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Immobilization of lead in anthropogenic contaminated soils using phosphates with/without oxalic acid

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ABSTRACT

Understanding the effects of oxalic acid (OA) on the immobilization of Pb(II) in contaminated soils by phosphate materials, has considerable benefits for risk assessment and remediation strategies for the soil. A series of phosphate amendments with/without oxalic acid were applied to two anthropogenic contaminated soils. We investigated the immobilization of Pb(II) by KH_2PO_4 , phosphate rock (PR), activated phosphate rock (APR) and synthetic hydroxyapatite (HAP) at different phosphate:Pb (P:Pb) molar ratios (0, 0.6, 2.0 and 4.0) in the presence/absence of 50 mmol oxalic acid/kg soil, respectively. The effects of treatments were evaluated using single extraction with deionized water or CaCl_2 , Community Bureau of Reference (BCR) sequential extraction and toxicity characteristic leaching procedure (TCLP) methods. Our results showed that the concentration of water extractable, exchangeable and TCLP-Pb all decreased with incubation time. The concentration of water-extractable Pb after 120 days was reduced by 100% when soils were amended with APR, HAP and HAP + OA, and the TCLP-Pb was <5 mg/L for the red soil at P:Pb molar ratio 4.0. Water-soluble Pb could not be detected and the TCLP-Pb was <5 mg/L at all treatments applied to the yellow-brown soil. BCR results indicated that APR was most effective, although a slight enhancement of water-soluble phosphate was detected at the P:Pb molar ratio 4.0 at the beginning of incubation. Oxalic acid activated phosphates, and so mixing insoluble phosphates with oxalic acid may be a useful strategy to improve their effectiveness in reducing Pb bioavailability.

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Introduction

Heavy metals and metalloids remain of much environmental concern. Lead (Pb), a naturally occurring metal, has always been present in soils, surface and underground water. It is considered to be a toxic element for both plants and animals. There are several sources of Pb contamination to the environment such as flaking paint, use of leaded gasoline, waste incineration, application of pesticides and mining operations (Silvetti et al., 2014). Accumulation of Pb in soils and its transport through the soil matrix are potential threats to human health, especially to children by ingestion of Pb-contaminated soil (Melamed et al., 2003). Lead in the human body can replace Ca and Zn in

proteins (e.g., synaptotagmin, amino levulinic acid dehydratase) and deactivates them (Shahid et al., 2012). It is therefore very important to identify factors determining the bioavailability and bioaccessibility of Pb. The biogeochemical behavior of lead strongly depends on its chemical speciation (Shahid et al., 2012). The bioavailability of lead can be decreased by forming compound with various materials so as to decrease its toxicity (Tang and Yang, 2012; Almaroai et al., 2014; Saoiabi et al., 2013; Wu et al., 2013). In contaminated soils it can be effectively decreased by converting labile Pb species into pyromorphite $\text{Pb}_5(\text{PO}_4)_3\text{Cl}$, which can be achieved by amendment with various phosphates (Maneck et al., 2006). For example, addition of insoluble phosphates to Pb-contaminated soil is effective in

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converting Pb from forms with high availability to the most strongly bound Pb fractions (Miretzky and Fernandez-Cirelli, 2008). The effectiveness of such Pb immobilization has been evaluated by single/sequential extraction and toxicity leaching procedure. Efficient Pb immobilization using phosphate amendments requires an increase in the solubility of the phosphate phase and of the Pb species phase by inducing acid conditions (Miretzky and Fernandez-Cirelli, 2008). Although soluble phosphate addition seems to be highly effective, excess phosphate in the soil can bring about the eutrophication of surface water (Shahid et al., 2014). The use of mixed treatments may be a useful strategy to improve their effectiveness in increasing lead immobilization and reducing the phosphate leaching (Cao et al., 2004; Jiang et al., 2012). Phosphate solubilizing bacteria (PSB) increase the solubilization of insoluble phosphate compounds such as phosphate rocks (PRs) through the production of organic acids and soluble phosphates (Park et al., 2011a).

Low-molecular-weight organic acids (LMWOAs) occur widely in water and soil as natural products of root exudates, microbial secretions, and exudation from dead or living cells (Gao et al., 2003). LMWOAs have been shown to enhance phytoremediation of soil contaminated with trace elements (Qin et al., 2004; Jiang et al., 2011). These organic acids have chelating characteristics, or being biodegradable, can form humified substances which solubilize heavy metals. When a plant is stressed by a high concentration of heavy metal, or low concentrations of calcium and phosphorus, its root release some LMWOAs which complex with the heavy metal (Qin et al., 2004; Evangelou et al., 2007; Magdziak et al., 2011; Ghnaya et al., 2013). Organic acids may thus play a significant role in PR dissolution. Some studies have shown that organic acids are effective in releasing phosphate from low and medium reactive PRs, but are ineffective in releasing phosphate from high reactive PRs (Kpombekou-A and Tabatabai, 2003). Some results have also demonstrated that organic acids have potential as amendments for increasing plant-available phosphate in PR-treated soils. However, Elliott and Herzig (1999) reported that oxalic acid was ineffective in removing Pb from a highly contaminated soil. Previous studies have addressed the effect of different forms of phosphate such as H_2PO_4 , K_2HPO_4 , $(NH_4)_2HPO_4$, $Ca(H_2PO_4)_2 \cdot H_2O$, and $Ca_5(PO_4)_3X$ ($X = F, Cl, Br, \text{ or } OH$) on the immobilization of metals (e.g., Pb, Zn, Cu, Cd) (Basta and McGowen, 2004; Kamiishi and Utsunomiya, 2013). All these forms ranging from relatively soluble amorphous phosphates to more thermodynamically stable hydroxyapatite have been used for remediating trace metal contaminated soils. LMWOAs are effective in releasing phosphate from low and medium reactive PRs (Kpombekou-A and Tabatabai, 2003; Jiang et al., 2012). Although the effects of several organic acids on heavy metal sorption and desorption on various clay minerals and oxides have also been studied (Gao et al., 2003; Qin et al., 2004; Yuan et al., 2007), there is limited information on the immobilization of heavy metals by phosphates, as affected by organic acids, especially the interactions between heavy metals, organic acid and phosphates in natural soils (Qin et al., 2004; Wang et al., 2009).

In the present research, we used incubation experiments simulating field conditions to investigate the effectiveness of phosphate-containing materials and organic acids on lead immobilization. It is possible that soluble phosphate (KH_2PO_4) compounds effectively immobilize Pb in soil, but induce to phosphate leaching. However, insoluble (phosphate rock, activated phosphate rocks and synthetic hydroxyapatite) compounds in the presence of oxalic acid enhance the immobilization of Pb and decrease the leaching of both phosphate and Pb (Wei et al., 2014). The aim of this study was to evaluate the leaching of phosphate and Pb after incubation by both soluble and insoluble phosphates (in the presence and absence of oxalic acid) compounds in two anthropogenic contaminated soils by single/sequential extraction and toxicity characteristics leaching procedure methods.

1. Materials and methods

1.1. Sampling, preparation and soil analysis

Buck soil samples were collected from 0 to 20 cm depth in Xianning (red soil, an udic ferralsol) and Xiaogan (yellow-brown soil, an alfisol) County, Hubei Province, China. The two soils were air dried and grounded to pass through a 2-mm mesh.

To simulate Pb contamination, $Pb(NO_3)_2$ was added to the sieved soils to achieve a total Pb contamination of 625 mg/kg. The Pb-amended soils were thoroughly stored at room temperature for 1 month to equilibrate, maintaining a soil moisture content of 20%. Sub-samples of the soils were characterized for pH, organic matter (OM), cation exchange capacity (CEC), particle size and Pb content using standard methods (Bao, 1999). Selected properties are presented in Table 1.

1.2. Soil amendments

Phosphorus and LMWOA amendments used in this study include potassium dihydrogenphosphate (KH_2PO_4), a natural phosphate rock (PR) from Zhongxiang, Hubei Province, China, oxalic acid-activated phosphate rock (APR) and synthetic hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$, HAP) and the oxalic acid (OA).

Synthesis of HAP: Aqueous solutions of ammonia hydrogen phosphate (0.3 mol/L) were slowly added drop by drop into calcium nitrate (0.5 mol/L) at a rate of 20 mL/min in a flask in a water bath at 40°C. Ammonium hydroxide (25%) was added to adjust the pH to 10. The resultant solution was allowed to react for 24 hr and was then diluted with distilled water and pure ethanol until the conductivity of the filtrate was <2 mS/m. The resulting residues were then dried at 80°C for 24 hr and mechanical polished (Guo et al., 2004). All chemicals were of analytical grade and purchased from Sinopharm Chemical Co. (Shanghai, China).

Activation of PR: The PR was mixed with 0.5 mol/L oxalic acid at a liquid–solid ratio of 10:1 and equilibrated at $(28 \pm 1)^\circ C$ for 6 days (Jiang et al., 2012).

Characterization of materials: The materials obtained were characterized by powder X-ray diffraction. The patterns were obtained using a D8 Bruker Advance X-ray diffractometer (Bruker D8 Advance, Germany) with Cu K α radiation (35 kV, 30 mA). The patterns of diffraction were obtained in a 2θ scanning range from 5° to 70° , with 0.05° and 5 sec of scan step and time, respectively (Fig. 1).

1.3. Incubation experiment

Experiments were conducted in a 250-mL plastic container (7 cm diameter and 5 cm height) containing 200 g soil. Phosphate amendments were produced at four levels according to P:Pb molar ratios 0, 0.6, 2.0 and 4.0, applied with and without oxalic acid. The amount of phosphate required for each treatment was calculated on the basis of the total phosphate content of the amendments. Treated soils were thoroughly mixed with various amendments prior to potting, and then were stored at room temperature for 120 days, maintaining soil moisture 20% (W/W). Sampling was triplicated for each treatment. During incubation, approximately 30 g soil was collected at 3, 7, 49, and 120 days, and ground to pass

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