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Redistribution of cadmium and lead fractions in contaminated soil samples due to experimental leaching



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ABSTRACT

The sequential extraction procedure (SEP) is often applied to soils for identification of binding phases but rarely is it used to monitor binding dynamics temporally. In this study, contaminated soils were subjected to two different leaching scenarios, one analogous to a hydrostatic or stagnant water exposure, and one that represents a hydrodynamic event such as heavy precipitation. A four step SEP was applied before and after each stage of leaching and the adsorption dynamics of cadmium and lead were studied. In the hydrostatic experiment, removal of Cd and Pb occurred largely from the residual fraction (from initially >30%-zero Cd after the 720 h experiment, 40%-<20% Pb, and from 50%-20% Cd, >50%-40% Pb, from organic and mineral soil horizons respectively). There was a significant redistribution onto the Fe/Mn oxides (from initially 30%-60% Cd after 720 h, 40%-65% Pb, and from 15%–30% Cd, 45%–60% Pb for organic and mineral soils respectively). A significant redistribution of Cd onto the organic matter was observed (from initially zero, to 20% after 720 h in organic soil, and from 1%-15% in mineral soil). After the experiment, fraction distribution of the organic soil sample closely matched the distribution pattern of a slightly contaminated fluvisol sample that was taken in a river valley adjacent to the studied soils. When a CaCO₃ amendment was applied, dissolution of both elements decreased, >50% less Cd and Pb released from organic soil, and ~80% less Cd released from mineral soil with amendment. However, the fraction distributions remained similar to that of a non-amended sample. In the hydrodynamic leaching experiment, important binding phases of Cd were identified as organic matter (from initially 0%-15% after leaching and from ~2%-~10% in organic and mineral soils respectively) and metal oxides (from initially 40%-50% after leaching in organic soil). Residual Pb increased after leaching, and losses occurred from the Fe/Mn oxides (>50% decrease in organic soil). The CaCO₃ amendment had little influence on both element distribution and concentrations in hydrodynamic column leaching. It is anticipated that this study could be a precursor for modelling heavy metal fractionation of contaminated soils based on water regime in the environment.

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1. Introduction

The transfer of potentially toxic elements (PTE), in gaseous, particulate, aqueous or solid phase, from anthropogenic sources to the pedosphere can result in persistent contamination of surface and subsurface soil. Such occurrences of PTE present a challenge for authorities and landowners to maintain PTE values below soil guideline limits. A diverse array of remedial strategies has evolved in response to the demand for the mitigation of polluted soils,

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and many studies have focused on the physical chemistry of soil dynamics. This serves to increase understanding of the processes controlling the pollutant mobility and toxicity, often as a precursor to remediation technology development. Amendments aimed at immobilizing inorganic contaminants decrease trace metal leaching and reduce their bioavailability by inducing a range of sorption processes, e.g., adsorption to mineral surfaces, formation of stable complexes with organic ligands, and surface precipitation (Kumpiene et al., 2008). A number of studies have focused on the immobilisation of PTE with the addition of basic amendments (Alvarenga et al., 2008; Lin and Zhou, 2009; Kim et al., 2013). For example, the addition of calcium carbonate to a soil strongly reduces metal scavenging by the solution from all soil fractions. Houben et al. (2012) studied CaCO₃, and other amendments, for the potential



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immobilisation of Cd and Pb in contaminated soil. A reduction in Cd leaching was observed mainly due to increases in pH caused by amendment additions. However, only CaCO₃ addition resulted in a strong reduction of metal plant uptake. The addition of calcium carbonate during production of card products inspired recent research by Prica et al. (2013), who focused on the potential for cardboard mill pulp as an immobilizing amendment to Cd enriched sediments. The study showed promising results, with significant reductions in Cd mobility when pulp was incorporated with the sediments. The sequential extraction procedure (SEP) was developed for the purpose of characterizing soil binding fractions, by selectively targeting and releasing metals bound in certain geochemical phases (Larner et al., 2006). Identification of the main binding sites and pedochemical phase associations of trace metals in soils helps toward predicting their mobilisation potential and the likely risks they pose to the environment (Gleyzes et al., 2002). Although the SEP is utilized frequently as a descriptive tool in the study of contaminated soils (Padmavathiamma and Li, 2010; Tang et al., 2008; Yang et al., 2011), little consideration has been given to the dynamics of PTE binding with time, across the spectrum of different soil fractions. For example in leaching or extraction experiments that are synonymous to a particular leaching scenario. Over a prolonged exposure of contaminated soil to a given solvent, a question can be formulated as to which of the operationally defined fractions contribute to the "soluble" portion of metal during dissolution, and what are the dynamics between the metal fractions that are being leached. To date there exists a gap in the literature concerning the temporal changes to the PTE distribution in contaminated soils.

The aim of this study is to provide a description of the temporal changes in Cd and Pb concentrations between binding phases of contaminated soil samples after leaching in hydro-static and hydro-dynamic scenarios. Furthermore, we compare the binding fractions of a contaminated organic soil sample after leaching with a soil that is frequently saturated due to flooding. The study also seeks to determine the effects of a lime treatment (CaCO₃) on the Cd and Pb binding dynamics in the studied soils. It is intended that results from this study will provide new insight into the behaviour of Cd and Pb in contaminated soils that are subjected to periodic and prolonged saturation.

2. Materials and methods

2.1. Study site and sampling

Soil was collected from the edge of a smelter waste heap in the historic mining district of Kutná Hora, approximately 60 km to the east of Prague in the Czech Republic (Fig. 1). The area is situated on the Kutná Hora Crystalline Unit, represented by micaschist and orthogneiss zones. An extensive description of the geology of KH can be found in Kachlík (1999). Mining operations here followed the path of Agenriched quartz veins which formed from metamorphosed igneous material (Pauliš and Mikuš, 1998). Soils around the mining wastes in this area are highly enriched in Cd and Pb (plus other PTE) as evidenced in previous studies (Ash et al., 2014). The presence of metal bearing minerals (e.g., arsenopyrite, chalcopyrite, tetrahedrite, galena and sphalerite) in the material that was mined and smelted, plus fluxes used in the smelting process, resulted in a metal rich slag. Fragments of slag at the studied site were shown to be reactive in water: Cd exhibited increasing dissolution with contact time, Pb showed lesser solubility but was still extractable from the material (Ash et al., 2013). Furthermore, Cd and Pb, which may exist in elemental form or bound with sulphates and carbonates, are readily dissolved in soil solution. Hence, chemical weathering of slag is a likely source of PTE into the studied soils. By means of isotopic composition analysis, Ettler et al. (2004) confirmed that metallurgy and smelter activities play a predominant role in severely contaminating local soils, and that downward migration of Pb in smelter-impacted soils can occur at a rate of approximately 0.3 to 0.36 cm/yr. Although the operation of smelters at our studied vicinity ceased sometime in the 1800s, mining and ore processing continued in the nearby locality of Kaňk (approx. 3 km north) up until 1991 (Vrátný, 1998). In addition to any atmospheric fallout, physical weathering and aeolian deposition of metallic dust from the slag heap are likely to compound the contamination of the studied soils.

One pit was dug on a flat area adjacent to the smelter waste heap (marked A in Fig. 1) where a thin layer of leaf litter and vegetation debris has accumulated and grass is growing sparsely. The pit was dug to sufficient depth so as to expose the mineral layer which underlies the organic horizon. Soils at the edge of the waste heap have developed over the last

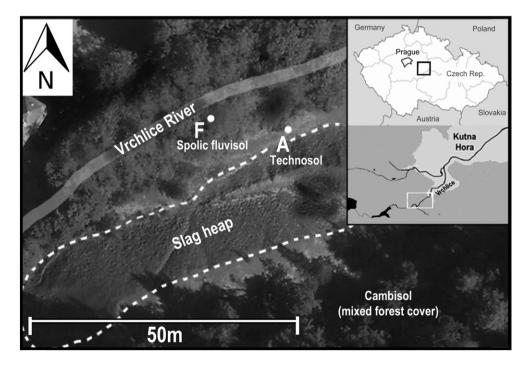


Fig. 1. Map of study area to the south west of Kutná Hora showing soil type, Vrchlice River, and position of sample pits.

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