



## Behaviors of exogenous Pb in P-based amended soil investigated with isotopic labeling method coupled with Tessier approach



Zhi-Yong Huang<sup>a,\*</sup>, Jian Li<sup>a</sup>, Ying-Lan Cao<sup>a</sup>, Chao Cai<sup>b</sup>, Zhi Zhang<sup>a</sup>

<sup>a</sup> College of Bioengineering, Jimei University, Xiamen 361021, China

<sup>b</sup> Key Laboratory of Urban Environment and Health, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China

### ARTICLE INFO

#### Article history:

Received 1 June 2015

Received in revised form 5 October 2015

Accepted 18 October 2015

Available online 26 October 2015

#### Keywords:

Agricultural soil

Exogenous Pb

Passivation

P-based compound

Isotopic labeling method

Tessier approach

### ABSTRACT

Lead (Pb) pollution in soil has attracted public attention because of its serious impacts on human health through food chain. However, the chemical and biological behaviors of exogenous Pb in soil remain unknown, specifically in the passivated soil with phosphate (P)-based materials. By spiking soil with stable enriched isotopes of <sup>206</sup>Pb (>99%), the exogenous Pb<sup>2+</sup> in soil was simulated in the present study; the distribution, transformation, and bioavailability of exogenous Pb in soil passivated with the P-based compound were quantitatively investigated using an isotopic labeling method coupled with a Tessier approach of sequential extraction procedures. The results showed that exogenous Pb in water-soluble fraction (F1) accounted for 0.58% of the total spiked isotopes, which was slightly higher than that of the intrinsic Pb (0.34%) in soil. Nevertheless, the exogenous Pb percentages in exchangeable fraction (F2), carbonate bound fraction (F3), Fe/Mn oxide fraction (F4), and organic matter bound fraction (F5) were about twice more than those of the intrinsic Pb in the four fractions. But exogenous Pb in residual fraction (F6) was only 45.6%, while intrinsic Pb was 70%. After the soil was passivated with P-based compound consisting of diammonium phosphate, bone meal, and hydroxyapatite, more than 90% of exogenous Pb in F2 could be immobilized, but small Pb percentage in F1 was immobilized. Additionally, Pb species in F2, F3, and F4 might be converted into different forms in F5 and F6, but the immobilized Pb in F5 and F6 was labile because more than 50% of the exogenous Pb in the two fractions was lost after planting. The results demonstrated that the isotopic labeling technique coupled with Tessier approach was a novel method that can evaluate the distribution, transformation, and bioavailability of exogenous Pb in soil. These results are helpful for further immobilization of exogenous Pb in agricultural soil.

© 2015 Elsevier B.V. All rights reserved.

### 1. Introduction

Lead (Pb) contamination in soil is among the most important ecological problems worldwide that has attracted more attention because of its non-degradation and persistence in soil and its potential threat on human health through the food chain. High Pb levels in agricultural soil in China are usually due to industrial emissions, mining, misuse of chemical fertilizers, and sewage waste discharge (Huang et al., 2012a; Yu et al., 2012; Zhao et al., 2010). Mining is among the main sources of Pb for agricultural soil contamination, specifically in mine areas, because of exploitation, tailing, wastewater, and mining dust (Liu et al., 2005). Hence, revealing the chemical and biological behaviors of exogenous Pb in soil is essential because the new metal contaminant may present different chemical and biological behaviors compared with those of the original Pb (marked as intrinsic Pb) in soil.

Passivation based on various phosphorus (P) materials is an alternative chemical method to immobilize Pb in soil in situ, in which Pb can be

mainly converted into insoluble forms, such as pyromorphite (Basta and McGowan, 2004; Cao et al., 2004; Chen et al., 2006; Chen et al., 2007; Miretzky and Fernandez-Cirelli, 2008; Zhu et al., 2004). Many P-based materials, including soluble, moderately soluble, and insoluble phosphates, such as (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub>, K<sub>2</sub>HPO<sub>4</sub>, CaHPO<sub>4</sub>, Ca(H<sub>2</sub>PO<sub>4</sub>)<sub>2</sub>, bone meal, hydroxyapatite, and apatite, have been used as the passivating reagents. The multi-component consisting of two or more phosphate materials have also been applied (Park et al., 2011). Significant efforts have been made to evaluate the immobilization effectiveness of Pb in soil passivated with P-based materials (Chen et al., 2007; Hettiarachchi and Pierzynski, 2002; Miretzky and Fernandez-Cirelli, 2008; Park et al., 2011; Ryan et al., 2001); BCR and Tessier approaches, which are based on the combination of specific extractions, are typically used (Gleyzes et al., 2002). For example, phosphate rocks can convert Pb species into water soluble, exchangeable, carbonate, Fe–Mn oxide, and organic fractions into residual fraction forms by the Tessier approach of sequential extraction procedures (SEP), hence effectively reducing Pb bioavailability in soil (Ma and Rao, 1997). After mixing with hydroxyapatite for 240 d and partitioning with a Tessier approach, Ryan et al. (2001) found that more than 45% of Pb in soil is converted into insoluble

\* Corresponding author.

E-mail address: [zhyhuang@jmu.edu.cn](mailto:zhyhuang@jmu.edu.cn) (Z.-Y. Huang).

forms, thereby reducing Pb labile pools by immobilization. However, SEP approach cannot distinctly distinguish the different behaviors of exogenous and intrinsic Pb in soil under P-based passivation.

With several advantages over the traditional radioisotope labeling technique, stable isotopic labeling method is becoming popular in the investigation of metals' mobility and potential bioavailability in soil, specifically for exogenous Pb in soil matrices (Huang et al., 2011; Pickhardt et al., 2002; Rodríguez-Cea et al., 2006; Xie et al., 2012). Recently, the stable isotopic labeling technique combined with SEP approach has been successfully applied to qualitatively investigate the chemical and biological activities of exogenous mercury and Pb in soils in our previous reports (Huang et al., 2012b; Huang et al., 2014). The isotopic labeling method coupled with sequential chemical extraction is a novel approach that can enable us to investigate the distribution, transformation, and bioavailability of heavy metals in soil.

To date, few data are available on the evaluation of chemical and biological behaviors of exogenous Pb in passivated soil. In the present study, the contamination of exogenous Pb in soil is simulated by spiking the soil with the stable enriched  $^{206}\text{Pb}$  isotopes, and a quantitative method using isotopic labeling technique coupled with Tessier approach of SEP is applied to investigate the chemical and biological behaviors of the exogenous and intrinsic Pb in soil passivated with P-based materials. The present study aims to investigate the distribution, transformation, and bioavailability of the exogenous Pb in soil passivated with P-based materials.

## 2. Materials and methods

### 2.1. Soil sampling

Soil samples collected from a large piece of vegetable-growing land were suspected for Pb pollution caused by emissions from an abandoned lead–zinc mine. This land is located at the Liancheng County of Fujian Province, China. At least six subsamples of topsoil (0–20 cm) were collected and blended. After being transported to the laboratory, the soil samples were oven-dried for 72 h at 40 °C, ground with a glass mortar, passed through an 80 mesh sieve, and finally stored in polypropylene bags.

### 2.2. Analysis of soil characteristics

The soil pH value, contents of organic matter, and cation-exchange capacity were measured according to the methods described in our previous report (Xie et al., 2012).

### 2.3. Simulation of exogenous Pb in soil by spiking stable enriched isotopes

To simulate the contamination of exogenous Pb in soil sample, an enriched  $^{206}\text{Pb}$  isotopic reagent (>99%, Isoflex, USA) was spiked in the form of  $\text{Pb}(\text{NO}_3)_2$  at a ratio of 75.00 mg of  $\text{Pb}^{2+}$  per 1000 g of soil. The addition of enriched isotopes spiked in soil was based on the following: (1) minimum influence of the spiked isotopes on soil properties (Xie et al., 2012); and (2) minimum analytical error in the measurement of isotope ratios ( $^{208}\text{Pb}/^{206}\text{Pb}$ ) (Zeng et al., 2013). The spiked soil was mixed with pure water up to 70%–80% of moisture and incubated at room temperature for 30 d. The sample was blended regularly at least twice a day to ensure full contact of the spiked  $^{206}\text{Pb}$  with the intrinsic Pb in soil.

### 2.4. Immobilization of Pb with P-based compound

After spiking the enriched  $^{206}\text{Pb}$ , about 1000 g of moist soil was mixed with the optimized phosphate compound consisting of 10 g of bone meal, 20 g of diammonium phosphate, and 15 g of hydroxyapatite. The spiked soil without treatment with P-based compound was selected as a control group. All the soil samples were again incubated at room

temperature for 60 d. Subsequently, the soil samples were subjected to the sequential chemical extraction based on Tessier approach for the measurement of  $R_{\text{Pb}}$  ( $^{208}\text{Pb}/^{206}\text{Pb}$ ) and Pb concentrations in each Pb fraction.

### 2.5. Pot experiment

Each of the 500 g soil samples was placed into a perforated plastic pot (about 800 ml) for pot experiment. Four seedlings (~5 cm) of Chinese mustard (*Brassica juncea*), which is considered to be the species of readily accumulating soil Pb (Zaier et al., 2010), were planted. Four replicates were carried out for this experiment. The plants were grown in a controlled environment at 20 °C and 12 dark/12 h light. The moisture content was maintained at 80% with pure water daily. Approximately 20 ml of mixed nutrient solution consisting of  $\text{CO}(\text{NH}_2)_2$  (0.5 g l<sup>-1</sup>),  $\text{KH}_2\text{PO}_4$  (0.3 g l<sup>-1</sup>),  $\text{MgSO}_4$  (0.05 g l<sup>-1</sup>),  $\text{KNO}_3$  (0.7 g l<sup>-1</sup>), and  $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$  (0.0006 g l<sup>-1</sup>) was added per 20 d. After 60 d of growth, the plants were harvested, rinsed in 50 mmol l<sup>-1</sup> of EDTA for 5 min, and washed with deionized water and pure water. The plants were dried at 60 °C for 72 h, ground, and digested with 5 ml of 16 mol l<sup>-1</sup>  $\text{HNO}_3$  to measure  $R_{\text{Pb}}$  ( $^{208}\text{Pb}/^{206}\text{Pb}$ ) and Pb concentrations.

At the end of the pot experiment, the rhizospheric soil adhering to the root system was carefully collected by shaking the roots. After drying, the rhizospheric soil was partitioned with Tessier approach for the measurement of  $R_{\text{Pb}}$  ( $^{208}\text{Pb}/^{206}\text{Pb}$ ) and Pb concentrations in each Pb fraction.

### 2.6. Fractionation of Pb species in soil with Tessier approach

Based on the methods in previous reports (Kabala and Singh, 2001; Salbu et al., 1998), Pb species in soil samples were partitioned with a Tessier approach, as briefly described in Table 1. Six fractions of Pb species in soil were operationally defined as follows: water-soluble fraction (F1); exchangeable fraction (F2); carbonate bound fraction (F3); Fe/Mn oxide fraction (F4); organic matter bound fraction (F5); and residual fraction (F6). The Pb concentrations and the isotope ratios of  $R_{\text{Pb}}$  ( $^{208}\text{Pb}/^{206}\text{Pb}$ ) in each of the fractions were measured with atomic absorption spectroscopy (AAS) and inductively coupled plasma mass spectrometry (ICP-MS), respectively.

### 2.7. Measurement of Pb concentrations and isotope ratios ( $R_{\text{Pb}}$ )

The Pb concentrations in soil and plants and various chemical fractions were measured with a ZEEnit 700 AAS instrument. The quality assurance of Pb measurement in soil by AAS was achieved by adding Pb standards with an average recovery of  $89.8 \pm 4.5\%$ .

The isotope ratios of  $R_{\text{Pb}}$  ( $^{208}\text{Pb}/^{206}\text{Pb}$ ) in soil and chemical fractions were measured with a 7500a instrument (Agilent, USA) of ICP-MS according to the method described in our previous report (Huang et al., 2012a). About 50 ng ml<sup>-1</sup> of Pb 981 (National Institute of Standards and Technology, USA), a reference material consisting of natural isotopic composition, was used for the quality assurance of  $R_{\text{Pb}}$  measurement. The measured Pb isotope ratio was 2.1681 with an average precision (expressed as relative standard deviation, RSD) of 0.242%. Moreover, 50 ng ml<sup>-1</sup> of Tl (GSB G62070-90, National Analytical Center of Steel Material, China) was used as an internal standard for the correction of mass bias and signal fluctuation during the long-term  $R_{\text{Pb}}$  ( $^{208}\text{Pb}/^{206}\text{Pb}$ ) measurement.

### 2.8. Quantitative evaluation of the exogenous and intrinsic Pb distributions in samples

The exogenous and intrinsic Pb concentrations in soil samples and plants were calculated based on the method described in our previous report (Huang et al., 2014). Briefly, the calculation method was based on the mass balances of isotopes in soil samples, in which the mole

Download English Version:

<https://daneshyari.com/en/article/4573089>

Download Persian Version:

<https://daneshyari.com/article/4573089>

[Daneshyari.com](https://daneshyari.com)