasing incubation time while those for samples under 70% FWC incubation changed only slightly, which implies increasing environmental risks of Pb from washed soils that are flooded. Therefore, the redistribution and potential environmental risks posed by heavy metals in washed soils, especially in paddy rice soils, should be taken into account when the suitability of a washing agent is evaluated. In the present study, CA was found to be unsuitable for washing Pb-contaminated agricultural soil, whereas the FeCl₃ solution was the most stable washing agent tested, as it had high removal efficiency and low release of Pb after washing.

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1. Introduction

Soil contaminated with Pb, due to activities such as mining and industrialization, has gained much attention (Arunakumara et al., 2013; Fang et al., 2014; Li et al., 2014). Pb in soils can enter into livestock and humans through the food chain (Dong et al., 2011; Nabulo et al., 2010). Excessive intake of Pb can be damaging to human health (e.g., affecting nervous, skeletal, and enzymatic systems; Zhang et al., 2012). Pb affects intelligence and skeletal development of children, and children younger than 6 years are more sensitive to Pb than older children (Qin et al., 2010). Therefore, soils contaminated by Pb must be remediated immediately.

Soil washing can be used to permanently remove toxic metals from soils (Dermont et al., 2008). Soil washing can remove most of the toxic

metals associated with labile fractions and can slightly affect metal pools in non-labile fractions (Hernandez-Soriano et al., 2011; Wuana et al., 2010; Ye et al., 2014). It is well known that toxic metals associated with labile fractions are much more toxic, mobile, and bioavailable to livestock than metals associated with non-labile fractions (Zhang et al., 2010). On the other hand, most studies have focused on the removal efficiency and redistribution of heavy metals after washing (Ash et al., 2015; Finzgar et al., 2014); therefore, the fate of the residual fraction is still unclear. Re-establishing the equilibrium that is disturbed during soil washing could initiate the transformation of non-labile fractions to labile fractions (Udovic and Lestan, 2009). Although it is obvious that residual heavy metals after soil washing may be toxic to the surrounding environment if they are transformed into mobile and bioavailable fractions, the extent to which this occurs is still not well known. The transformation of residual heavy metals should be affected by various abiotic and biotic soil factors (Udovic et al., 2007). Soil pH, soil redox potential (Eh), and organic matter affect the characteristics of heavy metals by altering their fractionation, mobility, and metal-complexes (Kim and Owens, 2009); these alterations may pose ecological risks to the surrounding environment. Changes in environmental conditions could cause heavy metals in less mobile fractions to redistribute to labile fractions (Bourg and Loch, 1995; Jean et al., 2007). It is

Abbreviations: CA, citric acid; DDW, double distilled water; Eh, soil redox potential; F1, exchangeable and acid extractable fraction; F2, easily reducible fraction; F3, oxidizable fraction; F4, residual fraction; FWC, field water capacity; P1, 7 days of soil incubation; P2, 15 days of soil incubation; P3, 30 days of soil incubation; P4, 60 days of soil incubation; P5, 90 days of soil incubation; P6, 120 days of soil incubation; P7, 150 days of soil incubation; P8, 180 days of soil incubation.

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important to understand whether residual heavy metals in washed soils are immobile, stable, or would become mobile if conditions were to change. Because the ultimate goal of soil washing is re-introducing the washed soils for agricultural production, the stability of residual heavy metals in washed soils determine the success (immobile and stable) or failure (mobile) of a soil washing process.

The objective of this study was to assess the long-term transformation of Pb in soils washed with various extractants under flood and 70% field water capacity incubation conditions and to evaluate the utility of each soil washing process from the perspective of Pb release for Pb-contaminated agricultural soils.

2. Materials and methods

2.1. Study area and soil

The study area closes to a former smelter in the town of Meixian, Youxi County, Fujian Province, China (latitude: 26°15'6"N, longitude: 118°15'20"E; elevation is about 90 m above sea level). It belongs to the subtropical maritime monsoon climate with annual average temperature and rainfall of 19.6 °C and 1600–1700 mm, respectively. The mining activity in this area began >30 years ago. The contaminated soil used in this study was collected from the Ap horizon (0–20 cm) of an agricultural field, in which rice and vegetables used to be rotationally cultivated. The soil was classified as sandy loam, mixed, thermic, Typic Endoaquepts (Soil Survey Staff, 2014). Soil was transported back to the laboratory to air-dried, ground and passed through 10-mm and then 2-mm laboratory test sieves, and homogenized. Soil passed through a 10-mm sieve was used for soil washing and the soil passed through a 2-mm sieve was used for soil physical and chemical analysis. Total Pb of this studied soil was 1550.2 mg kg⁻¹.

Soil pH was measured using a pH meter with a complex glass electrode (pHS-3E, Shanghai Precision & Scientific Instrument Co., Ltd, China) at a soil-to-water ratio of 1:2.5. The cation-exchange capacity (CEC) was determined by the ammonium acetate (1 mol L⁻¹ NH₄OAc, pH = 7.0) method, and particle size was determined by the sedimentation method (SSSC, 1999). The organic matter content was determined by wet oxidation with H₂SO₄–K₂Cr₂O₇ (SSSC, 1999). The soil was digested with HNO₃–HCl–HClO₄–HF to determine the total Pb concentration. Pb concentration in the digested solution was determined using inductively coupled plasma-mass spectrometry (ICP-Mass, Nexion 300, Perkin Elmer, NY). A soil sample with a certified concentration of Pb (GBW07407), provided by the China National Center for Standard Materials, was used as a reference. The recoveries of Pb from the reference soils were 95–108%. Soil field water capacity (FWC) was determined by the laboratory Wilcox method (see Supplementary method) (Zhu, 1996). Physicochemical characteristics of the contaminated soil are shown in Table 1.

2.2. Soil washing

Five hundred grams of air-dried soil was placed in a 2000 mL plastic bottle and mixed with a 0.1 mol L⁻¹ solution of one of four reagents (citric acid, Na₂EDTA, HCl, or FeCl₃) at a soil-solution ratio of 1:2. The

bottle was shaken on a back-and-forth shaker (HY-8, Changzhou, China) for 2 h at 25 ± 2 °C (210 rpm). After shaking, the supernatant was drained off. This washing process was repeated twice, as previous studies have indicated that this optimizes washing numbers for Pb removal (Chen et al., 2015). The washed soil was then rewashed with double distilled water (DDW) to remove the mobilized metals and was air-dried. In order to obtain enough washed soil for further use, washing process with one of the extractant as described above was repeated for thirty times. The washed soils of the same extractant were mixed together for incubation. The Pb concentrations in the washing solutions are presented in Table S1. Washing with citric acid (CA), EDTA, FeCl₃ and HCl removed 9.0, 39.5, 29.7 and 18.6% of total soil Pb, respectively.

2.3. Incubation

Samples (200 g) of air-dried washed or untreated soils were placed into individual 250-mL bottles. To stimulate paddy field and dry land environments, the soil samples were subjected to flood (about 1 cm water above the soil surface) or to 70% FWC conditions with DDW. Totally, there were twenty-four treatments for each washed and untreated soil under either flood or 70% FWC incubation. Samples were covered with cling film to decrease water evaporation, and incubated at 25 ± 2 °C. During the incubation period (180 days), DDW was regularly replenished to the soils to maintain flood or 70% FWC conditions. Soils were sampled after 7, 15, 30, 60, 90, 120, 150 and 180 days of incubation, which correspond to period 1 (P1), period 2 (P2), period 3 (P3), period 4 (P4), period 5 (P5), period 6 (P6), period 7 (P7), and period 8 (P8), respectively. At each sampling time, the soils in three bottles of each kind of washed soil were taken, air-dried and passed through 2 mm sieve for further analysis. The pH and Eh of the soil solutions were measured at each sampling time using an automatic ORP depolarization tester (FJA-6, Nanjing, China).

2.4. Determination of available soil Pb

The available Pb in each soil sample was extracted with a diethylenetriaminepentaacetic acid (DTPA) solution. The extraction solution contained 0.005 mol L⁻¹ DTPA, 0.01 mol L⁻¹ CaCl₂, and 0.1 mol L⁻¹ triethanolamine (TEA) and was adjusted to pH 7.30 ± 0.05. In a 100 mL polyethylene centrifugation tube, 5.00 g of soil was shaken with 10 mL of the DTPA extraction solution for 2 h on a horizontal shaker at approximately 180 rpm. After shaking, samples were centrifuged for 15 min at 3000 rpm (DT5-2, Beijing, China) and filtered through a 0.45 μm Millipore filter. The Pb concentration in the filtrate was determined using ICP-Mass. Extractions were conducted in triplicate.

2.5. BCR sequential extraction

A three-stage modified BCR sequential extraction procedure (Rauret et al., 2000) was used to determine the fractionation of Pb, Mn, and Fe in the soils after incubation for 7, 90, and 180 days. Four operationally defined fractions were sequentially determined: exchangeable and acid extractable fraction (F1), easily reducible fraction (F2), oxidizable fraction (F3), and residual fraction (F4). All data reported were means of triplicate analyses. A soil sample with certified concentrations of Pb (GBW080037) was provided by the China National Center for Standard Materials and was used as a reference. The recoveries of Pb from the reference soil were 88.3 to 105.6%. Pb in untreated soil was mostly bound to F2 and F4 (66.8 and 21.8% of total Pb, respectively), with very little Pb in F1 and F3 (Chen et al., 2015).

2.6. Lead mobility in soils

The mobility factor developed by Gusiatin and Klimiuk (2012) was used to describe the mobility of Pb in incubated soil samples. The Pb

Table 1
Chemical and physical properties of the contaminated soil.

Soil sample	pH	OM ^a (g kg ⁻¹)	CEC ^b (cmol (+) kg ⁻¹)	Clay Silt Sand			Texture	Total Pb (mg kg ⁻¹)
				%				
MX	6.4	23.5	9.0	12.1	15.1	72.8	SL ^c	1550.2

^a Organic matter.

^b Cation-exchange capacity.

^c Sandy loam.

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