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Effects of tree vegetation and waste amendments on the fractionation of Cr, Cu, Ni, Pb and Zn in polluted mine soils

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HIGHLIGHTS

► Planting pines or eucalyptus to mine areas decreased Cr and Cu contents in soils.
► Cr and Cu were mobilised from non-mobile fractions in soils vegetated with trees.
► Amending with wastes also attenuated pollution by Cr and Cu in mine soils.
► Wastes could add considerable concentrations of Ni, Pb and Zn.

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ABSTRACT

Soils at a depleted copper mine in Touro (Galicia, Spain) are physically and chemically degraded and have also polluted the surrounding area. Due to these environmental problems and the large area of these mine soils, the reclamation strategies carried out at Touro have consisted of planting trees (pine or eucalyptus), amending with waste material (sewage sludge and paper mill residues), or using both treatments. Tree planting has been carried out for 21 years and waste amending for 10. Two different zones were selected in the mine (the settling pond and mine tailing) in order to evaluate the effect of the different reclamation practices on the chemical fractions of Cr, Cu, Ni, Pb and Zn. The results showed that soils in the untreated sites were polluted by Cr and Cu. Planting pines and eucalyptus on mine soils decreased the concentration of these heavy metals in non-mobile soil fractions. Amendments also attenuated pollution by Cr and Cu as the wastes that were used had lower concentrations than the untreated mine soils. Planting trees increased Ni, Pb and Zn retention in the non-mobile fractions, preventing them from being leached into surrounding areas. However, caution should be exercised when adding organic wastes, as they can lead to increase concentrations of Ni, Pb and Zn and their phytoavailable form. The results also showed that changes in the chemical fractionation of heavy metals in soils was more influenced by the clay percentage and both dissolved and soil organic carbon (SOC and DOC) than by soil pH or cation exchange capacity.

1. Introduction

Waste material produced by metal mining is accumulated in two different sites. Material with a diameter of more than 1 mm is deposited in what are known as mine tailings, while fine materials, produced during the last step of metal extraction, are accumulated in settling ponds (also known as flotation banks). Settling ponds dry over time once mining activity has ended. Mine tailings and dry settling ponds are accepted as soils and defined as Technosols by the FAO (2006), since they have properties and pedogenesis dominated by their technical origin (made by humans). Mine soils resulting from metal mining have significant limitations for the survival of living organisms, such as extreme pH, low cation exchange capacity, pollution by heavy metals and low organic matter and nutrient concentrations (Akala and Lal, 2001; Vega et al., 2005; Asensio et al., 2011; Barrutia et al., 2011). Mine areas can cause significant environmental problems in the surrounding areas if their soils are not reclaimed. The first problem is the acid mine drainage produced when the sulphides in the rock fragments are oxidised (Johnson and Hallberg, 2005; Lin et al., 2007). The second problem is the increase in the solubility of metals due to the low mine soil pH (McBride et al., 1997; Lombi et al., 2002; Fandiño et al., 2010). Because of all these environmental problems, it is necessary to improve the quality of mine soils to avoid pollution and help plant species to develop. Soil reclamation strategies usually involve planting vegetation and amending. As the reclamation of mine soils requires large amounts of money due to their huge area, one of the most popular mine soil

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amendments used in recent years is to add waste material (Calace et al., 2005; Tandy et al., 2009; Baker et al., 2011; Karami et al., 2011). An important factor in evaluating the effect of reclamation treatments on mine soils is to determine the distribution of heavy metals in the different soil fractions. A knowledge of the concentration of each heavy metal in each soil phase provides both the current and the potential phytoavailable concentrations. Total concentrations of heavy metals in soils only provide limited information on their toxic effects, since they are associated with various soil components in different ways (Jeng and Singh, 1993; Ahumada et al., 1999).

The effect of organic amendments and/or vegetation on mine soils polluted by heavy metals have been evaluated by several authors (Vega et al., 2004, 2005; Shu et al., 2005; Conesa et al., 2007; Lottermoser et al., 2008; Karami et al., 2011). However, only a few have carried out chemical fractionation of the retained metals (Tandy et al., 2009; Abbaspour and Golchin, 2010), and there is still a lack of information on the chemical fractionation of heavy metals in reclaimed mine soils under field conditions. The novelty of the current study is that we carried out a sequential extraction of heavy metals in mine soils under field conditions (within the mine) treated by planting trees or/and amending with wastes. The sampling site was located at the mine in Touro (Galicia, Northwest Spain). The mine tailing at this mine was formed by materials left over after extracting copper from the ore. There was also a settling pond in this mine, consisting of fine materials from the copper flotation process. Copper was mined in Touro for 15 years, between 1973 and 1988. Since then, the mine has been used for the extraction of material for road construction. The company "Tratamientos Ecológicos del Noroeste" (Ecological Treatments of the Northwest) has carried out different reclamation treatments at Touro mine soils: planting eucalyptus and pine trees, amending with wastes (mainly sewage sludge and paper mill residues) and both tree planting and waste amending at the same time. Therefore, the main aim of this study was to evaluate whether these three treatments are able to decrease the bioavailability of heavy metals in polluted mine soils. For this reason, both the mobility and phytoavailability of Cr, Cu, Ni, Pb and Zn in different soil fractions were determined in reclaimed soils from a copper mine. We hypothesized that planting eucalyptus or pine trees, amending with sewage sludges and paper mill residues or combining the two treatments could decrease the mobility of heavy metals in mine soils.

2. Materials and methods

2.1. Study areas and soil sampling

The mine is located in Touro (Galicia, Northwest Spain) (Lat/Lon (Datum ETRS89): 8° 20′ 12.06″ W 42° 52′ 46.18″ N) (Fig. 1). The climate in this zone is Atlantic (oceanic) with precipitation reaching 1886 mm per year (with an average of 157 mm per month) and a mean daily temperature of 12.6 °C. The average relative humidity is 77% (AEMET, 2012).

The settling pond (B) and the mine tailing (M) of the mine were studied. One untreated site (B1), two vegetated sites (B2v and B3v) and one amended site (B4w) were sampled in the settling pond, whereas one untreated site (M1), one vegetated site (M2v), one amended site (M3w) and one vegetated-amended site (M4vw) were selected in the mine tailing (Table 1). Wastes added to the amended sites had pH 8–10, total organic C higher than 150 g kg^{-1}, total Cu higher than 100 mg kg^{-1} and total Zn higher than 300 mg kg^{-1} (Camps Arbestain et al., 2008). Five soil samples were randomly collected in points sufficiently spaced to be representative of each site on 9th March 2010. Samples were stored in polyethylene bags, dried at room temperature and sieved to <2 mm prior to being analysed. All of the soils only had one horizon, except for two, where both horizons were sampled (described below). The soils were classified according to the latest version of the FAO (2006).

2.1.1. Settling pond samples

The settling pond site was completely dry at the sampling time. The control sample from this zone (B1) was in an untreated area. B1 is a Spolic Technosol located 336 m above sea level, covering an area of 1.9 ha, with an AC horizon 40 cm deep and without vegetation. The second settling pond sample was chosen for its old vegetation (B2v). Pine trees (*Pinus pinaster* Aiton) were planted here in 1989 (21 years). B2v is also a Spolic Technosol, with spontaneous vegetation: eucalyptus (*Eucalyptus globulus* Labill), gorse (*Ulex* sp.), heather (*Erica* sp.), *Agrostis* sp. and bryophytes. The B2v soil covers 6200 m^2 and is 340 m above sea level, with an AC horizon 20 cm deep. The third settling pond sample was chosen because of its young vegetation (B3v), in order to compare it with B2v and to observe the effect of trees over time. The B3v area was vegetated with eucalyptus in 2004, and is also a Spolic Technosol. This area also has spontaneous vegetation: pines (*P. pinaster* Aiton), *Agrostis* sp., broom (*Cytisus* sp.), *Acacia* sp. and bryophytes. The B3v soil covers 1.15 ha and is 335 m above sea level, with an AC horizon 20 cm deep. The fourth settling pond sample was located in a recently amended area (B4w). Sewage sludge mixed with paper mill residue had been added 5 months before the sampling date. These wastes were directly added with trucks and then spread on the soil surface, without being mixed in with the mine soil. The final depth of this new layer was 20 cm, with a volume of 280 tons per ha. Only spontaneous herbaceous vegetation grew on B4, measuring 100 m^2 and at a height of 339 m above sea level. B4w was an Urbic Technosol with two horizons: AC (B4Aw) and C (B4Bw). B4Bw was also sampled to

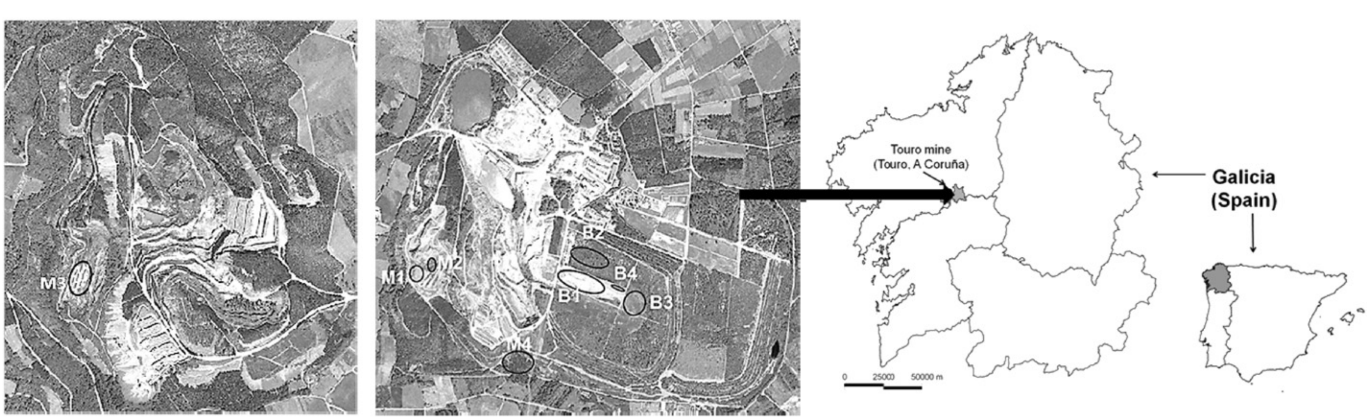

Fig. 1. Location of sampled sites in the Touro mine. Soil description is shown in Table 1.
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