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Nickel, Lead and Zinc Sorption in a Reclaimed Settling Pond Soil

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ABSTRACT

The wastes used to amend soils sometimes have high concentrations of metals such as nickel (Ni), lead (Pb) and zinc (Zn). To determine the capacity of soils to retain these metals, the sorption capacities of different mine soils with and without reclamation treatments (tree vegetation and waste amendment) for Ni, Pb and Zn in individual and competitive situations were evaluated using the batch sorption technique. The untreated settling pond soil had low capacity for Ni, Pb and Zn retention. The site amended with wastes (sewage sludges and paper mill residues) increased the sorption capacity most, probably because of the higher concentrations of soil components with high retention capacity such as carbon and clay fraction. No significant competition was observed between metals in the competitive sorption experiment, indicating that the maximum of sorption was not achieved by adding 0.5 mmol L^{-1} of metal. We can conclude that, despite the possible additions of Ni, Pb and Zn from wastes to degraded soils, sewage sludges and paper mill residues have a high sorption capacity that would prevent the metals from being in a mobile form.

Key Words: metals, mine soil, retaining capacity, tree vegetation, waste amendment

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INTRODUCTION

The most commonly used reclamation treatments with mine soils are planting vegetation and amending with wastes. The main purpose of reclamation treatment is to improve the physical, chemical and biological properties of the degraded mine soils in order to stabilize the land and avoid inappropriate runoff and drainage, amongst other problems. Also, in metal mine sites it is crucial that reclamation treatments reduce the bioavailable concentrations of the metals in soils. The waste amendments and the soil conditions that vegetation creates can favour the retention of heavy metals, but as the retention mechanism of metal ions at the soil surface is often unknown, the term "sorption" is preferred (Sparks *et al.*, 1999). This term involves the loss of a metal ion from an aqueous to a contiguous solid phase, and consists of three important processes: adsorption, surface precipitation and fixation (Apak, 2002; Bradl, 2004). Although there are several published studies on the effect of reclamation treatments on mine soils (Bendfeldt *et al.*, 2001; Brown *et al.*, 2003; Shrestha and Lal, 2008; Baker *et al.*, 2011), there is hardly any information on the effect of waste amendments on the sorption capacity of these type of soils. Moreover, as there is usually more than one metal in amended soils, it is very important to evaluate the competition and preference of metals for binding sites. Synergies and antagonisms are often reported, and the extent of the metal sorption can differ significantly if more than one metal is present (Vega *et al.*, 2006; Covelo *et al.*, 2007a). Both planting vegetation and amending with wastes (such as sewage sludge, manure or ashes) significantly increase the concentration of organic matter, carbonates and clay in degraded mine soils (Filcheva *et al.*, 2000; Bendfeldt *et al.*, 2001; Alvarenga *et al.*, 2008; Asensio *et al.*, 2013a). Because of the characteristics of these soil components, the increase in their contents would imply a higher metal retaining capacity than that before reclamation. Iron (Fe) and manganese (Mn) oxides are sometimes common in mine soils, and also have a high sorption capacity (Vega *et al.*, 2006). It is expected that the combination of organic matter, clay, carbonates and Fe and

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Mn oxides from the reclaiming treatments or from the former soil can create new conditions for retaining metals. Based on this hypothesis, samples from a mine soil from the Touro Mine in Galicia, Spain were taken in order to find out if the vegetation planted and waste added increased the metal sorption capacity of these soils.

Previous studies have shown that the wastes used to reclaim mine soils often contain considerable concentrations of nickel (Ni), lead (Pb) and zinc (Zn) that could enter the soils (Nyamangara, 1998; Singh and Agrawal, 2008; Asensio *et al.*, 2013b). Therefore, sorption studies on mine soils reclaimed with sewage sludge must also take into account the additional input of those metals in the soils. Some studies have evaluated the sorption capacities of soils from reclaimed sites (Vega *et al.*, 2006), but without a comparison with untreated soils. We only know one study that compares the sorption capacities of soils from treated and untreated sites, although in this case the metals are cadmium (Cd), copper (Cu) and Pb (Vega *et al.*, 2009). Therefore, the information available on the sorption of Ni, Pb and Zn in reclaimed metal mine soils is limited. The main objective of this study was to evaluate the sorption capacity of Ni, Pb and Zn of the soil formed in the settling pond of a depleted Cu mine. Previous data showed that samples from treated sites had higher concentrations of Ni, Pb and Zn than the untreated sites, and that unexpectedly, the highest proportion was in the residual fraction of the soil (bound to clays) (Asensio *et al.*, 2013b). The present study aimed to go further by showing whether the settling pond soils were capable of retaining high concentrations of Ni, Pb and Zn, making it possible to estimate whether mine soils can reduce the availability of metals when they are amended with wastes.

MATERIALS AND METHODS

Description of the study area and soil sampling

Soil samples were taken at the Touro Mine in Galicia, Northwest Spain (8°20′12.06″ W, 42°52′46.18″ N). There were two opencast mines at Touro, Arinteiro and Bama, from which chalcopyrite and pyrrotine were extracted between 1973 and 1988 with the aim of obtaining Cu. The rocks at this site are amphibolites with a high garnet content, and these minerals form part of a great schist and amphibolite area. The Cu extraction stopped in 1988 and, since then, materials for road construction were extracted at Touro. The mine tailing covered a surface of 760 ha, formed by great piles of coarse rock fragments with strong slopes. In addition to Cu, Fe and sulfur (S), chromium (Cr) was present in high concentration in the ore.

A settling pond of 71 ha was created during the flotation process for extracting Cu while mining. The area has been completely dry for several years, and because of the later accumulation of waste material, the resulting soil can be considered as a Spolic Technosol according to the latest version of the FAO classification (FAO, 2014). Four sites were selected at the settling pond area: i) untreated site (B1, the control sample), ii) the old-vegetated site (B2v), where *Pinus pinaster* Aiton was planted 21 years before the sampling date, iii) the young-vegetated site (B3v), where *Eucalyptus globulus* Labill was planted 6 years before sampling and iv) the amended site (B4w), where sewage sludges and paper mill residues were added 5 months before sampling. The wastes were added with trucks and then spread on the soil surface, without being mixed with the mine soil. The municipal sewage sludges were a dewatered anaerobic digested sludge (AN) obtained from a wastewater treatment plant in Ourense (Spain) and a previously CaO-treated aerobic sludge (AE), obtained from a water treatment plant in Santiago de Compostela (Spain). The paper mill residues (sludges and ashes) were obtained from a paper production plant (Iurreta Corporation SA, Basque Country, Spain) where it is produced during the cogeneration of energy by burning tree bark. The range of the pH of the wastes used as amendments was 7–11.5, the total organic C varied between 150–230 g kg^{-1}, the total Cu ranged from 100 to 500 mg kg^{-1}, total Ni from 30 to 60 mg kg^{-1}, total Pb from 50 to 180 mg kg^{-1} and total Zn from 130 to 870 mg kg^{-1} (Camps Arbestain *et al.*, 2008). On March 9, 2010, a single horizon of all of the soils was sampled to 20 cm, except from B4w, where two soil horizons were sampled from 0 to 20 cm (B4Aw) and 20 to 40 cm (B4Bw), in order to observe possible changes in the subsurface horizon. Soil samples were randomly collected in each selected site from areas spaced sufficiently far apart to be representative. The samples were stored in polyethylene bags, dried at room temperature and sieved to < 2 mm before being analysed.

General characterization of the soils

Some of the characteristics of the soils we studied that generally influence metal sorption are summarized in Table I. The standard procedure of the pipette method was used to determine soil particle size distribution, *i.e.*, the percentages of sand, silt and clay (Kroetsch and Wang, 2008). Clay and silt fractions were determined by pipette, based on the Sto-

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