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Stabilization of Pb- and Cu-contaminated soil using coal fly ash and peat

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Copper and lead mobility and bioavailability in soil can be effectively reduced by using a combination of coal fly ash and peat as soil amendments.

Abstract

The stabilization of metal contaminated soil is being tested as an alternative remediation method to landfilling. An evaluation of the changes in Cu and Pb mobility and bioavailability in soil induced by the addition of coal fly ash and natural organic matter (peat) revealed that the amount of leached Cu decreased by 98.2% and Pb by 99.9%, as assessed by a batch test. Metal leaching from the treated soil was lower by two orders of magnitude compared to the untreated soil in the field lysimeters. A possible formation of mineral Cu- and Pb-bearing phases and active surface with oxides were identified by chemical equilibrium calculations. Low metal leaching during a two-year observation period, increased seed germination rate, reduced metal accumulation in plant shoots, and decreased toxicity to plants and bacteria, thereby demonstrating this stabilization method to be a promising technique for in situ remediation of Cu and Pb contaminated soil.
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1. Introduction

High metal mobility is characteristic of sites contaminated with high levels of metals that pose a threat to human health and the environment. The mining industry is a major source of soil contamination with trace elements, e.g. Cu and Pb. The metals are spread through ore transshipment, mine waste storage, acid drainage generation from sulfide containing mine wastes that cause soil acidification, and contaminant leaching.

Soil amendment with minerals such as Fe oxides, organic matter, alkaline materials can reduce metal mobility and availability in soil by adsorption, complexation, (co)precipitation or a combination thereof, and help to restore soil properties (Basta et al., 2005). Iron oxides (FeOx) have a high sorptive capacity for metals (Chen et al., 2000; Lombi et al., 2002; Brown et al., 2005). Numerous studies have demonstrated the successful immobilization of metals, especially Pb, by phosphorus containing materials due to the precipitation of pyromorphite-type minerals (McGowen et al., 2001; Cao et al., 2003, 2004; Chen et al., 2003; Melamed et al., 2003; Brown et al., 2005). Industrial by-products, like red mud, fly ashes, berringite, biosolids, dolomitic residues, have been shown to a varying extent contribute to metal immobilization in soil (Mench et al., 2000; Basta and McGowen, 2004; Brown et al., 2004; Garrido et al., 2005).

However, application of soil amendments may not always sufficiently reduce Cu and Pb mobility or improve soil quality and vegetation establishment. Hartley et al. (2004) tested several iron amendments (goethite, metallic Fe, iron(II)/(III) sulfates plus lime), but none of the treatments were efficient for Cu stabilization. The surface charge of FeOx is pH dependent

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and metal adsorption decreases with decreasing pH (Cornell and Schwertmann, 2003). Therefore, by using FeOx in acidic soils, additional pH adjusting materials as well as fertilizers are needed to increase the sorptive capacity of FeOx and facilitate revegetation of the soil (Bleeker et al., 2002).

Despite the reported high treatment efficiencies, P added to soil can easily form other minerals, e.g. apatite and manganese hydrogen phosphate rather than Pb pyromorphites (Porter et al., 2004). Increasing the amount of added P could help to overcome this problem, although an excess of soil P can be a potential source of the eutrophication of surface waters (Brown et al., 2004) and high amounts of precipitated apatite-like minerals can adversely change the soil structure (Porter et al., 2004).

Industrial by-products, like coal and biofuel combustion fly ashes (CFA), are alkaline materials with high sorptive capacity, mainly composed of ferroaluminosilicates, and can be used as ameliorants for acidic soils. Fly ashes are suggested to solve problems related to acid mine drainage and metal solubility (Misra et al., 1996; Iyer and Scott, 2001; Xenidis et al., 2002). Fly ashes increase the surface area available for element adsorption, improve the physical properties of soil (Gorman et al., 2000), neutralize the pH of acidic soils and render most cationic metals less mobile (Ciccu et al., 2003). CFA contains alkaline (K) and alkaline earth (Ca, Mg) metals that are important plant nutrients. A mixture of CFA with organic matter (OM) is expected to further enhance biological activity in the soil (Jala and Goyal, in press), reduce leaching of major nutrients (Sajwan et al., 2003) and be beneficial for vegetation (Rautaray et al., 2003; Tripathi et al., 2004).

The use of fly ashes as soil amendments would solve several problems by reducing the amount of landfilled soil and ashes, reduce the consumption of soil ameliorants (fertilizers, lime) and decrease the metal mobility and availability in soil (Ciccu et al., 2003; Mittra et al., 2005).

The aims of this study were to determine whether a combined addition of CFA and natural OM (peat) to an acidic soil contaminated with sulfidic ore deposits can reduce Cu and Pb mobility, change their distribution between metal-binding soil fractions, as well as reduce the metal toxicity to microorganisms and uptake by plants.

2. Material and methods

2.1. Soil

The soil used was sampled from a Cu ore transshipment station in Slagnäs, Sweden. A 400 kg composite soil sample of similar texture was excavated from the surface and to a depth of 20 cm. The soil was air dried, homogenized and sieved to a <4 mm fraction for the batch leaching tests and to <2 mm for pH_{stat} leaching tests, sequential extractions and toxicity assessment.

2.2. Amendments

The tested coal fly ash was generated by wood and coal combustion at Öresundskraft, Sweden. The pH was 12.4 and the electrical conductivity (EC) of the CFA was 20 mS cm^{-1}. Total solids were 999 g kg^{-1}, the loss of ignition was 6%, and the average particle size was 6.9 μm. Metal concentrations were (±SD, $n = 3$): Ca 15.3 ± 0.8; Fe 6.0 ± 0.7; Al 4.1 ± 0.5; Mg 2.8 ± 0.2; K 1.9 ± 0.2; Na 1.4 ± 0.2; Mn 0.5 ± 0.1%, and Zn 425 ± 34; Cu 71 ± 3; Pb 32 ± 3; Ni 31 ± 3; Cr 19 ± 4; Cd 2.4 ± 0.4 mg kg^{-1}.

Peat was obtained from Norrlandsjord, Luleå, Sweden. According to the von Post peat decomposition classification (Wiklander, 1976), the peat is defined as ‘‘almost completely unhumified, containing plenty of fibers’’. The peat pH was 3.9 and the EC was 0.1 mS cm^{-1}. The percentage of ash after combustion at 550 °C was 17%.

About 200 kg of the air dried, non-sieved soil was manually mixed with 10 kg dry weight (DW) CFA and 10 kg DW peat.

2.3. Lysimeters

Of 4 polyethylene (PE) lysimeters (100 L), 2 were filled with the treated soil and 2 with the untreated soil. One lysimeter from each pair was sowed with a seed mixture supplied by Veg Tech AB, Vislanda, Sweden. The plant seed mixture, typically used for re-vegetation of sandy, nutrient deficient soils, was composed of 90% grass and 10% herb species (Table 1). A porous PE plate was placed 10 cm from the lysimeter bottom, thereby forming an empty space for the leachate accumulation. The plate was covered with a geotextile filter to prevent wash out of small particles. The lysimeters were placed in holes to a depth of 0.5 m and exposed to natural precipitation and temperature. Leachate samples were pumped from the bottom of the lysimeters.

The experiment was conducted from July 2003 to November 2004. Leachate samples were collected 10 times over a 500-day period, mostly after considerable rain events and snow melting, excluding the winter period when the soil was frozen.

2.4. Leaching tests

A batch leaching test was used to estimate the water soluble fraction of metals prior to and after the soil treatment following two weeks of equilibration time. A randomized triplicate 2^2 full factorial design (Montgomery, 2001) was applied. The low levels of the factors CFA and OM were set to 0% to examine if any leaching of contaminants was due to the single factors or to their interactions. The high levels of the factors were set to 5%, (i.e. the amount of amendments).

The air-dried soil was mixed with the two amendments according to the factorial design and left at room temperature for two weeks to equilibrate at a humidity level corresponding to 50% of the soil water holding capacity. The samples (100 g of each) were then filled into 1.5-L PE bottles and mixed with water acidified to pH 4 using HNO_3 at a liquid-to-solid ratio (L/S) 10 L kg^{-1}. The samples were shaken for 24 h using an orbital shaker. pH and EC were measured in unfiltered samples.

The untreated soil and the soil treated with both amendments were leached for 24 h at L/S 10 in a pH_{stat} mode by adding 1 M HNO_3 or 1 M NaOH solutions to maintain pH 3 or pH 7 conditions, respectively.

Table 1
The seed mixture of plants used for soil revegetation

Herbs (10%)	Grasses (90%)
Achillea millefolium	*Agrostis capillaris*
Anthyllis vulneraria	*Anthoxanthum odoratum*
Campanula rotundifolia	*Bromus hordeaceus*
Hieracium pilosella	*Deschampsia flexuosa*
Hieracium umbellatum	*Festuca rubra*
Hypericum maculatum	*Festuca ovina*
Leucanthemum vulgare	
Lotus corniculatus	
Linaria vulgaris	
Rumex acetosa	
Silene dioica	
Thlaspi cearulescens	
Vicia cracca	

Proportions of the species of herbs and grasses in the mixture are given in parentheses.

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