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Evaluation of solid residues quality after enhanced Cu leaching of polluted soils

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HIGHLIGHTS

- Cu is leached from polluted soils using acidic process wastewater from incineration.
- The Cu content in the soils was reduced five times or more after leaching.
- The solid residues were safe enough to be deposited as non-hazardous waste.
- The soil function to filter/buffer for metals was not highly affected by leaching.

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ABSTRACT

Excavation followed by landfilling is one of the most common methods for treating soils contaminated with metals. Removing the metals through soil washing not only allows valuable substances to be recovered, but also results in cleaner soil residues. In this project a method for leaching and recovering Cu from polluted soils using acidic wastewater is further developed and evaluated, with special attention to the leaching process. In addition, the qualities of the soil residues are assessed in order to investigate how the proposed remediation method affects the soil properties. Soil samples highly polluted with copper (Cu) were collected from two sites in Sweden. After acidic leaching and water washing, the Cu content of the soil samples was reduced five times or more. The original soils could not even be deposited in landfills for hazardous waste; however after treatment of the soils according to the proposed method, the Cu leaching decreased six-fold and the solid residue was safe enough to be deposited in landfills for non-hazardous waste. The soil function "soil as filter and buffer for heavy metals" was evaluated using the TUSEC (technique for soil evaluation and categorization for natural and anthropogenic soils) manual. Originally the soils were of "low" i.e. class 4 or "very low capacity of binding and buffering heavy metals" i.e. class 5, while after the remediation process, both soils were categorized as Class 5. To summarize, the proposed method clearly shows potential not only for remediation of Cu polluted soils but also indicate a potential for recovery and reuse of Cu from the leachates generated. Even though the previously highly polluted soils could not be directly put back at the original sites, the solid residues could be deposited in landfills for non-hazardous waste, which is an improvement, considering the original soils could not even be deposited in a landfill for hazardous waste.

1. Introduction

World-wide, the number of contaminated sites in need of remediation is enormous, and only in Western Europe the number of estimated potential contaminated sites is more than 2.5 million (Panagos et al., 2013). Municipal waste and industrial waste contribute most (38%) to the soil contamination; mineral oil and toxic metals are the main soil polluters (60%). Remediation of soils contaminated with metals is difficult, time consuming and expensive because of the toxicity and persistence of the metals. Excavation followed by landfilling, so-called dig-and-dump, is due to the Swedish EPA the most common method for treating soils contaminated in Sweden (Helldén et al., 2006); the reason for this is the relatively low disposal costs and the often urgent remediation. In addition to the fact that landfilling is not sustainable in the longer perspective, valuable metals potentially present in the landfilled masses are not utilized and returned into the societal cycle, but left in the landfill with risks of future leaching into the surrounding area and groundwater, posing significant risks to the environment and human health. Consequently, there is a need for alternative treatment methods.

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Fig. 1. Photographs of original dried soil samples: (1) clayish fine sand/coarse silt and (2) slightly clayish sand and coarse silt.

One interesting method, which was discussed already in the 1990's by for instance the US EPA, is soil washing combined with metal recovery (U.S. EPA, 1995; 1999; Karlfeldt Fedje et al., 2013; Ortega et al., 2008; Dermont et al., 2008). Until now the use of these methods are limited due to e.g. technical limitations and high costs, but as the demand for metals is increasing this becomes more interesting. In addition, removing the metals not only allows valuable substances to be recovered but also results in cleaner soil residues, which might reduce the needs for landfills.

The purpose of soil washing is to leach out the metals into a liquid in a form that is as concentrated as possible using different chemical leaching agents, e.g. water, acids, salts, chelants, surfactants, reducing and oxidising agents (Voglar and Lestan, 2013; Bisone et al., 2012; Moon et al., 2012; Lo et al., 2011; Dermont et al., 2008). In a study of soils from industrial timber treatment sites, contaminated with Cu, Cr and As, it was shown that addition of humic substances promoted the leachability of Cu at high pH. This was explained by Cu forming soluble Cu-humate complexes with dissociated carboxylic and phenolic groups (Hartley et al., 2014). At lower pH the humic substances precipitated and was adsorbed on the soil surface and acted as a barrier to metal leaching. This has been widely recognised and it is explained by the protolysis of the acidic groups in the humic substances making them dens and curled because the repellent negative charges in the carboxylic and phenolic groups do not keep them dissolved at low pH. The adsorbed humics inhibited the metals release by chelating, steric blocking, modification of the surface properties and adsorption of metal-humate complexes (Yip et al., 2010). Several leaching agents, including HCl, NH_4Cl, lactic acids, EDDS and acidic by-product process waters from solid waste incineration have been tested and compared on soil, bark and bark-ash samples (Karlfeldt Fedje et al., 2013). The results showed that the acidic process water was the most efficient leaching agent, resulting in as much as 90–100% of the Cu being leached. To combine the chemical extraction procedures with pre-physical separation methods based on e.g. size, density or flotation properties have been successfully used where the concentration of particulate forms of metals into smaller volumes of soil will make the subsequent chemical extraction step more effective (Sierra et al., 2011, 2014; Dermont et al., 2008).

As discussed above there is much research on the mobility and release of metals from contaminated soils, but the research on use and reuse of these soil materials is limited in the scientific literature, highlighting the importance of doing research in this area. Among the available articles are immobilisation and encapsulation, mainly prior to landfilling-like applications (Camenzuli et al., 2015; Travar et al., 2015). Another area is phytoremediation projects, e.g. Chigbo and Batty (2015) and Ghosh and Singh (2005). However, the initial aim of soil remediation is to make the soil residue clean enough to, preferably, be reused in society or where this is not possible, be deposited in a less restricted landfill site. The challenge in soil washing projects is that during the chemical extraction there is an impending risk that the soil matrix will be degraded or destroyed, resulting in a humic and nutrient poor soil useless for primary production (Makino et al., 2007). There is also a risk that the physical separation followed by the chemical processing will destroy the physical structure of the soil, causing changes in the permeability and shear strength, which will affect the long-term soil functions (Zihms et al., 2013), and make the washed soil less useful as filling and construction material. The soil washing will affect soil properties such as texture, water holding capacity, organic matter and concentration of total nitrogen (Im et al., 2015). In order to evaluate contaminated soil residues after treatment, the soil function concept is used. Here, it is important to not only look at chemical contamination concentrations, but to also evaluate the physical and biological soil properties after treatment (Bone et al., 2010a, 2010b). The soil function evaluation criteria have been used in combination with multi-criteria decision analysis (MCDA) for sustainable appraisal of remediation alternatives (Volchko et al., 2014), resulting in a method tested on a real case for evaluation of the effects of remediation alternatives on selected ecological soil functions. This method has been further developed to create the SCORE (Sustainable Choice of Remediation) MCDA method, where economic, environmental and social criteria are all taken into account in the assessment of different remediation methods for contaminated soils (Rosén et al., 2015).

In this article, contaminated soils from two sites in Sweden are studied with the aim to leach and recover metals from the soils. The process for enhanced acidic soil washing was optimized with respect to physical pre-treatment effects, acidic liquid concentrations, time, liquid-to-soil ratio (L/S) and after-washing steps. Special attention was given to the soil residues, in order to transform the original hazardous soils into less toxic materials. For this reason, the characteristics of the original and leached soils were thoroughly evaluated according to their potential future usage. In addition, the soil function "soil as filter and buffer for heavy metals" was also evaluated.

2. Materials and methods

2.1. Soil samples

Soil samples with different characteristics from two sites – Köpmannebro (1) in western Sweden and Björkhult (2) in eastern Sweden – were used in this study. Both sites were formerly used for timber industry activities and are strongly contaminated with Cu, historically used for impregnation of wood with $CuSO_4$ according to the Boucherie method. Representative soil samples from several locations at each site were collected from depths of 50–>100 cm at Site (1), and of 20–60 cm at Site (2). The sub-samples were mixed to produce homogenous soil samples, then gently dried at 105 °C until constant weight, i.e. 100% dry solids (DS) (Fig. 1).

2.2. Leaching experiments

Highly acidic process wastewater (pH around 0) was used as leaching agent, as it has been used in previous studies with satisfying results (Karlfeldt Fedje et al., 2013, 2015). The process water was produced using the wet flue gas cleaning process from incineration of municipal solid and industrial waste in full scale. Consequently, the process water contained metal ions and chlorides that were present in the flue gases. The concentrations of different ions in the process water vary

Table 1
Concentrations of major ions present in the acidic process water used for the leaching experiments.[a]

Process water	Cl	Na	K	Zn	Fe	Si	B	Pb	Ca	Al	Cu	Sb	P	Mg	Hg
[mg/L]	56 000	270	94	62	35	32	1.0	11	11	7.0	3.0	3.0	2.0	2.0	1.0

[a] All other analysed ions (As, Ba, Be, Cd, Co, Cr, Li, Mn, Mo, Ni, Se, Ag, Sr, Tl, U and V) were detected in concentrations <1.0 mg/L.

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