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Removal of heavy metals and arsenic from a co-contaminated soil by sieving combined with washing process

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ABSTRACT

Batch experiments were conducted with a heavy metals and arsenic co-contaminated soil from an abandoned mine to evaluate the feasibility of a remediation technology that combines sieving with soil washing. Leaching of the arsenic and heavy metals from the different particle size fractions was found to decrease in the order: <0.1, 2–0.1, and >2 mm. With increased contact time, the concentration of heavy metals in the leachate was significantly decreased for small particles, probably because of adsorption by the clay soil component. For the different particle sizes, the removal efficiencies for Pb and Cd were 75%–87%, and 61%–77% for Zn and Cu, although the extent of removal was decreased for As and Cr at <45%. The highest efficiency by washing for Pb, Cd, Zn, and As was from the soil particles >2 mm, although good metal removal efficiencies were also achieved in the small particle size fractions. Through SEM-EDS observations and correlation analysis, the leaching regularity of the heavy metals and arsenic was found to be closely related to Fe, Mn, and Ca contents of the soil fractions. The remediation of heavy metal-contaminated soil by sieving combined with soil washing was proven to be efficient, and practical remediation parameters were also recommended.

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Introduction

In recent decades, the intensification of human activities such as mining, smelting, electroplating, and coal combustion has resulted in a broad range of new environmental problems. Among these has been the widespread generation of enormous quantities of complex, heavy metal-contaminated soils, which constitute a major health challenge of global concern (Boulabaha et al., 2006). Many heavy metal-contaminated sites have been identified (Dermont et al., 2008). In the United States, heavy metals are prevalent at 77% of the Superfund sites, 72% of the Department of Defense sites, and 55% of the

Department of Energy sites, and over 50 million m³ of heavy metal-contaminated soil exist at current National Physical Laboratory sites (USEPA, 2004). In Europe, heavy metal-contaminated soils encompass several million hectares, accounting for about 37.3% of the total contaminated soil (EEA, 2014). Similar trends are also found in China, where 16.1% of the soil has been proven to be contaminated, primarily by heavy metals, and in regions characterized by heavily polluting enterprises, industrial wastelands, industrial parks, and mining areas, the contamination levels can be as high as 30% (Ministry of Environmental Protection and the Ministry of Land, 2014).

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The mobility and bio-availability of metals are significantly affected by the particle's size and composition of the soil. Fine soil fractions can adsorb more contaminants than coarse sand because of their larger specific surface areas. In general, elemental adsorption (particularly heavy metals) increases with decreasing aggregate size (Zhang et al., 2003). Anderson et al. (1999) pointed out that contaminants were concentrated in the finer (0.05 mm or less) silt and clay fractions. In addition, metal accumulation in the clay fraction is attributed to the presence of clay, organic matter, minerals, and Fe/Mn oxides in the micro-aggregates, which act as delivery vehicles for metals (Rodriguez-Rubio et al., 2003; Quenea et al., 2009). As a result, heavy metals and arsenic in clays are difficult to remove due to their strong adsorption. Cheng et al. (2008) indicated that the concentration of Cu in clays was as high as 670.6 mg/kg, 4.77 times the concentration achieved in gravel. Gong et al. (2013) drew a similar conclusion in a soil survey in Hainan, wherein the smallest fraction (<0.053 mm) had the highest heavy metal concentrations.

Soil washing is one of the few treatment alternatives for the elimination of heavy metals from contaminated soils, which can be applied to pilot/full-scale field remediation (Dermont et al., 2008). The advantages of the technology are: high adaptability, a wide range for effective concentration of the contaminants, simple operation, strong regulation, short repair cycle period, lower cost, wide applicability, and huge practical value. Juwarkar et al. (2007) used 0.1 mol rhamnolipids to wash Cd-contaminated soil and obtained 91% Cd removal at pH = 10. Torres et al. (2012) demonstrated that Cd, Zn, and Cu could be washed with efficiencies up to 85.9%, 85.4%, and 81.5%, respectively. Maity et al. (2013) proved that soil washing can achieve high efficiencies, with the removal of 98% Pb, 95% Cu, and 56% Zn.

Physical separation technologies are primarily applied in the mining and mineral processing industries to separate particulate forms of metals. These technologies include sieving, centrifugation, flocculation, hydrocyclone classification, magnetic separation, and flotation. As these technologies concentrate contaminants into smaller volumes, they have been applied in the field of soil remediation in recent years (Dermont et al., 2010; FRTR, 2007). Lin et al. (2001) removed 84%–88% Pb from soil by jigging and density separation, and achieved the efficient removal of Pb by soil washing. Reynier et al. (2013) adopted soil washing combined with flotation to eliminate heavy metals in four soils, and removal efficiencies of 82%–93%, 30%–80%, and 79%–90% were obtained for As, Cr, and Cu, respectively.

Previous studies have shown that physical separation and soil washing can both result in the effective removal of heavy metals from soil, and even the combined technology has been attempted to remediate heavy metal-contaminated soil. Considering that soil washing is insensitive to the type of contaminant and suitable for sandy soils (Impellitteri et al., 2002), and that separation technology can reduce the volume of waste (Anderson et al., 1999), we plan to use sieving combined with soil washing to treat heavy metal-contaminated soil. The objectives of the research are to: (1) discover the occurrence, distribution characteristics, and influence of heavy metals in soil particles; (2) evaluate the removal efficiency and potential volume reduction of contaminants in the soil fractions; (3) probe the

mechanism of heavy metal adherence to soil particles during the process of soil washing.

1. Materials and methods

1.1. Soil samples and main physicochemical properties

Soil sample was collected from the 0–50 cm surface layer of a metallurgical industry disposal site (E113°04'24"; N27°52'38") located in Hunan Province, China. The sample was air-dried and homogenized at room temperature in the laboratory, and the coarse debris and visible plant materials were removed before further processing. The primary physical and chemical properties of the soil were as follows: pH 6.65; organic matter, 66.9 g/kg; total nitrogen, 0.13%; total phosphorus, 0.11%; Fe, 3.47%; Ca, 5.42 mg/kg; and cation exchange capacity (CEC), 15.7 cmol/kg.

1.2. Soil particle separation and metals distribution according to particle size

For the separation procedure, the air-dried sample (250 g) was weighed and dry-sieved. The soil was subjected to 3 × 10 min automatic shaking in a series of graded sieves positioned in a vibrating screen instrument (ZBSX-92A, Lei Yun Test Equipment Manufacturing Co., Ltd., China). The sieved material was divided into six graduated fraction sizes, >2, 2–1, 1–0.25, 0.25–0.1, 0.1–0.053, and <0.053 mm, which corresponded to 14.63%, 12.33%, 32.05%, 17.62%, 15.07%, and 8.30% of the total original sample. The distribution of the soil was determined by the weight of each size after sieving. The sample was classified as a sandy soil according to the particle size definition by the American Society for Testing and Materials (Fig. 1).

The concentrations of trace elements (including heavy metals, Ca, and S) were analyzed by inductively coupled plasma-optical emission spectrometry (ICP-OES, Perkin-Elmer Optima 5300DV) after complete sample (0.05 g) decomposition by mixed acid digestion (HF–HNO₃–HClO₄) (a modification of the GB-T 17141-1997 method, NSPRC (National Specifications of the People's Republic of China), 1997). Two reference standard soils

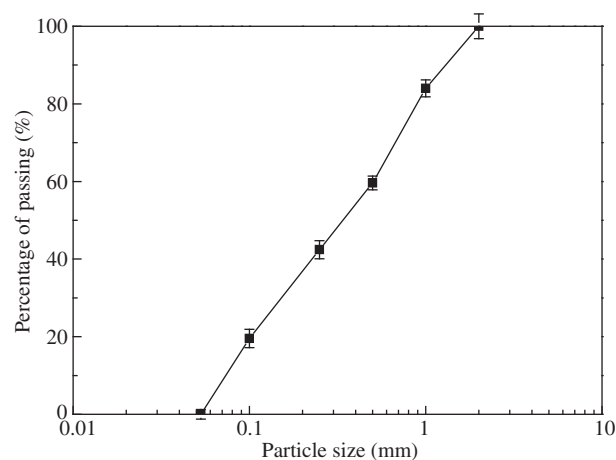


Fig. 1 – Particle-size distribution in the bulk soil sample.

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