



Research article

Immobilization and reduction of bioavailability of lead in shooting range soil through hydrothermal treatment



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ABSTRACT

Immobilization of Pb in contaminated soil by hydrothermal treatment (HT) under subcritical conditions was investigated using a lab-scale apparatus. The Pb immobilization was evaluated through investigating the transformation of Pb fractions and by single chemical extraction. The results showed that HT and treatment temperature significantly affected the immobilization through redistribution of Pb fractions. The results of bioavailability and eco-toxicity assessment demonstrated a drastic decrease in the bioavailable fraction from 41.33 to 14.66%, and an increase in the non-bioavailable fraction from 2.90 to 15.76%. Moreover, the leaching potential of Pb was significantly reduced after treatment. Based on the risk assessment code (RAC), the treated soil residues exhibited medium risk (21.7–14.6 of RAC value for treated soil), which represented a decrease from high risk (41.3 for untreated soil). Immobilization was associated with Pb fractionation from weakly bound to stable fractions. A variety of mechanisms including adsorption and precipitation were suspected to be responsible for Pb immobilization. The findings of this study suggest that the HT may facilitate quantitative reduction of the bioavailability and eco-toxicity of Pb-contaminated soil. Considering the effectiveness of HT in remediating contaminated soil with organic pollutants, this approach might enable remediation of soil co-contaminated with organics and heavy metals.

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1. Introduction

Heavy metal contamination, in particular Pb, in soil is a serious problem due to its potential threats to human, especially at army shooting ranges, as Pb is a hazardous environmental pollutant. Moon et al. (2013) reported that approximately 1400 active small arms firing ranges can be found in the Republic of Korea and more than 3000 in USA. Shooting ranges around the world are generally considered for a significant sink and potential source for toxic trace metal pollution in soil and groundwater (Sanderson et al., 2014). Due to lead bullets and their fragments, Pb is the dominant metal presence in shooting range soils (Cao et al., 2003). However, many sites could be co-contaminated with organic hazardous compounds and heavy metals, in particular army shooting range soils. The use of organic compounds, such as petroleum products and explosives, in military operations has often caused for soil co-contamination.

The stabilization/solidification (S/S) process is commonly used to immobilize Pb-contaminated soils (Aydın and Aydın, 2014; Chen

et al., 2007; Houben et al., 2012; Ok et al., 2010). Although various S/S agents, such as chemicals, lime materials, fly ash, cement and cement kiln dust have been successfully employed to immobilize Pb in contaminated soils (Cao et al., 2009; Houben et al., 2012; Ok et al., 2011; Sanderson et al., 2015), their ability to eliminate organic pollutants from soil is limited. Therefore, the development of a single remediation technique that allows remediation of co-contaminated soil by extracting/degrading organic pollutants together with stabilization of Pb is of great interest. It has already been proved that the hydrothermal treatment (HT) process (e.g., vapor extraction, sub-/supercritical water extraction/oxidation) is very effective for extraction or degradation of various organic pollutants in contaminated soils (Chang et al., 2011; Islam et al., 2012, 2014a,b).

In recent years, sub-/supercritical water technology had also become a promising approach to stabilize the heavy metals in various sludge residues during their liquefaction (Chen et al., 2014; Huang et al., 2011; Islam et al., 2015; Leng et al., 2014; Li et al., 2012; Shi et al., 2013a,b; Yuan et al., 2015; Zhai et al., 2014) and in medical waste incinerator fly ash (Bo et al., 2009; Jin et al., 2010). Most studies reported that the most available fractions of Pb were

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transformed mainly into relatively stable fractions during HT, resulting in decreasing its direct toxicity and reducing the environmental risk. However, to date, stabilization of heavy metals in contaminated soil by sub-/supercritical HT process has not been investigated; therefore, the feasibility of remediating inorganic contaminated soil, by reducing Pb bioavailability and eco-toxicity is unknown. It would be worthwhile to study the effectiveness of subcritical HT for heavy metal immobilization in contaminated soils, due to its advantages of high extractability and degradability of organics. The objective of this study was to evaluate the Pb immobilization in shooting range soil using HT under subcritical condition.

After weathering, shooting range soil is composed mostly of lead-carbonate related minerals (Yin et al., 2010). Chen et al. (2008) reported that the exchangeable and carbonate fractions are likely to be affected by the change of ionic composition in water. It is well known that the concentrations of ionic products of water molecules are high under subcritical conditions ($100 \leq T \leq 374$ °C and <22.1 MPa) (Kuhlmann et al., 1994; Oh et al., 2011). Accordingly, it is hypothesized that increased concentration of OH⁻ ions could be a factor in Pb immobilization via adsorption process by increasing negatively charged sites on soil particles (Ahmad et al., 2012a; Ok et al., 2011). In addition, organic fraction can be degraded by subcritical water, leading to the release of soluble Pb and thus formation of PbO under oxidizing conditions which can be precipitated on soil particles. Similar finding was reported by Song et al. (2013) and Devi and Saroha (2014) that the formation of insoluble PbO was occurred during high-temperature HT of paper mill sludge, resulting in the immobilization of Pb in the sludge. These observations suggest that HT may immobilize Pb in contaminated soil.

2. Materials and methods

2.1. Soil sample and analysis

Soil samples were collected from the surface (0–30 cm) at a shooting range site, located close to Seoul, Republic of Korea, which was heavily contaminated with Pb. Several samples from different sampling points were mixed together, homogenized and sieved to <2 mm. The soil sample was characterized for pH, electric conductivity (EC), soil organic matter (SOM), cation exchange capacity (CEC), distribution of particle size and sequential and total Pb content. Detailed soil sampling methodology and physicochemical analytical procedure were reported previously (Islam et al., 2016). The mineralogical composition of the soil sample was determined by X-ray diffraction (SC-XRD, AXS Smart APEX II, Bruker) and the elemental composition and dot map of the soil samples were carried out by scanning electron microscopy (FE-SEM, JSM-7500F, Jeol Inc.) equipped with an energy dispersive X-ray spectroscope (ISIS 310 EDX, Oxford Inst.).

For sequential extraction of Pb, the five fractions, including readily available and exchangeable Pb (F1), carbonate/specifically adsorbed fraction (F2), Pb bound to Fe and Mn oxides (F3), organic and sulfide fraction (F4), and Pb in residue (F5) were measured following the Tessier method (Tessier et al., 1979). The residual and total content of Pb in soil samples were subjected to acid digestion with aqua-regia [1 mL of HNO₃ (70%) and 3 mL of HCl (35%)]. In all cases, the extracted solution was filtered through a 0.45 µm filter, and the concentration of Pb metal was determined by an inductively coupled plasma optical emission spectrometer (ICP-OES, Optima7300 DV, Perkin Elmer). Each test was performed in triplicate and the average values were presented.

2.2. HT procedure

The HT experiments were carried out in a 2 L enclosed stainless steel reactor at 220 and 270 °C under subcritical conditions to maintain the water in a liquid state. In each test, 500 g air-dried sieved soil were mixed with 1.5 L of tap water in the reactor and heated to the set temperature for 1 h. After HT, the soil-water mixture was cooled to ambient temperature, and then soil residue and liquid samples were collected after separation. The soil residue was air-dried and stored for further testing. The soil residues from treatment at temperatures of 220 and 270 °C were assigned the codes SR220 and SR270, respectively. Finally, the untreated (directly using the shooting range soil) and treated soil residues were subjected to a leaching and fractionation test. The aqueous samples were also analyzed for determination of Pb concentration.

2.3. Leaching experiment

To examine Pb immobilization, the Korean standard leaching procedure was adopted to assess the leaching rate of Pb under acidic conditions; this was used widely for testing the toxicity characteristics of heavy metal-contaminated soil (Lim et al., 2013; Moon et al., 2011; Ok et al., 2011). Pb in untreated soil and treated soil residues was extracted with 0.1 N HCl solution at a solid/liquid ratio of 1:5 (g:mL) and shaking for 1 h at 30 °C. To evaluate the reduction of the most available Pb fraction in soils, extraction with 1 M MgCl₂ at a 1:10 soil:solution ratio was performed. All suspensions were filtered through a 0.45 µm filter and the concentration of Pb was determined by ICP-OES.

2.4. Bioavailability and eco-toxicity assessment

The immobilization of Pb in soil residue was also evaluated by the changes of five fractions. These fractions may be considered to decrease in lability from fraction F1 to F5 and they can further be three grouped based on their stability as bioavailable (Cbio: F1+F2, easily releasable), potentially bioavailable (Cp-bio: F3+F4, relatively stable) and non-bioavailable (Cn-bio: F5, stable) fraction. These three parts reflect the eco-toxicity in the environment as direct toxicity, potential toxicity, and no toxicity, respectively (Li et al., 2012; Shi et al., 2013a; Vela et al., 1993). Because the risk due to heavy metals in soil depends strongly on the weakly bound fractions (F1+F2), a risk assessment code (RAC) was used for risk evaluation of Pb before and after HT, following the other studies (Perin et al., 1985; Singh et al., 2005; Yuan et al., 2015). RAC was proposed to assess the bioavailability of Pb in soil residue based on the proportion of the first two fractions (F1+F2) of Tessier's method to the total concentration. When the RAC value (%) is < 1, 1–10, 11–30, 31–50, and >50, generally risk are assigned as no risk, low risk, medium risk, high risk, and very high risk, respectively (Leng et al., 2014; Perin et al., 1985; Singh et al., 2005).

3. Results and discussions

3.1. Characterization of soil sample

The selected properties of the contaminated soil are presented in Table 1. The soil was sandy loam with a pH value of 6.1. The SOM and CEC of the soil sample were 7.8 (wt.%) and 44 cmol_c/kg, respectively. The soil had the typical exchangeable Ca content (5.30 cmol_c/kg) of Korean upland soil (Ahmad et al., 2012b). The Pb concentration (3436 mg/kg) in soil was about fivefold higher than the Korean standard level for shooting site (700 mg/kg) (MOE, 2010). The mineralogical composition of shooting range soil

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