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Stability of heavy metals in soil washing residue with and without biochar addition under accelerated ageing

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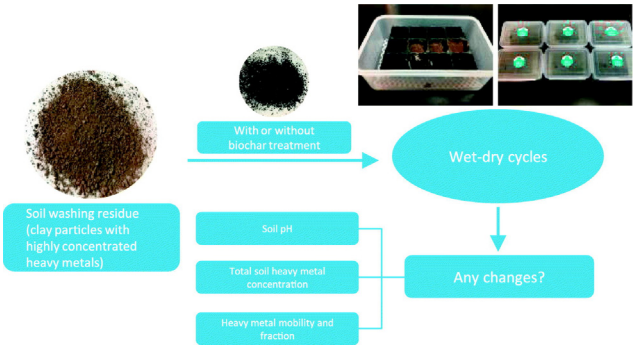
HIGHLIGHTS

- Cd^{2+} and Pb^{2+} in soil washing residue (SWR) exceeds TCLP regulation limit.
- Heavy metal mobility in SWR was reduced by biochar addition.
- Accelerated ageing mobilised heavy metals in SWR regardless of biochar addition.
- Accelerated ageing show SWR poses long-term risks to the environment.

GRAPHICAL ABSTRACT



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ABSTRACT

Soil washing residue (SWR), which typically concentrates the washed toxic metals and is comprised of high contents of clay particles, may pose risks to the surrounding environment. This study aims to simulate accelerated ageing to assess the stability of selected metals (Cd^{2+} (132 mg/kg), Cu^{2+} (248 mg/kg) and Pb^{2+} (3470 mg/kg)) in a SWR (89.68% of clay) with and without biochar treatment. The soil was incubated under constant moisture and wet-dry cycles (accelerated ageing), respectively, and the mobility and fractions of heavy metals in the soils with and without biochar treatment were examined. Under the constant moisture condition, biochar addition at 5% w/w reduced the leached Cd^{2+} (by 1.81%) and Cu^{2+} (by 8.70%) from SWR at day 1 and the leached Cu^{2+} (by 51.08%) and Pb^{2+} (by 25.36%) from SWR at day 14; however, the leached metals in the TCLP solution from the biochar-amended soils still exceed the regulatory limits (1 mg/L for Cd^{2+}, 5 mg/L for Pb^{2+}, no regulatory limits for Cu^{2+}). Conversely, accelerated ageing (14 days) significantly increased the fractions of exchangeable Cd^{2+} (from 3.63–3.94% to 6.21–6.29%) and Pb^{2+} (from 0.025–0.027% to 0.034–0.041%) as well as the TCLP leachabilities of Cd^{2+} (from 2.91–3.28% to 3.46–3.73%), Cu^{2+} (from 0.08–0.10% to 0.03–0.06%) and Pb^{2+} (from 0.25–0.35% to 0.52–0.57%) in the soils, as compared with those incubated under constant moisture, regardless of biochar addition. This study reveals challenges associated with stabilising SWR due to the presence of residual fine-grained particles.

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1. Introduction

Various contaminants, including toxic metals, are released into soils due to human activities such as oil drilling, mining, chemical processing and waste disposal (Guemiza et al., 2017; Hou and Al-Tabbaa, 2014). The contaminants, once accumulated in soil, can cause serious ecosystem damage and have the potential to cause human health problems. For instance, heavy metals can accumulate in internal organs and cause chronic damage to the body (Gavrilescu, 2004).

Soil contamination by toxic metals is a major environmental problem worldwide. China is the most recent country to conduct a national soil survey, and according to the results that were released by the Chinese government, 19.4% of tested agricultural land and 34.9% of tested former industrial land is contaminated (Qu et al., 2016; Zhao et al., 2015; Hou and Li, 2017).

Soil remediation aims to control, modify or destroy pollutants that present unacceptable ecosystem or health risk (Hou and Al-Tabbaa, 2014). Chinese researchers are exploring innovative remediation approaches in a variety of ways (Ma et al., 2015; Ma et al., 2014; Ma et al., 2016). Solidification/stabilisation (S/S) is among the most widely-used conventional technologies in soil remediation (Hou et al., 2016a), which aims to immobilise contaminants on-site or for landfill disposal (Wang et al., 2016). However, contaminants are not removed from the contaminated site after S/S treatment, which results in uncertainty about the long-term stability of contaminants at the site. Soil washing is one of the few permanent treatments to separate metals and organics from soils, which could result in a thorough clean-up of the contaminated site (Dermont et al., 2008; Zhang et al., 2010; Hou et al., 2014a).

Soil washing uses (1) physical separation, (2) chemical extraction or (3) a combination of both to remove contaminants from soil. Physical separation is more commonly used and is far more cost effective ($ 70–187 m^{-3}) than is chemical extraction ($ 358–1717 m^{-3}) (Dermont et al., 2008). S/S is typically regarded as a cost-effective way to treat contaminated site. The costs for ex-situ S/S and in-situ S/S are $ 90–245 m^{-3} and $ 50–330 m^{-3} respectively according to US Environmental Protection Agency (USEPA, 2010). Therefore, the cost of soil washing using physical separation is comparable to or even less than S/S, which makes it feasible to be applied in large-scale in developing countries such as China regardless of several limitations such as 1) that it is preferable for sandy soil rather than clay and 2) that it may generate secondary pollution during operation. The reuse of contaminated soil treated by soil washing can promote green and sustainable remediation (Hou et al., 2015), which is a relatively new concept that has drawn great attention in the remediation field (Hou, 2016; Hou et al., 2014b, 2014c; Hou et al., 2016a, 2016b). In the case of metals remediation, physical separation uses a range of technologies (e.g. mechanical screening, hydrodynamic classification, gravity concentration, froth flotation, magnetic separation, electrostatic separation, and attrition scrubbing) to remove weakly bonded heavy metals and separate from the soil the relatively coarse particles that have been cleaned (Dermont et al., 2008). The cleaned coarse particles (typically in excess of 50–70%) can then be returned to the site; however, this process also enriches metals into the remaining fine fraction of the soil (predominantly clay). These clay particles, enriched with heavy metals, may be unsafe if exposed to the environment. The heavy metals in the soil are retained on clay mainly through cation exchange (Shen et al., 2016a), and so these exchangeable heavy metals are bioavailable and potentially can be desorbed to the environment under field conditions (Filgueiras et al., 2002).

This introduces a problem of the soil washing technology which is similar to that of S/S: that the long-term stability of the residual soil (defined as soil washing residue (SWR) hereafter) that contains elevated metal concentrations relative to the bulk soil is uncertain (Isoyama and Wada, 2007). If the SWR is safe, it could be reused on site. If it still poses environmental risks, it should be disposed of in a hazardous waste landfill directly or after S/S treatment, until it meets regulatory requirements. It is therefore important to assess the long-term stability of heavy metals in SWR. However, such efforts are very limited. For instance, Ko et al. (2005) assessed the fractions of As, Zn^{2+} and Ni^{2+} in soil after pilot-scale acid washing using the Community Bureau of Reference three-step sequential extraction, and observed that acid washing significantly reduced the exchangeable, Fe/Mn oxide and organic/sulfides fractions of As, Zn^{2+} and Ni^{2+} in the coarse particles (0.25–0.42 mm and 0.841–10 mm), whereas the influence on fine particles was insignificant. This indicates the distribution of the original heavy metals bound to fine particles was little changed during soil washing. However, the mobility and fractions of the metals in SWR, when exposed to the environment over longer terms, remains unclear.

Based on this evidence, the stability of heavy metals (Cd^{2+}, Cu^{2+} and Pb^{2+}) in a SWR was assessed. The SWR underwent accelerated ageing under wet-dry cycles and the mobility and fractions of heavy metals in the soils were investigated. Biochar was mixed with the SWR as a S/S treatment and its potential impact on the long-term stability of heavy metals in the SWR was additionally investigated, with and without accelerated ageing. Biochar was selected for S/S because it is a promising remedial material that has potential to be applied in large-scale S/S and soil mixing treatments, based on the results of the authors' previous work (Shen et al., 2015; Shen et al., 2016b). Biochar maybe made from various biological waste materials (Dennehy et al., 2017; Feng and Zhu, 2018; Lin et al., 2017). Its usage can result in substantial sustainability gains (Hou and Al-Tabbaa, 2014; Hou et al., 2017a, 2017b; Song et al., 2018). This study aims to assess the long-term stability of heavy metals in SWR, with and without biochar treatment, using accelerated ageing.

2. Materials and methods

2.1. Soil and biochar

The SWR used is from a contaminated site in Guangzhou, China that belonged to one of the biggest steelmaking companies in China. A site investigation showed that soil contamination on this site covered an area of 158,000 m^2 and approximately 520,000 m^3 of soil was contaminated. A range of contaminants were detected in the soil including heavy metals: Cd, Hg, As, Pb, Cr, Cu, Zn, Ni, Se, Co, V, Sb (≤ 54,100 mg/kg), polycyclic aromatic hydrocarbons (≤4190 mg/kg) and petroleum hydrocarbons (≤44,000 mg/kg). A systematic treatment was employed based on soil washing and thermal desorption to treat the contaminated soils in 2015–2016. Briefly, the contaminated soils were washed and the coarse particles (≥0.075 um) were separated and returned to the site or reused for other purpose (after being tested and compared to regulatory limits). When necessary, the relatively fine particles (≤0.075 um) (concentrated with contaminants after soil washing) were also treated by thermal desorption to remove organic contaminants. Then the SWR was collected for the present study.

The SWR was oven dried at 40 °C for 48 h and ground and sieved to smaller than 2 mm for the experiments. The SWR sample was analysed for particle size distribution, pH and organic matter and heavy metal contents. The SWR was pretreated in a 20 g/L sodium hexametaphosphate solution in an ultrasonic cleaner for 5 min and its particle size distribution was analysed using a laser particle size analyser (mastor2000, Malvern Instruments Ltd., UK) in wet mode. Soil pH was tested using a solid to liquid ratio of 1:20 g/mL, and soil total organic content (TOC) was tested using a TOC analyser (TOC-VCPH, Shimadzu, Japan). The heavy metal concentrations in the SWR were obtained by the sum of each fraction in sequential extraction as per Section 2.4. The results of these tested properties of SWR are shown in Table 1. The SWR was alkaline with a pH value of 9.14. Clay and silt comprise 89.68% and 10.32% based on particle size analysis, respectively, of the solids in the SWR, indicating that in-situ separation was successful as no coarse particles were mixed in the SWR. Cd^{2+}, Pb^{2+} and Cu^{2+} exceeded regulatory limits and were the primary