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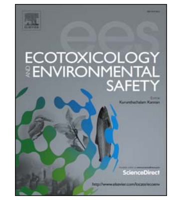

Potential use of lime combined with additives on (im)mobilization and phytoavailability of heavy metals from Pb/Zn smelter contaminated soils

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ABSTRACT

This explorative study was aimed to assess the efficiency of lime alone and in combined with additives to immobilize Pb, Cd, Cu and Zn in soil and reduce their phytoavailability for plant. A greenhouse pot experiment was performed by using low and heavily contaminated top soils viz. Tongguan contaminated (TG-C); Fengxian heavily contaminated (FX-HC) and Fengxian low contaminated (FX-LC). The contaminated soils were treated with lime (L) alone and in combined with Ca-bentonite (CB), Tobacco biochar (TB) and Zeolite (Z) at 1% and cultivated by Chinese cabbage (*Brassica campestris* L). Results revealed that all amendments ($p < 0.05$) significantly reduced the DTPA-extractable Pb 97.33, Cd 68.06 and Cu 91.11% with L + TB, L + CB, L + Z in FX-LC soil and Zn 87.12% respectively, with L + CB into TG-C soil. Consequently, the application of lime alone and in combined with additives were drastically decreased the dry biomass yield of *Brassica campestris* L. as compared with control. Thus, these feasible amendments potentially maximum reduced the uptake by plant shoots upto Pb 53.47 and Zn 67.93% with L + Z and L + TB in FX-LC soil, while Cd 68.58 and Cu 60.29% with L + TB, L + CB in TG-C soil but Cu uptake in plant shoot was observed 27.26% and 30.17% amended with L + TB and L + Z in FX-HC and FX-LC soils. On the other hand, these amendments were effectively reduced the potentially toxic metals (PTMs) in roots upto Pb77.77% L alone in FX-HC, Cd 96.76% with L + TB in TG-C, while, Cu 66.70 and Zn 60.18% with L + Z in FX-LC. Meanwhile, all amendments were responsible for increasing soil pH and CEC but decreased soils EC level. Based on this result, these feasible soil amendments were recommended for long term-study under field condition to see the response of another hyper accumulator crop.

1. Introduction

Soil contamination with potentially toxic metals (PTMs) had been a worldwide challenge for food security and health hazardous (Srinivasarao et al., 2014; Mahar et al., 2016; Ali et al., 2017a, 2017c). It is also a serious problem in China, and has become gradually more severe with the development of mine exploration, metallurgy industry, solid waste disposal, paint pigments, and irrigation of wastewater. Subsequently, due to the metal/gold mining and smelting both are important sources of trace metals to the environment (Lee et al., 2014; Shen et al., 2017). Mine soils usually have high metal concentration, even several years after the mine has been abandoned (Forján et al., 2016). Approximately, one-sixth of the total farmland area in China has been polluted with (PTMs) including Cd, Pb, Cu and Zn (Liu et al., 2005; Zhuang et al., 2009; Ali et al., 2017d). Thus, the accumulation of these PTMs can resulted to decrease of soil fertility, soil microbial activities and biodiversity, and collectively responsible for crop losses, and even has become more serious implications for both animal and human health (Pérez-Esteban et al., 2014). Moreover, these soils often display less fertility conditions, including degraded physical structure, unbalanced texture, extreme pH values, low cation exchange capacity (CEC), low OM and nutrient contents, which limit the establishment of vegetation and intensify erosion by rain and wind (Asensio et al., 2013). However, various remediation methods have been developed for the

List of abbreviation: TG-C, Tongguan contaminated soil; FX-HC, Fengxian heavily contaminated soil; FX-LC, Fengxian low contaminated soil; PTMs, Potentially toxic metals, L, Lime; CB, Calcium bentonite; TB, Tobacco biochar; Z, Zeolite; DTPA, Diethylenetriamine pentaaceticacid

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treatment of contaminated soils, including land excavation, soil washing, phytoremediation, and stabilization to reduce PTMs bioavailability (Fellet et al., 2011; Beesley et al., 2014). But most of the current technologies for soil remediation are time consuming or too expensive. Therefore, it is imperative to develop techniques that can treat and stabilize contaminants in situ in an efficient and cost effective manner (Wenzel et al., 1999). To overcome this globally problem, soil dilution or turnover has also been practiced as an alternative long-term approach as well as robust promising technology to reduce the concentration of PTMs from polluted sites (Hseu et al., 2010; Ahmad et al., 2012). Consequently, in this regards soil dilution technique and stabilization with additives are considered as an attractive option for remediation of metal mining or smelter contaminated soils. Since, last few years several amendments have been employed for the immobilization of metal-laden contaminated soils (Gonzalez-Alcaraz et al., 2013; Bolan et al., 2014). However, there is an increasing demand for novel, large scale applicable, and effectiveness of potential soil additives. While, factors manipulating for HMs immobilization mostly including adsorption, cation exchange, precipitation, pH, temperature and surface complexation (Jung et al., 2011; Kim et al., 2012). Whereas, the precipitation and co-precipitation become as salts can also contribute to reduce the mobility of contaminants into the soils (Kumpiene et al., 2008). Furthermore, soil amendments should have a high sorption capacity; they should be abundant, available, biodegradable, and cheap in order to use them as economically feasible as amendments for remediation of PTMs contaminated sites (Mahar et al., 2015). However, using excess amount of alkaline as soil amendments can make the soil alkaline and compacted, further decreasing agricultural productivity. In some cases, an amendment might be effective at fixation one pollutant but may enhance the mobility of another metal (Hartley and Lepp, 2008). Hydrated lime ($Ca(OH)_2$) is an oldest and most widely employed as a soil PTMs immobilizing agent (Chlopeckaand Adriano, 1996). Because, the addition of lime to PTMs polluted soils is a well-known strategy to increase soil pH, trigger precipitation of metal carbonates, oxides or hydroxides and decrease metals solubility (Castaldi et al., 2005b). But, natural zeolite is a type of porous alumino-silicates mineral characterized by negative charges, which can potentially able to reduce the Pb, Cd and Zn bioavailability (Castaldi et al., 2004, 2005a). Also, bentonites are argillaceous materials that can be effectively employed as adsorbents for many wastewater pollutants, namely PTMs ions and organic compounds due to high surface area when they are hydrated. Furthermore, it has strong colloidal properties and its volume increases several times when coming into contact with water, forming a gelatinous and viscous liquid. The special properties of bentonite (hydration, swelling, water absorption, viscosity and thixotropy) render it as a valuable material for a wide range of uses and applications (Dimirkou et al., 2002). Finally, biochar (BC) is a solid and carbon-rich by-product similar to activated carbon, obtained from synthetic conversion of biological material under minimal oxygen supply, which is attracting more and more attentions during recent years, due to its outstanding abilities for the immobilization of PTMs from soil or aqueous systems (Li et al., 2015; Park et al., 2016). Subsequently, its porous structure, active functional groups and normally high pH and CEC were also support the immobilization process (Park et al., 2011). Nonetheless, highly effective and low cost practices are very much needed to solve this problem. One such method that is obtaining interest is the use of BCs that can absorb PTMs and decrease their bioavailability to plants and prevent uptake and food chain transfer (Puga et al., 2015). However, last few year numbers of scientific papers are published on lime alone applied as soil amendment (Gray et al., 2006; Lee et al., 2009; Mallampati et al., 2012) or combined with other inorganic additives e.g, limestone + sepiolite, for soil remediation (Wu et al., 2016). Besides, Zhou et al. (2014b) used limestone + sepiolite and hydroxyhistidine + zeolite for stabilization of multi-metals polluted soil. More recently, (He et al., 2017) have applied lime, slag, and bagasse alone or in combined as amendment for remediation of Cd-contaminate paddy soil, but upto date no attempt has been made to study the efficiency of lime combined with Ca-bentonite (CB), biochar, and Zeolite (Z) on the immobilization and its underlying mechanism for phytoavailability of PTMs from contaminated soil by conducting pot experiment. While, Chinese cabbage (*Brassica campestris* L.) is a nutritious leafy vegetable, commonly grown in many parts of China and have much economic value.

Therefore, the aimed of this study was 1) to assess the efficacy of emerging and low cost alternative mixture of additives for immobilization of Cd, Pb, Cu and Zn and their uptake by Chinese cabbage (*Brassica campestris* L.) from Pb/Zn smelter contaminated soils. Chinese cabbage was chosen as an indicator plant because it has highly potential to resist higher concentration of PTMs, higher biomass and short duration of plant; 2) to quantify the effectiveness of immobilizing agents on soil chemical properties.

2. Materials and methods

2.1. Soil collection and characterization

Three different soils samples were collected from Tongguan (TG), (34°29.584"N , E 11018.271"E), Fengxian (FX) (36° 56' 54.36"N, 106° 31' 29.41"E) counties of Shaanxi Province, China. While, TG soil was collected from artisanal gold mining region of Tong County mainly contaminated with Pb and Cu and mentioned as Tongguan contaminated (TG-C). Actually, FX soil was collected from contaminated with Cd and Zn mainly due to a Zinc smelter, while this soil collection was divided in two sub-category such as one heavily contaminated Fengxian soil (FX-HC), which close to the smelter and one low contaminated Fengxian soil (FX-LC) was consider, which far from the smelter. All the soil samples in bulk were collected from surface layer (0–20 cm depth), then stored in polyethylene bags and all sub-samples were bulked together in the laboratory to obtain an “average” sample. Soils were homogenized, air-dried, and manually crushed. Samples were ground to pass through, 2 mm sieve and analyzed for their basic physico-chemical properties according to Ding et al. (2013). Subsequently, sieved samples were stored at 4 °C prior to use and analyzed physico-chemical characteristics of studied soils (Table 1).

2.2. Analysis and characteristics of soil amendments

Hydrated lime as ($Ca(OH)_2$), was purchased from local market of Yangling City, Shaanxi Province, China. The Ca-bentonite (CB) was purchased from Weifang Huawei Bentonite Group Co. Ltd., China, in a Ca-based form. Tobacco biochar (TB) was derived from stalk residues by the pyrolysis process at 500 °C in Yixin Biological Energy Science and Technology Development Co., Ltd., Shaanxi, China. While, the natural zeolite was purchased from Zhejiang Shenshi Mining Industry Group Co., Ltd., China, which has a diameter of 3.5–4.0 (Ai) and CEC of 120–160 meq/100 g. The pH of additives was measured in deionized water (DI) 1:5 H_2O for inorganic materials and 1:10 H_2O organic materials, by using USEPA Method 9045D and ASTM D1125, respectively. Total PTMs concentration of additives were determined by using acid digestion according to USEPA Method 3051A and analyzed by AAS (Z-2000, Japan).

2.3. Experimental set-up

The impact of additives on immobilization and phyto-availability of Pb, Cd, Cu and Zn to the *Brassica campestris* was investigated in a greenhouse using the Pb/Zn smelter contaminated soils. The studied additives viz. L alone and in combined with CB, TB and Z were applied at 1% (dry weight basis), while prior to use as a soil amendment among the all soil additives were homogenized and ground to pass through a sieve of 2 mm. In a portion of 3.0 kg of each air-dried samples, were added 30.0 g of each additives, which were placed into 20 cm-diameter