

Competitive sorption and desorption of heavy metals in mine soils: Influence of mine soil characteristics

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Abstract

Many mine soils are chemically, physically, and biologically unstable and deficient. They are sometimes amended with sewage sludge and ashes but often contain heavy metals that increase the already high mine soils' heavy metal contents. Cd, Cr, Cu, Ni, Pb, and Zn in mutual competition were added to five mine soils (Galicia, Spain). Soil capacities for heavy metal sorption and retention were determined by means of distribution coefficients and selectivity sequences among metals. Influence of soil characteristics on sorption and retention was also examined. Retention selectivity sequences indicate that, in most of the soils, Pb is the preferred retained metal, followed by Cr. The last metals in these sequences are Ni, Cd, and Zn. Soil organic matter content plays a fundamental role in control of Pb sorption. Gibbsite, goethite, and mica influence Cr retention. Soil organic matter, oxides, and chlorite contents are correlated with $K_d \sum_{sp} \text{medium}$. Heavy metals are weakly adsorbed by soils and then desorbed in high amounts. To recover these soils it is necessary to avoid the use of residues or ashes that contain heavy metals due to their low heavy metal retention capacity.

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1. Introduction

Many mine soils (soils developed over mine spoil) are mechanically, physically, chemically, and biologically deficient: they are shallow, texturally unbalanced, and unstructured, and their strong acidity, due to rapid, intense oxidation [1,2], results in their leaching large quantities of cations (Al^{3+} , Fe^{2+} , Mn^{2+} , Pb^{2+} , Cu^{2+} , Zn^{2+} , etc.), thereby creating an unfavorable environment for biological development. Moreover, these soils and the underlying spoil are highly susceptible to erosion by wind and rain due to their lack of internal cohesion and constitute dangerous sources of pollution due to their resulting pH, Eh, conductivity, and ion content of surface and subterranean runoff.

Attempts to restore the environmental health of areas occupied by mine soils [3,4] generally treat them with organic waste together with large doses of mineral fertilizer. Replenishment

of soil C and N, which is essential for sustainable revegetation of mine soils, may be accelerated by the use of organic amendments [5]. Several studies also reported that the application of ashes favors the reclamation of acid mine spoils, although is necessary to consider their low N and P content, high salinity, and potentially toxic elements [6]. The organic waste supplies carbon and other biogenic elements in forms that pioneering fauna can in principle make available to plants by metabolization, thereby favoring the development of a soil that is structurally and otherwise suitable for the growth of fauna and flora [7,8]. However, both mineral fertilizer and industrial, agricultural, and domestic waste can contain levels of heavy metals, e.g., lead, chromium, nickel, cadmium, and arsenic [9], that, though possibly acceptable when applied to normal soils, are unacceptable when applied to soils that, like mine soils, can themselves have high heavy metal content. Such waste should therefore be used for mine soil amendment only if the soil characteristics in regards to the sorption—and desorption—of heavy metals are known. For example, depending on these characteristics, it may be desirable for organic waste and mineral fertilizers to be applied together with natural soil components (clays, ox-

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ides, etc.) that can immobilize heavy metals in forms that will not be taken up by organisms. In this respect it should also be borne in mind that although experiments performed to determine sorption and desorption characteristics may be prolonged until equilibrium is reached, predictions of heavy metal fixation and transport in a dynamic environment should not assume equilibrium conditions [10,11].

Since the threat of heavy metal pollution posed by mine soils generally concerns more than one metal, studies of the sorption and desorption of heavy metals in these soils should take into account competition between metals for binding sites. However, there have in fact been few studies of the simultaneous sorption of more than one heavy metal by any kind of soil [10,12–15], and in the case of mine soils we know of none. More has been published on the simultaneous sorption of several heavy metals by isolated silicate minerals [16], Fe, Al, and Mn oxides [14], humus [17], and organic and inorganic colloids [18–22]; but, as in the case of whole soils, results have been highly contradictory as regards the order in which different metals are sorbed and desorbed. In fact, the very relevance of competition between metals has been controversial, possibly as a result of (i) the failure to test the importance of competition of cations in many studies; (ii) the confounding of ionic strength effects by covarying cation concentrations; (iii) the dependence of competing cation effects on pH and loading; and (iv) differences among cations in their competitive ability and concentration effects. For the purposes of ranking metals according to the extent of sorption when in competition, a useful parameter appears to be K_d , the distribution coefficient for partition of the ion between soil and soil solution; on the basis of K_d values it is possible to calculate the mobility and fate of competing metals in the soil [23–25].

The main goals of this study were to evaluate the competitive sorption and desorption of Cd, Cr, Cu, Ni, Pb, and Zn, in mine soils from two mines in the province of A Coruña (Galicia, northwest Spain), to establish the selectivity sequences of sorption and desorption of these metals in the soils by means of distribution coefficients and to evaluate the different soils' capacity for sorbing and retaining the group of metals that is being introduced into the soils through treatment with sewage sludge and coal ash.

2. Materials and methods

2.1. Soils

The mine soils studied (all Anthropogenic Regosols) [26] are located at the former copper mine at Touro (Spain), from which road material is currently extracted, and at the lignite mine at Meirama. The spoil at Touro consists mainly of amphibolites, chalcopryrite, limonite, garnets, and iron and copper sulfides. Samples of two soils were taken at this mine: TE, the soil of a mine tailing that was laid in 1986 but is only now undergoing initial recovery efforts involving treatment with sewage sludge and mineral fertilizers; and TB, the virtually vegetationless soil of an area that was formerly a sedimentation lagoon

Table 1
Characteristics of the soils studied (means of triplicate determinations)

	TE	TB	MH	MI	ME
Texture (%)					
Sand	75a	43d	49bc	48c	52b
Silt	17d	40a	34b	35b	27c
Clay	8c	17b	17b	17b	21a
Clay mineralogical analysis (%) (in clay fraction)					
Kaolinite	59.6d	40.8e	94.9a	70.8c	90.6b
Chlorite	nd	21.1a	nd	nd	nd
Quartz	nd	13.8a	nd	nd	nd
Smectite	nd	nd	2.8c	10.6a	4.3b
Gibbsite	5.3b	7.8a	nd	1.8c	nd
Goethite	6.2b	7.4a	nd	nd	nd
Mica	24.6a	8.9c	2.4e	16.9b	5.1d
Talk	4.2a	nd	nd	nd	nd
Acidity and effective cationic exchange capacity					
pH	3.62e	4.78c	6.11a	4.66d	5.71b
CECe cmol kg ⁻¹	9.47a	6.02d	6.84b	6.03d	5.34c
Organic matter (g kg ⁻¹)					
O.M. (organic matter)	0.83c	15.04a	0.64c	4.08b	0.80c
Oxides content (g kg ⁻¹)					
Aluminum oxides	2.57b	21.1a	1.70d	1.52d	2.11c
Iron oxides	2.44c	3.62a	1.43d	3.15b	1.61d
Manganese oxides	0.1b	1.20a	0.03d	0.07c	0.02d
Density (mg m ⁻³)					
Bulk density	0.84bc	1.65a	0.60c	1.56a	0.96b
Density	2.77a	2.24d	2.50c	2.64b	2.66ab
Porosity (%)					
	70a	26d	76a	41c	64b

Note. Values of the same parameters followed by a different letter are significantly different ($p < 0.05$). nd: not detected.

(where spoil from the flotation plant was deposited) and was subsequently covered with a mixture of wood ash and coal ash.

At Meirama, samples of three soils were taken: MI, a shallow soil developed over a mine tailing laid in 2000, in which year its revegetation was also begun; MH, from an area of older mine tailing now converted in wetland; and ME, from a 10-yr-old tailing.

For each of the above five soils, five samples of the top 30 cm were taken using an Eijkelkamp sampler and were pooled in the field. The pooled samples were transported to the laboratory in polyethylene bags, air dried, passed through 2-mm-mesh sieves, and homogenized with a Fritsch Laborette 27 rotary sample divider.

Profile description, physicochemical characteristics, and heavy metal contents of topsoils and limiting factors for reforestation of these mine spoils have been discussed in two previous papers [2,27]. Soil characteristics are summarized in Table 1.

2.2. Sorption and desorption experiments

As the retention mechanism of metal ions at soil surfaces is often unknown, the term “sorption” is preferred [28], which in general involves the loss of metal ions from an aqueous to a contiguous solid phase and consists of three important processes: sorption, surface precipitation, and fixation [29].

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