



Remediation of Cu, Pb, Zn and Cd-contaminated agricultural soil using a combined red mud and compost amendment



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ABSTRACT

Red mud and compost are two inexpensive and effective materials available for the in situ treatment of soil contaminated with heavy metals. In order to investigate the specific process involved, a detailed analysis of the synergistic effects of combined red mud and compost was carried out. The efficiency of three different treatment systems, namely compost (C), red mud (R), and a mixture of compost and red mud (R + C), on the movability, ecological risk, and biological effective Cu, Zn, Cd, and Pb over time of incubation in agricultural soil was evaluated. Results showed that the pH, water extractable organic carbon (WEOC) and total organic carbon (TOC) showed significant changes after addition of these amendment materials to the target soil. The combination of red mud and compost mixture as amendment material (R + C) resulted in the largest reduction in efficiency of the available heavy metals. In addition, the microbial biomass in the treated soil increased after addition of the amendment materials, highest increase was observed with R + C. The results indicate that these amendment materials, especially R + C could lower the ecological risk of heavy metals in soil.

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1. Introduction

In recent years, as population growth and the continuous expansion of industrial production, agricultural and industrial activities have caused serious heavy metal contamination in widespread areas on the planet (Beech and Cheung, 1995; Cheung and Gu, 2007; Jackson et al., 2009; Kumar et al., 2012; Tsekova et al., 2010; Xu et al., 2004; Zhou et al., 2015, 2016). Heavy metals as pollutants entering soils, especially agricultural soils, expose a major threat to human health and ecosystem risk due to its stability and non-microbial degradation and mineralization (Aryal et al., 2016; Colin et al., 2012; Luo et al., 2011; Nirola et al., 2016; Sayara

et al., 2011; Sultana et al., 2014; Zhuang et al., 2009). Consequently, formulation of effective ways to treat and control heavy metal pollution in soils is a great challenge in both research and application.

There are at least two ways available to remediate heavy metal contaminated soils: one is to modify the chemical states of heavy metals in soil so that mobility, biological availability and environmental risk can be reduced by stabilization of heavy metals; the other is to eliminate heavy metals from soils, making the soil heavy metal content close to or reach the soil background values (Sar et al., 1999; Shi et al., 2009; Wuana and Okieimen, 2011). The stabilization technology to remediate heavy metal pollution of soils has received widespread attention because of its effectiveness, short time period for treatment, competitive price, wide applicability, and small impact on the eco-environment (Kumpiene et al., 2008; Lee et al., 2009). Therefore, many stabilizing agents have been tested to treat and control heavy metal pollution in soil system, such as lime, red mud, biochar and compost (Bolan et al., 2014; Ding et al., 2016; Du et al., 2010; Gray et al., 2006; Huang et al.,

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2016; Li et al., 2016; Ruyters et al., 2011; Shi et al., 2009; Wang et al., 2016; White et al., 1995; Xu et al., 2016). Compost (C), the main product of stabilization process from agricultural solid waste in particular, is one of the most inexpensive materials available for the stabilization of heavy metal polluted soil in situ, because of other technologies of physical and chemical treatments are either inadequate or too expensive in comparison (Beesley et al., 2010; Clemente and Bernal, 2006; Karami et al., 2011; Walker et al., 2004). Compost was used for the remediation of Cu-Zn mine spoil, reduced the bio-availability and biotoxicity of heavy metals, and accelerated establishment of *Bromus carinatus* biomass (O'Dell et al., 2007). Manure and compost were implemented for the amendment of pyritic mine waste contaminated soil to promote bud growth and decrease the concentrations of Cu, Zn, and Mn in the bud, and the phytoavailability of heavy metals to plants (Walker et al., 2004). Actinobacteria was used for the remediation of co-contaminated soils with metals and organic compounds, reduced the concentrations of lindane and Cr (VI) (Polti et al., 2014). *Geotrichum* sp. and *Bacillus* sp. were implemented for the amendment to remove Cr (VI) efficiently from contaminated soils (Qu et al., 2016). The *Bacillus* sp. strain AKD1 were used for the remediation of co-contaminated with heavy metal and cypermethrin, reduced the concentrations of Li^{2+} , Fe^{2+} and V^{5+} (Tiwary and Dubey, 2016). A transgenic bacterium *Bacillus cereus* BW-03 (pPW-05) was used for the remediation of contaminated site for heavy metals, removed 96.4% inorganic mercury synergistically (Dash and Das, 2015). In our previous work, compost was used for the con of Cu, Zn in underground water and soil under the condition of simulated rainfall (Chen et al., 2010). We also used compost for the remediation of toxic organic pollutants and heavy metal pollution in waste water (Zeng et al., 2012).

Red mud (R), an industrial solid waste produced by extracting alumina from bauxite, has drawn public attention as a soil improvement material (Nadaroglu and Kalkan, 2012; Westman et al., 2009). It has many beneficial immobilization capabilities as a heavy metal stabilizer, due to its rich in iron-aluminum oxide, active functional groups, and high pH (Nadaroglu and Kalkan, 2012; Westman et al., 2009). And it also has been tested for its strong ability for the removal heavy metals. For example, Gray and co-workers assessed the validity of red mud, secondary product of aluminum production process, for the reduction of metal availability, and found that soil pH increased and the concentration of available heavy metals decreased after addition of red mud with dosage of 3% or 5% (Gray et al., 2006). Others reported that red mud had powerful ability for chemical fixation of heavy metals, and higher efficiency for reducing solubility and bioavailability of metals in soil (Santona et al., 2006). Red mud for the amendment of polluted soil reduced the concentration of available heavy metals and promoted soil enzyme activity and pH of soil (Garau et al., 2007). Besides, in our previous work, the exchangeable fraction of Pb, Zn and Cd in polluted farmland soil showed a significant reduction after the application of red mud (Luo et al., 2012; MeiRong et al., 2012).

Red mud and compost, as the effective and inexpensive soil amendment materials, have been used to restore and remediate the polluted soils. For example, the interaction of red mud and compost could provide an effective means to enhance the effectiveness of red mud or compost amendment when putting together (Ming, 2008; Zhou et al., 2012). Red mud had the best efficiency for reducing the extractable Zn and Pb fractions instantly, while compost and mud did not show any effect for Pb. Compost, obtained from the source-separated municipal solid waste, and red mud could decrease the leachability of Zn, Mn and Ni concentration in polluted mine soil effectively. In fact, composting is a biological process in which some pollutants were removed or immobilized by

microbes, using different raw materials, e.g., municipal solid waste, agricultural solid waste, industrial waste (Brunori et al., 2005; Chen et al., 2015). Variable results were obtained from the remediation of soil because of different concentrations of heavy metals in contaminated soils, long term or short time of contamination history, and also the different activity of soil microbes (Wu et al., 2016).

In this work, rice straw, an abundant supply of agricultural solid waste, was selected as raw material to produce the compost. The concentrations of Cu, Zn, Cd and Pb in soil pore water, available Cu, Zn, Cd and Pb in soil, and soil physical and chemical properties after the treatment with compost, red mud, or R + C to polluted soil. The effects of compost and red mud in combination as restoration materials for lowering the bio-availability and movability of heavy metals were investigated. Furthermore, the effect of compost and red mud on microbial biomass in soil and the ecological risk of heavy metals were also studied.

2. Materials and methods

2.1. Agricultural soil samples

In this work, the agricultural soil samples were obtained from a farmland of Fangzhuang town (Located in east longitude 113° 26' 51.91", latitude 35° 13' 23.75"), which is located in Jiaozuo city of Henan province, China (Fig. 1). Henan province is located in the north subtropical and warm temperate regions, which has mild climate, sufficient sunshine and abundant rainfall, and suitable for agriculture, forestry, animal husbandry and fishery. Because of discharge of the industrial effluents, the agricultural effluent and natural origin, the agricultural soil was contaminated by Cu, Zn, Cd, Pd etc. Six sampling sites were selected in the farmland, and approximately 5 kg soil was collected from 0 to 20 cm depth of soil surface layer at each site. All the collected soil samples were mixed thoroughly then air dried at ambient temperature, large particles and biological debris were removed, subsequently, sieved through a nylon mesh (2 mm).

Red mud is the secondary waste product from the alkaline lixiviating of monohydrallite in the Bayer Processing. All red mud used in this work was obtained from Chalco Zhongzhou Branch, Henan, China. It was sieved through 100 mesh sieve, washed, and air dried as received. Compost was obtained from rice straw, and the methods of operation were described in previous works (Zeng et al., 2011, 2015; Zhang et al., 2016). R + C was product of the red mud and compost after mixing at the proportion of 1:1 (w/w). Fig. 2 shows the chemical characteristics of these materials used in this study.

2.2. The design and procedure of experiments

Fig. 3 presents the whole process of the method. Soil was mixed with the compost materials thoroughly in the following ratio: (S) each pot with 500 g soil only; (S + C) each pot with 500 g soil and 25 g compost; (S + R) each pot with 500 g soil and 25 g red mud; and (S + B + C) each pot with 500 g soil, 25 g R + C. Finally, the mixed soil was transferred into pots with 1000 ml capacity. Each treatment was carried out in triplicate.

Soil pore water was extracted by attaching 30 ml plastic syringes to each samples by the method described in previous work (Beesley et al., 2014). The operation of this method was conducted as follows: firstly, non-ion water was sprayed to the soil of each pot to reach about 60% water holding capacity of the soil. Secondly, all of the pots were putted in an artificial climate chamber for 60 days under 30% relative humidity and 25 °C in order to reach about 60% the water holding capacity. The soil moisture in each pot was adjusted on a weekly basis. Thirdly, the pore water was acquired by

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