



Toxicity characteristic leaching procedure over- or under-estimates leachability of lead in phosphate-amended contaminated soils



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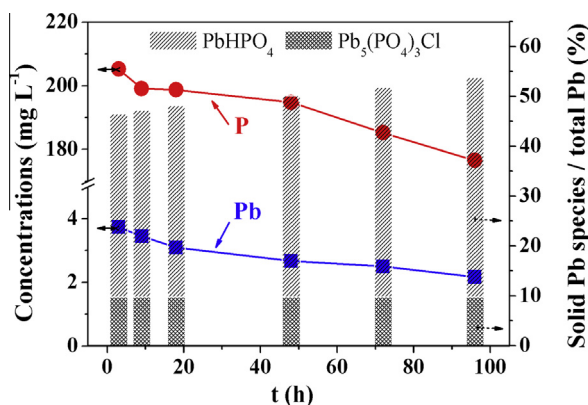
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HIGHLIGHTS

- Lead leachability does not reach equilibrium during the standard TCLP test.
- The acidic TCLP fluid induces more transformation of insoluble Pb–P precipitates.
- TCLP underestimates the leaching of soil Pb with less soluble PR amendment.
- TCLP overestimates the leaching of soil Pb with soluble TSP amendment.

GRAPHICAL ABSTRACT

The reaction process of $\text{Pb}(\text{NO}_3)_2$ -contaminated soil amended by TSP: the concentrations of Pb and P in TCLP leachate decreased but Pb–P precipitates increased as leaching time increased.



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ABSTRACT

In this study, $\text{Pb}(\text{NO}_3)_2$ -, PbSO_4 -, or PbCO_3 -contaminated soils were treated with triple super phosphate (TSP) or phosphate rock (PR) and then subjected to the toxicity characteristic leaching procedure (TCLP) to assess Pb leachability. Soluble TSP resulted in the transformation of Pb into insoluble Pb phosphate precipitates in all contaminated soils, and the transformation increased with extended leaching times. Consequently, Pb concentrations in the TCLP leachates treated with TSP were reduced by 97.3–99.7% compared with the untreated soils, and Pb leaching decreased over the extraction time and did not reach equilibrium even after 96 h of extraction. Precipitation of Pb phosphate minerals in the less soluble PR-treated soil was limited, and Pb leaching was controlled by the dissolution of the Pb compounds, resulting in elevation of Pb in the TCLP leachate. Pb leaching continued to increase with time due to continuous dissolution of PbSO_4 and PbCO_3 . The results indicated that Pb leaching is kinetically controlled by either Pb compound dissolution or phosphate mineral formation. The standard TCLP test using a designated 18 h incubation time can overestimate the leachability of Pb in soils contaminated with lead and amended with soluble TSP and underestimate the leachability of Pb in soils contaminated with Pb and amended with less soluble PR. Therefore, wide use of TCLP for assessing Pb leachability in all contaminated soils is insufficient, and development of a site-specific evaluation method is urgently needed.

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1. Introduction

The toxicity characteristic leaching procedure (TCLP), developed by the United States Environmental Protection Agency (USEPA), is designed to simulate the worst possible scenario for co-disposal of waste and municipal solid waste in a landfill (USEPA, 1992) and has been widely applied as a regulatory test to evaluate contamination levels (Mantis et al., 2005). The standard procedure recommends two types of leaching fluids: acetic acid solution ($\text{pH} = 2.88 \pm 0.05$) and acetate buffer solution ($\text{pH} = 4.92 \pm 0.05$) to simulate the presence of organic compounds in landfill leachates. However, the applicability of the TCLP for assessing the hazards posed by waste disposal in landfills has been challenged. Important factors, such as pH, redox potential, biological activity, retention time, and interactions with landfills with other compositions, are not considered in the TCLP leachates. The use of TCLP for predicting leaching may result in misclassification and lead to over or under estimates of heavy metal leachability in contaminated soils. Hooper et al. (1998) confirmed that the TCLP underestimated the concentrations of As oxyanion leaching when compared with real municipal solid waste (MSW) leachate and proposed that this was due to the complexation of As with organic compounds in the MSW leachate. Similar results were reported by Halim et al. (2004) and Ghosh et al. (2004), who argued that the TCLP did not simulate the pH-Eh conditions in landfills and demonstrated that the test underestimated As leaching from bearing solids and that equilibrium was not achieved within 18 h of standard exposure time. Cao and Dermatas (2008) investigated the leachability of Pb in shooting range soils using the TCLP and showed that Pb concentrations and solution pH values were not at equilibrium after the standard extraction time of 18 h due to continued dissolution of metallic Pb particles and the re-precipitation of Pb as cerussite. Rubinos and Barral (2013) reported that the standard TCLP largely underestimated the release of trace metals from bauxite red mud because of its high capacity for neutralizing acidity. Consequently, the acetic acid provided in the TCLP extraction fluid was quickly consumed. Some other procedures, such as the leaching environmental assessment framework (LEAF), were developed to combat TCLP deficiency to characterize metal leachability over a range of plausible and expected environmental conditions for several use and disposal scenarios (Kosson et al., 2014). For example, LEAF is currently being used by the USEPA to evaluate potential leaching from coal combustion residuals (e.g., fly ash and scrubber residues) due to the wider use of multi-pollutant controls at U.S. coal-fired power plants (Kosson et al., 2014).

In the last decade, the TCLP has been used extensively to test the leachability of heavy metals in amended contaminated soils to evaluate the efficiency of metal immobilization (Scheckel et al., 2013; Tsang et al., 2013). Phosphate materials are effective amendments for immobilizing heavy metals in contaminated soils (Hafsteinsdóttir et al., 2015; Moseley et al., 2008). For example, immobilization tests of Cd and Cu by $\text{NH}_4\text{H}_2\text{PO}_4$ over 35 days yielded the minerals $\text{Cd}_5\text{H}_2(\text{PO}_4)_4 \cdot 4\text{H}_2\text{O}$, $\text{Cu}_3(\text{OH})_3\text{PO}_4$ and CuNH_4PO_4 (Ayati and Lundager Madsen, 2000). The USEPA Best Management Practices Manual recommends phosphate application for binding Pb and preventing migration in soils at outdoor shooting ranges (USEPA, 2005). The purpose of using phosphate amendments is to form lead phosphates or pyromorphites $[\text{Pb}_5(\text{PO}_4)_3\text{X}]$, where $\text{X} = \text{Cl}^-$, OH^- , F^- , which are the most thermodynamically stable and insoluble Pb minerals over a large pH and Eh range (Scheckel et al., 2013). The corresponding Pb solubility is extremely low under many conditions of environmental significance. For example, the Ksp values for fluoropyromorphite $[\text{Pb}_5(\text{PO}_4)_3\text{F}]$, bromopyromorphite $[\text{Pb}_5(\text{PO}_4)_3\text{Br}]$ and hydroxypyromorphite $[\text{Pb}_5(\text{PO}_4)_3\text{OH}]$ are $10^{-71.6}$, $10^{-78.1}$ and $10^{-76.8}$, respectively, compared with $10^{-84.4}$

for chloropyromorphite $[\text{Pb}_5(\text{PO}_4)_3\text{Cl}]$. When the TCLP is applied to assess heavy metal leachability in P-amended soils, fluid chemicals may alter the chemical speciation of heavy metals, causing inaccurate assessments of heavy metal leaching. The strong acidity imparted during the TCLP may induce P amendment dissolution, which could enhance heavy metal phosphate precipitation. Ryan et al. (2001) and Scheckel and Ryan (2003) employed extended X-ray absorption fine structure (EXAFS) spectroscopy to prove that Pb speciation changes to more insoluble forms during the sequential extraction test. Similar results were reported when using the physiologically-based extraction test (PBET) by Scheckel et al. (2005). It was demonstrated that the speciation of extracted Pb in contaminated samples with phosphorus amendments was altered during the PBET test, suggesting that chemical extraction methods induced Pb precipitation and pyromorphite formation from soluble Pb. Dermatas et al. (2008) studied the performance of Pb occurring in alkaline-buffered and acidic-buffered firing range soils with a monobasic calcium phosphate amendment, and the results of X-ray powder diffraction showed that the brushite and cerussite peaks were significantly decreased and a small pyromorphite peak appeared in the TCLP residue, suggesting that the TCLP conditions favored Pb and P dissolution as well as Pb–P precipitation reactions.

The overall objective of this study was to determine the applicability of TCLP to assess Pb leaching in soils contaminated with different Pb forms and amended with different P sources. Specifically, this study investigated (i) the leaching behavior of $\text{Pb}(\text{NO}_3)_2$, PbSO_4 , and PbCO_3 in soils; (ii) the effects of soluble triple superphosphate fertilizer and less soluble phosphate rock on Pb leaching; and (iii) Pb speciation and potential Pb–P precipitation reactions in phosphate-amended soils during the TCLP test conditions.

2. Materials and methods

2.1. Soil and amendment characterizations

An uncontaminated soil was collected from the top 20-cm at a farm field in the suburb of Changshu city in eastern China. After air drying, the soil was crushed, passed through a 2-mm sieve, and homogenized. Selected physical and chemical properties of the soil are listed in Table 1. To meet the study objectives, fine powders of PbNO_3 , PbSO_4 or PbCO_3 were well mixed with the uncontaminated soil, bringing the Pb concentration in these soils to 15,000 Pb mg kg⁻¹, which was within the range of many Pb-contaminated soils in China (Deng et al., 2006; Li and Huang, 2007). Furthermore, a higher Pb percentage (i.e., 1.5%) was purposely spiked into the soil in an attempt to reach a sufficient Pb concentration to effectively identify potential Pb reaction products by using X-ray diffraction (XRD). These three artificially contaminated soils were referred to as PbNO_3 -soil, PbSO_4 -soil, and PbCO_3 -soil. The soils were equilibrated for two weeks at room temperature with 50–75% field moisture capacity before they were subjected to the amendment experiments.

The phosphate amendments included triple superphosphate fertilizer (TSP) and ground phosphate rock tailing (PR). The TSP was mainly composed of $\text{Ca}(\text{H}_2\text{PO}_4)_2 \cdot 2\text{H}_2\text{O}$, which is acidic and water-soluble (Dermatas et al., 2008), while the PR mainly consisted of fluoroapatite $[\text{Ca}_{10}(\text{PO}_4)_5\text{F}_2]$, which is alkaline and much less soluble (Cao et al., 2004). Selected physical and chemical properties of the P amendments are shown in Table 1.

2.2. Soil treatments

The phosphate amendments were applied evenly to all three Pb-contaminated soils at a 4:1 molar ratio of P/Pb. This ratio was

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