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Mobility and phytoavailability of As and Pb in a contaminated soil using pine sawdust biochar under systematic change of redox conditions

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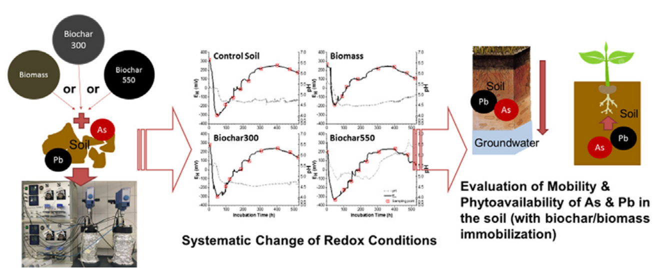
HIGHLIGHTS

- Reducing conditions increased and biomass addition decreased the mobility of As.
- Rhizosphere exudates may induce higher phytoavailable - As under oxic conditions.
- O-containing functional groups of biomass may control the mobility of As and Pb.
- The increment of pH caused by BC550 increases the mobility and availability of As.
- Alkaline conditions provided by BC550 reduce the phytoavailable Pb.

GRAPHICAL ABSTRACT



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ABSTRACT

Biochar has been adopted to control the mobility and phytoavailability of trace elements (TEs) in soils. To date, no attempt has been made to determine the mobility and phytoavailability of arsenic (As) and lead (Pb) in a contaminated soil with biochars as amendments under predefined redox potentials (E_H). Thus, in this study, a soil contaminated with As and Pb (2047 and 1677 mg kg^{-1}, respectively) was pre-incubated for 105 days with three amendments (pine sawdust biomass (BM) and two biochars produced from the same feedstock at 300 °C (BC300) and 550 °C (BC550)). The aged samples were then exposed to dynamic E_H conditions to evaluate the mobility and phytoavailability of As and Pb after immobilization. The BM amendment significantly decreased and the BC300 slightly reduced the mobility and phytoavailability of As and Pb, which may be related to the oxygen-containing functional groups on the surface of BM and BC300. In contrast, BC550 increased the mobility of As at −300 to −100 mV and

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100 mV, enhanced the phytoavailability of As under oxidizing condition (>100 mV), but reduced the phytoavailability of Pb, which might be caused by the properties of amendments and redox chemistry of the TEs. The effectiveness of BM and biochars for the stabilization of As and Pb varied under dynamic E_H conditions, which indicates that detailed investigations should be conducted before the applications of biochar as soil amendment under variable environmental conditions, especially for contaminated paddy soils.

1. Introduction

Contamination of soils with trace elements (TEs) due to anthropogenic activities threatens sustainability of agroecosystems (Park et al., 2016). For instance, arsenic (As) and lead (Pb) are highly toxic to plant, animals and humans, and they are listed among the priority hazardous contaminants (Shih et al., 2016). Simultaneous remediation on anionic and cationic TEs contaminated soils is known to be challenging (Fang et al., 2016). Total concentrations of TEs are not able to provide sufficient information about the potential mobilization and phytoavailability (Huang and Matzner, 2007; Rinklebe et al., 2016a,b). Therefore, soil remediation technologies should, besides reducing the total amount, also aim to minimize the easily mobilized and available amount of TEs (Shaheen and Rinklebe, 2015). Low-cost amendments such as carbonaceous recycled products (e,g., compost), agricultural wastes, inorganic minerals, and industrial by-products successfully reduced the mobility and phytoavailability of TEs in soil by adsorption, co-precipitation, surface complexation, and ion exchange (Ahmad et al., 2014b; Tsang et al., 2014; Zhang et al., 2015).

Biochar, as a by-product of pyrolysis, has become a popular amendment to immobilize TEs in contaminated soils (Beesley et al., 2011; Ahmad et al., 2016b). It can achieve cost-effective contaminant stabilization, carbon sequestration, and enhancement of soil quality by nutrient replenishment, water retention, and reinforcing the enzymatic and microbial activity (Awad et al., 2013; Ahmad et al., 2014b, 2016a). The properties of biochar are known to be affected mainly by the feedstocks and the pyrolysis temperature. In general, biochars produced at a lower temperature (~300 °C) have more oxygen-containing functional groups, while those produced at a higher temperature (500–700 °C) possess higher surface area and more micropores (Uchimiya et al., 2011b; Ahmad et al., 2012b; Rajapaksha et al., 2014), which influence their capacity for immobilization. It has been reported that the mobility of Pb was substantially reduced in many cases (Cao et al., 2011; Park et al., 2011a; Uchimiya et al., 2012), whereas the mobility of As was enhanced by the increase of soil pH, release of phosphorus (P), and increasing amount of arsenite due to reduction (Beesley and Marmiroli, 2011; Park et al., 2011b).

In spite of a large number of studies on the application of low-cost amendments, the stability of contaminant immobilization under variable environmental conditions is still questionable (Beesley et al., 2010; Tsang et al., 2013b; Beiyuan et al., 2016; Rizwan et al., 2016a). In particular, the variation of redox conditions influences the fate of TEs via changes in pH, dissolved organic carbon (DOC), microorganism activities, and the redox chemistry of iron (Fe), manganese (Mn), and sulfur (S) (Du Laing et al., 2009b; Frohne et al., 2011, 2014; Huang, 2014; Schulz-Zunkel et al., 2015; Rinklebe et al., 2016b). For instance, increasing redox potential (E_H) facilitated the oxidation processes that generate protons and reduce solution pH (Yu et al., 2007). The solubility of arsenate (As^V) increases in soil with increasing pH in a range of 3–8, whereas arsenite (As^{III}) acts in an opposite way (Fitz and Wenzel, 2002). In addition, Fe/Mn (hydr)oxides are reduced to Fe^{2+} and Mn^{2+} as E_H decreases, so the associated TEs could be mobilized in soil. Under reductive condition, an increase of As mobility can be observed as As^V is reduced to the more mobile and toxic As^{III}, which may be further enhanced by the addition of biochar (Frohne et al., 2011, 2015; Choppala et al., 2016; Vithanage et al., 2017).

The predefined change of E_H could affect the TEs immobilization by biochar, as revealed by our recent findings (Rinklebe et al., 2016a). However, up to date, no attempt has been made to determine the mobility and phytoavailability of As and Pb in a contaminated soil with well-characterized biochars as amendments under pre-definite E_H. Thus, our goal was to mechanistically study the impact of dynamic redox conditions and associated pH changes on the mobility and phytoavailability of As and Pb in the soil solid phase using a highly sophisticated automated biogeochemical microcosm system. In particular, the liability of pine sawdust (agricultural waste) and two biochars produced at different temperatures (300 and 550 °C) as soil amendment on a field-contaminated soil were used to study the difference of amendments under oxidizing and reducing conditions.

2. Methodology

2.1. Soil and amendments characterization

The contaminated soil was collected from the upper 30 cm of an agricultural field in Gongju city, Chungcheongnam-do Province, Korea near the Tancheon mine. The studied field was banned for agricultural usage after detection of a high content of As and Pb in the soil. Soil sample was air-dried and passed, after removing debris, through a 2-mm sieve before use. The contents of TEs in soil were analyzed according to the US EPA Method 3051a (Table 1). Biomass of pine sawdust (BM) was collected from a sawmill company in Seoul, Korea, and washed with deionized water, air dried, ground and sieved under 1 mm. Pine sawdust biochars, which showed good effectiveness in previous studies, were produced at 300 °C (BC300) and 550 °C (BC550) with a heating rate of 7 °C min^{-1} according to the described method in Lou et al. (2016a, 2016b). Table S1 provides the physiochemical characteristics of BM, BC300, and BC550.

2.2. Incubation experiment

A 105-day pre-incubation was conducted for all amendments including the control soil to allow a proper aging of the biochar in the soil. The control soil (CS), soil treated with BM (S&BM), soil with BC300 (S&BC300), and soil with BC550 (S&BC550) at a dosage of 5% (which is equal to an application rate of 70 t ha^{-1}) were incubated at 70% of water-holding capacity of the soil. After 105-day incubation (aging) at 25 °C, soil samples from each amendment (CS, S&BM, S&BC300, and S&BC550) were air-dried and characterized physically and chemically (Table 1).