Ecotoxicology and Environmental Safety 165 (2018) 589–596

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Distribution and transformation of lead in rice plants grown in contaminated soil amended with biochar and lime

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ARTICLE INFO

Keywords:
Biochar
Lime
Lead
Rice
Transformation

ABSTRACT

This study aimed to investigate the effects of rice straw biochar and lime (RBL) on the remediation of lead (Pb)-contaminated soil and mitigation of Pb translocation in rice plants by using pot experiments. Lead-contaminated soil collected from a farmland near a Pb-zinc (Zn) mine, biochar, limestone powder, and indica rice (*Oryza sativa* L.) were used in the present study. The experimental treatments included: (1) control (CK), (2) 2.5% biochar (RB1), (3) 5% biochar (RB2), (4) 0.6% lime (L1), (5) 1.2% lime (L2), (6) 2.5% biochar + 0.6% lime (RBL1), and (7) 2.5% biochar + 1.2% lime (RBL2). The results revealed that the treatment with RBL was more efficient than the treatment with only biochar or lime in decreasing $CaCl_2$-extractable Pb content in the soil by increasing soil pH and soluble sulfate content in the soil. Treatment with RBL reduced in the accumulation of Pb in the shoot of rice plants, this was mainly attributed to the decrease in the concentration of available Pb in the soil. The RBL2 treatment not only decreased the concentration of Pb in brown rice by 84.33% and Pb distribution in rice embryo, but also increased rice yield by 53.38% from that of the control. Further, unlike biochar treatment, RBL and lime treatments decreased the translocation of Pb in rice plants. The RBL treatment increased the proportion of Pb distributed in the cell wall and reduced the mobility of Pb in plant tissues. Thus, application of biochar and lime in combination is more effective than their individual application in reducing the availability of Pb in the soil and Pb accumulation in brown rice.

1. Introduction

Rice (*Oryza sativa* L.) is the staple food in many Asian countries and a dominant food crop in the tropical and subtropical regions of Southern China where the soil is mostly acidic (Zhao et al., 2015a). Large areas agricultural soil have been contaminated with lead (Pb) (Soylak et al., 2001; Soylak and Türkoglu, 1999), some of the areas near mine diggings have been used to produce rice, resulting in the accumulation of Pb in rice grains. An investigation on rice grains collected from mining-impacted paddy fields revealed that about 34% of the samples contained Pb higher than the limit set for Pb in rice in China (Williams et al., 2009).

Stabilization is a widely used method to reduce the bioavailability and transfer of heavy metals in the environment, mainly through adsorption, precipitation, and complexation reactions to redistribute the metals from a soluble phase to stable phase (Porter et al., 2004). Phosphate compounds, liming materials, organic composts, and ferrous/manganese (Fe/Mn) oxides are commonly used to immobilize heavy metals (McCann et al., 2015). Lime, an alkaline amendment, decreases the availability of heavy metals in soil by decreasing the hydrogen ion (H^+) concentration, thereby increasing negatively charged sites in the soil (Bolan et al., 2014). The addition of lime decreases the soluble and extractable contents of zinc (Zn), Pb, nickel (Ni), cadmium (Cd), and copper (Cu) contents in soil, further, it reduces the uptake of heavy metals in *Festuca rubra* (Gray et al., 2006). However, the over use of lime might harden the soil by increasing the linkage

Abbreviations: RBL, rice straw biochar and lime; Pb, lead; XRF, X-ray fluorescence spectrometry; HM, heavy metal; LA-ICP-MS, laser-ablation inductively coupled plasma mass spectrometry; EC, electrical conductivity; DCB, dithionite-citrate-bicarbonate; OCT, optimum cutting temperature; μ-SRXRF, micro-synchrotron radiation X-ray fluorescence microanalysis; SR, synchrotron radiation; TF, transfer factor

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https://doi.org/10.1016/j.ecoenv.2018.09.039
Received 3 January 2018; Received in revised form 5 September 2018; Accepted 7 September 2018
Available online 17 September 2018

between calcium (Ca)-ion and kaolinite particles (Chemeda et al., 2015). Furthermore, the application of lime was shown to adversely affect the germination of *Vaccinium myrtillus* seeds (Olsson and Kellner, 2002). Recently, biochar has been used as a novel carbonaceous material to adsorb Pb in polluted water or soil (Li et al., 2016). Biochar has a large surface area, active organic functional groups, micro-pore structure, and alkaline properties (Wu et al., 2012). The increase in the release of potassium (K), Ca, and phosphorus (P) from biochar was found to be related to high Pb immobilization. A study performed using X-ray diffraction analysis revealed that biochar produced from dairy manure contains high concentration of available P, which can combine with Pb and form stable hydroxypyromorphite ($Pb_5(PO_4)_3(OH)$) (Cao et al., 2011). Moon et al. (2013) conducted scanning electron microscopy/energy dispersive spectroscopy (SEM-EDX) and X-ray fluorescence spectrometry (XRF) analyses and showed that the effective immobilization of Pb after treatment with soybean stover-derived biochar is associated with the pozzolanic reaction products, chloropyromorphite, and Pb-phosphate. Biochar has shown positive effects on the immobilization of Pb, Cu, and chromium (Cr) in soil. It not only acts as a soil amendment for immobilizing heavy metals, but also improves soil properties and promotes nitrogen fixation in soil (Ultra et al., 2016). However, some studies have revealed that biochar can increase the mobility of some metals, such as arsenic (As), antimony (Sb), and Cu, in the soil, thereby increasing their concentration in plants (Beesley et al., 2010; Mendez et al., 2012). Furthermore, biochar increases the cost of agricultural production. Therefore, a combination of different amendments has been used to remediate heavy metal (HM)-contaminated fields. Rehman et al. (2017) revealed that a combination of limestone and biochar was relatively effective in minimizing Cd concentration in both rice and wheat plants with increased benefit-cost ratio. Thus, the application of biochar and lime (BRL) to Pb-contaminated soil was hypothesized to be more efficient in decreasing the transfer of Pb from soil to the rice system than the application of only biochar or lime.

The subcellular distribution of Pb in plant tissues determines the accumulation and detoxification of Pb in plants (Wang et al., 2015a). Zhao et al. (2015b) showed that the mechanism of Pb detoxification in plants involves the localization of Pb on the cell wall and in the soluble fraction of plant tissues with low biological activity via cell wall deposition and vacuolar compartmentalization. Lead in the root cells is mainly accumulated in the cell wall, cell membrane, cell voids, mitochondria, and peroxisomes (Malecka et al., 2008). Furthermore, Pb distributed in the soluble fraction of shoot tissue is the key source of Pb transfer to the grain; however, Pb in the cell wall fraction can restrict Pb transfer from the shoot to grain (Liu et al., 2015). The subcellular distribution of Pb in plants has been shown to be affected by amendments to soil. Ethylene diamine tetraacetic acid application in soil has been shown to promote Pb accumulation in the soluble and cellular organelles fractions, whereas diethyl aminoethyl hexanoate was found to increase the proportion of Pb in the cell wall fraction (He et al., 2013).

The information on the spatial distribution of HMs is crucial to understand how different tissues and cell types control their distribution, complexation, and storage. Further, understanding how this process affects the transfer of HMs in rice plants is necessary. Thus far, studies on the imaging and localization of elements, and in situ chemical speciation have used synchrotron-based techniques, mass spectrometry-based techniques, and laser-ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) (Zhao et al., 2014a). Moradi et al. (2010) used an LA-ICP-MS and observed higher amount of Ni stored in the cortex of Ni-spiked roots than that in the stele, whereas a contrary pattern was observed in the control plants. In the present study, differential centrifugation, micro-XRF, and LA-ICP-MS methods were used to reveal the transfer of Pb in rice.

This study aimed to investigate the effects of the combination of biochar and lime application on (1) Pb mobility in soil and (2) the transport of Pb in rice plants.

2. Material and methods

2.1. Soil sampling and amendments

Lead-contaminated soil was collected from the arable layer (0–20 cm) of a farmland near a mine in Youxi County, Fujian Province, China. The soil was air dried, ground, passed through a 2 cm sieve, and homogenized for the pot experiment. The physicochemical properties of the soil were determined, and the results are shown in Supplementary Table S1. The pH and electrical conductivity (EC) of a 5:1 water/soil suspension were determined using a pH meter (SevenCompact; Mettler-Toledo, Greifensee, Switzerland) and an EC meter (DDS-307A; INESA, Shanghai, China), respectively. The concentrations of carbon (C) and nitrogen (N) in the soil were analyzed using an elemental analyzer (vario MAX cube; Elementar, Hesse, Germany). The cation-exchange capacity of the soil was determined using 1 M ammonium acetate (pH = 7.0; Jackson, 1979). The soil particle size distribution was determined using the pipette method after hydrogen peroxide treatment (Jackson, 1979). Biochar was procured from Nanjing Qinfeng Crop-straw Technology Company (Nanjing, Jiansu Province, China). It was produced from rice straw at 500 °C for 2 h. Lime was obtained from Longyan, Fujian Province, China. The properties of biochar and lime were determined according to the procedure of Rehman et al. (2017), and are shown in Supplementary Table S1. The pH of amendments was measured in deionized water to H_2O ratio of 1:20 by using the pH meter (SevenCompact; Mettler-Toledo, Greifensee, Switzerland). The concentrations of C, N, and sulfur (S) of the amendments were analyzed using an elemental analyzer (vario MAX cube; Elementar, Hesse, Germany). The specific surface area was determined using the gas adsorption method (Micromeritics Instruments, TriStar II 3020, USA). The total concentration of Cd, Pb and Zn in the amendments were determined using an induced couple plasma-mass spectrometer (ICP-MS; NexION 300 ×; Perkin Elmer, NY, USA).

The pot experiments comprised seven treatments in triplicates, including a control treatment (CK) without amendments. For the experiment, 7.5 kg air-dried soil was added to each pot, and N, P and K fertilizers were applied at the rates of 0.15 g N kg^{-1}, 0.10 g P_2O_5 kg^{-1}, and 0.15 g K_2O kg^{-1} in the forms of urea, $NH_4H_2PO_4$ and K_2SO_4, respectively. The height and diameter of the pots were 25 and 20 cm, respectively. The fertilized soil was treated individually with 2.5% (w/w) biochar (RB1), 5% (w/w) biochar (RB2), 0.6% (w/w) lime (L1), 1.2% (w/w) lime (L2), 2.5% (w/w) biochar + 0.6% (w/w) lime (RBL1), or 2.5% (w/w) biochar + 1.2% (w/w) lime (RBL2), as reported by Shu et al. (2016) and Dong et al. (2016). Before rice transplanting, all the pots were maintained under flooded condition for 1 week. Rice seeds (Donglian 5 of conventional *O. sativa* L. ssp. *indica*) were washed with 0.5% active sodium hypochlorite for 15 min, soaked in deionized water, and germinated under moist condition for 3 d. After germination, the rice seedlings were transferred to beakers and grown for 17 d. Five uniform seedlings were selected and transplanted into each pot. Flooding condition (2–3 cm water level) was maintained throughout the growing period.

2.2. Rice harvest and analysis

Soil and plant samples were collected from each pot at tillering (50 days after transplanting) and maturity (114 days after transplanting) stages, to determine the soil properties, bioavailable Pb concentration of soil and Pb accumulation by rice at the vegetative and reproductive growth. After air drying, the soil sample was ground using a roller and passed through a 2 mm sieve. Next, 20 g of the 2 mm soil particles was ground using an agate mortar and passed through 0.149 mm sieve. The bioavailable Pb concentration in the soil sample was measured using 0.1 M calcium chloride ($CaCl_2$) solution. About 5 g of air-dried soil was weighted into a flask, mixed with 25 mL of $CaCl_2$ solution, shaken at