

Metal Uptake by Spontaneous Vegetation in Acidic Mine Tailings from a Semiarid Area in South Spain: Implications for Revegetation and Land Management

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Abstract Tailings are frequently a source of pollution in mining areas due to the spread of metals from their bare surfaces via wind or runoff water. Phytostabilization is an interesting and low-cost option to decrease environmental risks in these sites. In this study, an acidic mine tailing (pH 3–4) located in a semiarid area in Southeast Spain and the spontaneous vegetation which grow on were investigated. Soil samples were taken to characterize metal contamination, and three plant species, *Lygeum spartum*, *Piptatherum miliaceum*, and *Helichrysum decumbens*, were sampled in order to determine plant uptake of metals. The rhizosphere pH of *H. decumbens* was measured to be 6.7, which was significantly higher than the bulk soil (pH 3). The electrical conductivity values were around 2–5 dS m⁻¹. Total metal concentrations in soil were high

(9,800 mg kg⁻¹ for Pb and 7,200 mg kg⁻¹ for Zn). DTPA-extractable Zn and Pb were 16% and 19% of the total amount, respectively. The three selected plant species accumulated around 2–5 mg kg⁻¹ Cu in both shoots and roots. Zn concentration was 100 mg kg⁻¹ in *P. miliaceum* roots. DTPA-extractable Zn was positively correlated with Zn plant uptake. These plant species demonstrated to grow well in acid tailings taking up only low concentrations of metals and therefore are good candidates to perform further phytostabilization works.

Keywords Mining contamination · Semiarid climate · Phytostabilization · Plant uptake

1 Introduction

Soil contamination is of increasing worldwide concern because of its effects on environmental health, loss of soil productivity, and socioeconomic impacts. Mining activities are known to be responsible for many soil contamination sites (Conesa et al. 2006; Wei et al. 2009). Current mining activities in most developed countries are carried out within environmental laws and restoration works are normally achieved. However, mining operations which ceased some decades ago and worked under the absence of public environmental regulations left wastes on the ground which still imply

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a source of contamination and environmental risks for their surroundings (Conesa et al. 2006).

The Cartagena–La Unión Mining District (0–400 m above sea level; 37°37'20" N, 0°50'55" W–37°40'03" N, 0°48'12" W, area 50 km²) is located in Southeast Spain (Fig. 1). This semiarid region is characterized by Mediterranean climate with 18°C annual average temperature and 250–300 mm annual rainfall (which mainly occurs during spring and autumn). Mining in the Cartagena–La Unión District finished in 1991 after more than 2,500 years and was based on sulfides such as galena and sphalerite. Most of the wastes generated were disposed on the ground, forming tailings which currently cover approximately 160 ha (Martínez-Orozco et al. 1993). Some of these mine tailings have been restored by capping with non-polluted material and revegetation. However, short intensive rainfalls and wind have affected the restoration works, posing risk nearby agricultural and urban areas (Conesa et al. 2010). Water erosion was recognized to be the main erosion factor in this area (Conesa et al. 2006), although in the long term, wind erosion was also considered a risk. García-García (2004) found that dust layer deposits of more than 50 cm on soils surrounded some local tailings.

Capping tailings with non-polluted materials may reduce environmental hazards, but it is expensive and impractical due to the extent of the areas which are covered by tailings in the Cartagena–La Unión Mining District. In situ stabilization by chemical amendments is not favored because of their low durability and the need to carry out regular inspections, but they may result useful for the temporary stabilization of tailings prior to revegetation (Tordoff et al. 2000; Conesa et al. 2007a). The establishment of a stable plant cover is considered a suitable option to get long-term reclamation (Simon 2005). Former mining lands are considered to be suited for noninvasive and low-cost remediation technologies due to the huge areas to be treated. Among the available remediation technologies, phytostabilization

represents a promising tool (Mendez and Maier 2008a) and should include, according to Mendez and Maier (2008b): (a) the identification of regional and climatic specific plants which show low metal uptake in shoots and (b) the minimum plant-required amendments (compost, fertilizer, irrigation). This option emphasizes stabilizing the soils and not decontaminating them since mining soils are so heavily polluted that removal of metals using plants would take an unacceptable amount of time (Pratas et al. 2005).

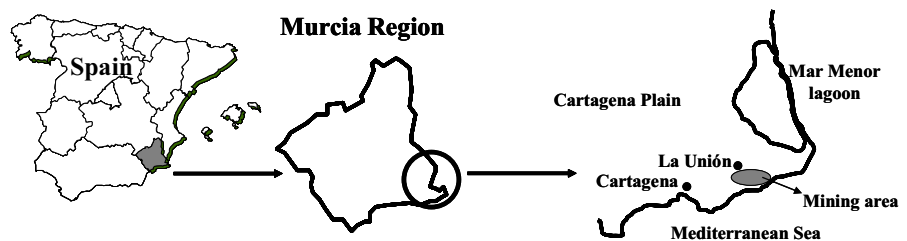
Some previous studies have shown the presence of metal-tolerant plant species in tailings from the Cartagena–La Unión Mining District (Conesa et al. 2006, 2007b). Most of these tailings showed neutral pH, and therefore, plant adaptation was favored in comparison to acid ones.

The aim of this study was to characterize the metal content in an acidic tailing in the Cartagena–La Unión Mining District and the metal uptake of the plants which spontaneously grow on, with the goal of setting a plant species catalog for further phytostabilization purposes. To this end, total and bioavailable metals were determined.

2 Materials and Methods

Sampling was conducted at a mine tailing situated close to La Unión town (SE Spain) (110 m a.s.l.; 37° 37'20" N, 0°50'55" W–37°40'03" N, 0°48'12" W). An experimental plot design (three plots of 8×8 m²) was performed on zones where spontaneous vegetation grew (Fig. 2). The percentage of plant covering was low and most of the area was bare. Soil samples were taken from the first 20 cm coincidental with the plot vertex. Three samples of each plant species (shoots and roots) were also taken, including their corresponding rhizospheric soils: *Piptatherum miliaceum* (L.) Cosson, *Lygeum spartum* L., *Helichrysum decumbens* (Lag.) Camb. Soil samples were air-dried,

Fig. 1 Location of the studied area



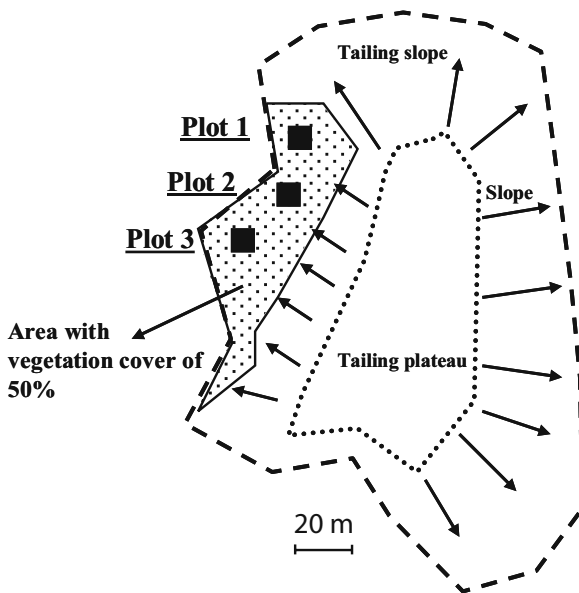


Fig. 2 Basic scheme of the tailing structure

sieved through 2-mm grain size, homogenized, and stored in plastic bags prior to laboratory analysis. The texture was determined using the Robinson pipette method combined with sieving. Equivalent calcium carbonate was determined using Bernard calcimeter. Electrical conductivity and pH in the 1:1 extract (soil/water) was determined using a conductivity meter Crison Micro CM 2200 and a pH-meter Crison Basic 20, respectively. Organic carbon was determined by the oxidation of organic matter using potassium dichromate, and total nitrogen was determined using the Kjeldahl method (Duchaufour 1970). The bioavailable metals were determined using extraction solutions containing 0.005 M DTPA, 0.01 M CaCl_2 , and 0.1 M triethanolamine (Lindsay and Norvell 1978) in 1:5 soil/DTPA solution ratio that was shaken for 2 h according to the recommendations of Norvell (1984) for the use of

DTPA in acidic and heavy metal-polluted sites. Copper, Pb, and Zn were measured in the resulting filtered extracts by flame atomic absorption spectroscopy (UNICAM 969 AA spectrometer). Total Cu, Pb, and Zn concentrations were determined using a $\text{HNO}_3/\text{HClO}_4$ digestion at 210°C for 1.5 h. Solution concentrations were measured using flame atomic absorption spectroscopy (UNICAM 969 AA spectrometer). This method was referenced using the CRM027-050 Certified Material (Resource Technology Corporation, USA). We obtained recoveries of 112% for Cu, 99% for Zn, and 94% for Pb.

Plants were carefully washed with distilled water and then dried at 65°C for 72 h. Shoots and roots were separated prior to grinding (Janke & Kunkel IKA® Labortechnik A-10). For each sample, 0.5 g was incinerated prior to a redilution using concentrated nitric acid. Metal concentrations (Cu, Pb, and Zn) were measured by inductively coupled plasma mass atomic spectroscopy (ICP-MS 4500 Agilent Technologies).

All the statistical analysis (ANOVA with LSD) was carried out with SPSS 14.0.0 (SPSS, Chicago, IL, USA). Differences at the $P < 0.05$ level were considered significant.

3 Results and Discussion

3.1 Soil Parameters

Soil analyses (Table 1) showed that the tailing samples were strongly acidic (pH 3–4). Rhizospheric soil samples from *H. decumbens* showed significantly higher pH (6.7) than bulk soil samples. Root exudates and related biogeochemical processes may have modified pH conditions around *H. decumbens* roots. Rhizosphere of some plant species may generate pH

Table 1 Soil properties from bulk samples ($N=12$) and rhizospheric samples ($N=3$)

Samples	pH H_2O	pH KCl	EC (dS m^{-1})	CaCO_3 (%)	Clay (%)	Silt (%)	Sand (%)
Bulk soil	4.1 (0.43)a	3.7 (0.41)a	5.5 (1.0)a	<2	7.0 (1.1)	25 (4)	68 (3.8)
Rhizosphere <i>H. decumbens</i>	6.7 (0.2)b	6.2 (0.2)b	2.4 (0.8)a	<2	n.m.	n.m.	n.m.
Rhizosphere <i>L. spartum</i>	3.7 (0.6)a	3.3 (0.6)a	2.5 (0.8)a	<2	n.m.	n.m.	n.m.
Rhizosphere <i>P. miliaceum</i>	4.9 (0.4)a,b	4.4 (0.4)a,b	4.1 (0.7)a	<2	n.m.	n.m.	n.m.

Numbers between brackets are standard errors. Different letters indicate significant differences ($P < 0.05$)

EC electrical conductivity, n.m. not measured

gradients in relation to bulk soil mainly due to the production and consumption of protons by plant roots to equilibrate for unbalanced cation–anion uptake (Hinsinger et al. 2003). Chaignon et al. (2002) observed that in the rhizosphere of oilseed and tomato plants, acidification occurred in a calcareous soil while alkalization occurred in an acidic soil. These pH changes and other rhizosphere-induced changes have influence in metal bioavailability and may condition, therefore, the growing of a site-adapted microflora. The pH changes were not so remarkable for *L. spartum* and *P. miliaceum* rhizospheres.

Due to the low pH, calcium carbonate was not detected in any sample. No significant differences were observed in relation to electrical conductivity values between bulk soils and rhizospheres. Both were in the range of slightly saline soils (2.5–5.0 dS m⁻¹). Sand was the most abundant fraction (70%), resulting in a sandy loam texture. Therefore, low water retention capacity is expected.

Organic carbon and total nitrogen (data not shown) showed percentages below 0.3% and 0.02%, respectively. It is known that polluted mining soils are characterized by low nutrient content and adverse conditions for plant growing (Wong 2003). In these conditions, the competition for main nutrients such as nitrogen or phosphorus represents a limiting factor for plants and soil microorganisms (Unterbrunner et al. 2007).

3.2 Total and DTPA Heavy Metal Concentrations

High total heavy metal concentrations were found (Table 1), especially for Pb and Zn. Lead showed maximums of around 15,000 mg kg⁻¹ and Zn 9,900 mg kg⁻¹.

In Spain, there are no national thresholds for heavy metals in soils, and guidelines must be established at a

regional scale based on the corresponding geochemical backgrounds (BOE 2005). In Murcia Region, official thresholds have been not yet established, but some scientific works have reported geochemical backgrounds and environmental guidelines. Soils from the nearby Cartagena plain (Fig. 1) were shown to have geochemical backgrounds of 12.6 mg kg⁻¹ Cu, 9 mg kg⁻¹ Pb, and 41 mg kg⁻¹ Zn (Martínez-Sánchez and Pérez-Sirvent 2007). However, reference levels in the soils from the mining district should be established separately due to its different geochemical basis. Some agricultural soils located around La Union town have shown concentrations of 21 mg kg⁻¹ Cu, 500 mg kg⁻¹ Pb, and 900 mg kg⁻¹ Zn (Conesa et al. 2010). In comparison with the thresholds settled by other European countries such as the Netherlands, Pb and Zn concentrations exceeded by far the intervention levels (530 mg kg⁻¹ for Pb and 720 mg kg⁻¹ for Zn; M. H.S.P.E. 2000). Moreover, Pb and Zn concentrations obtained by Simon et al. (1999) (360 and 650 mg kg⁻¹, respectively) in the soils affected by the toxic spill in Aznalcollar (South Spain in 1998) were widely surpassed.

A relatively high percentage of the total metal content in bulk soil samples was extracted by DTPA (Table 2) especially for Pb (6.5%) and Zn (2%). *P. miliaceum* rhizosphere soil samples showed higher significant DTPA-extractable Cu and Zn concentrations compared to bulk soil. It is known that geochemical parameters within the rhizosphere may affect metal solubility in relation to bulk soil (Wang et al. 2002; Wenzel 2009).

3.3 Plant Metal Uptake and Implications for Remediation

The spontaneous plant colonization in mine tailings is usually slow since the physicochemical characteristics

Table 2 Total and DTPA-extractable metal concentrations for bulk ($N=12$) and rhizosphere soil samples ($N=3$)

Metal	Bulk soil		Rhizosphere soils		
	Total concentration		<i>H. decumbens</i> DTPA-extractable	<i>L. spartum</i>	<i>P. miliaceum</i>
Cu	450 (50)	3.0 (0.45)a	4.2 (0.7)a,b	3.7 (1.3)a,b	7.3 (2.6)b
Pb	9,900 (790)	640 (180)a	1,100 (250)a	840 (700)a	1,600 (790)a
Zn	7,200 (530)	150 (40)a	270 (26)a,b	130 (97)a,b	330 (100)b

Numbers between brackets are standard errors. Different letters indicate significant differences ($P<0.05$) among DTPA extractable concentrations. Data are in milligrams per kilogram

of these sites are not suitable for most of the plant species. Nevertheless, some of the tolerant plant species can spread easily in these environments due to the lack of competitors (Macnair 1987). Conesa et al. (2007c) reported a complete characterization of local plant communities which grow in local tailings, showing that most of the species belong to *Gramineae* (*P. miliaceum*, *L. spartum*, *Phragmites australis*) and *Compositae* (*Dittrichia viscosa*, *H. decumbens*, *Phagnalon saxatile*, *Senecio vulgaris*, *Sonchus tenerrimus*).

The heavy metal concentrations taken up by roots and shoots are shown in Fig. 3. Copper concentrations in shoots and roots were in the same range for the three plant species studied ($\sim 5 \text{ mg kg}^{-1}$). In relation to Pb and Zn, *P. miliaceum* and *L. spartum* showed higher concentrations in roots in relation to shoots, while the opposite occurred for *H. decumbens*. The highest Zn (57 mg kg^{-1}) and Pb (31 mg kg^{-1}) concentrations in shoots were obtained for *H. decumbens*. However, *P. miliaceum* showed the highest Zn and Pb concentrations in roots: 46 mg kg^{-1} Pb and 106 mg kg^{-1} Zn. Metal uptake by *L. spartum* and *P. miliaceum* were within the thresholds that Chaney (1989) proposed in relation to dry foliage: $3\text{--}20 \text{ mg kg}^{-1}$ Cu, $15\text{--}150 \text{ mg kg}^{-1}$ Zn, and $2\text{--}5 \text{ mg kg}^{-1}$ Pb. However, Pb concentration in *H. decumbens* shoots surpassed the aforementioned thresholds.

The DPTA-extracted Zn had a positive correlation ($r=40$) with the metal uptake in the plant species studied. However, no positive correlation was found for DTPA–Pb and DTPA–Cu. Sims and Johnson (1991) also found that DTPA-extractable Zn was a better indicator of Zn plant uptake. In any case, the results of DTPA-extracted metals in the acid metal-polluted samples must be carefully interpreted (National Research Council 2003).

Plant metal accumulation is often expressed as a bioconcentration factor (BF). It is calculated as the metal concentration in dry plant tissue/metal concentration in soil quotient (Mattina et al. 2003). In our study, the BF for all metals was below 0.1. McGrath and Zhao (2003) considered <0.2 as normal for plants growing on polluted materials. The accumulation factor (AF; Fitz and Wenzel 2002), or also called translocation factor (Mattina et al. 2003), is the ratio between the concentration of metals in shoots and roots and defines the effectiveness of the plant to translocate the metals to the shoots. Tolerant plants

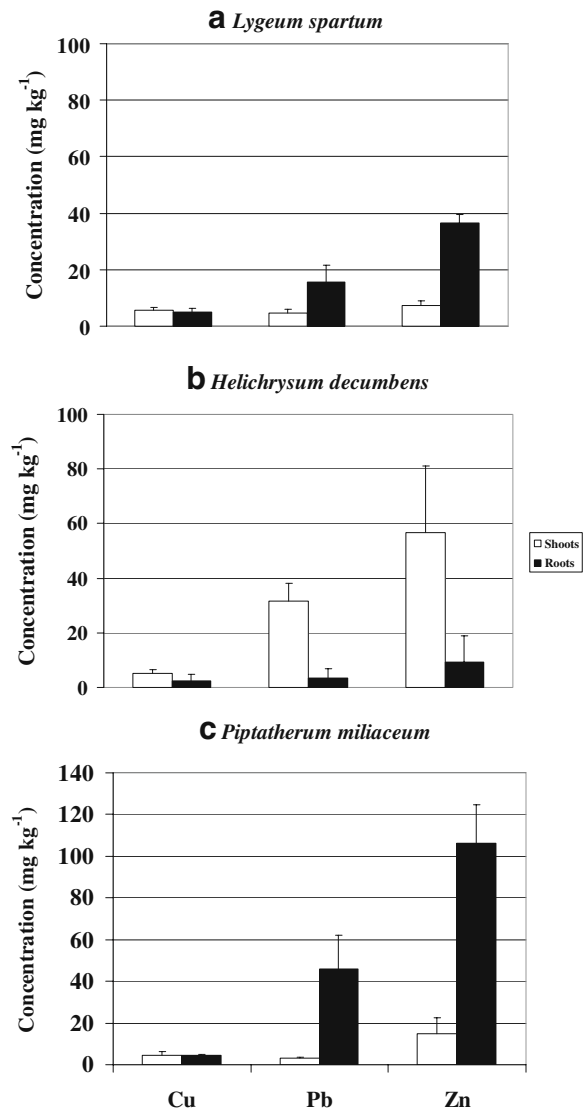


Fig. 3 Copper, Pb, and Zn uptake in *L. spartum* (a), *H. decumbens* (b), and *P. miliaceum* (c). Black bars on columns are standard error ($N=3$)

have values $\ll 1$ and hyperaccumulators $\gg 1$. In our case, *L. spartum* had AF values of 1.1 for Cu and 0.2–0.3 for Pb and Zn. For *P. miliaceum*, the accumulation factors were 1 for Cu and <0.2 for Pb and Zn. Finally, *H. decumbens* showed an AF of 0.6 for Cu, 2.5 for Pb, and 1.7 for Zn. From these data, we can confirm that the metal translocation is in general low, except for *H. decumbens* and Pb. This fact should be taken into account in order to prevent the entry of metals in the food chain.

Previous studies in this area have shown that *L. spartum* grew on other acidic tailings (Conesa et al. 2006). However, Conesa et al. (2007a) showed in a pot experiment using homogenized acidic tailings that *L. spartum* showed low rate survival and high metal uptake ($\sim 50 \text{ mg kg}^{-1}$ Cu, $\sim 70 \text{ mg kg}^{-1}$ Pb, and $\sim 4,000 \text{ mg kg}^{-1}$ Zn). The latter authors hypothesized that plants gradually invade the tailings from areas where soil conditions are less aggressive for initial plant establishment (i.e., better water retention) and grow selectively into the soil, taking advantage of the heterogeneity in the distribution of metals. Our study may support this hypothesis. This plant species (*L. spartum*) is widespread in the Mediterranean area and is able to tolerate extreme conditions of aridity, high temperatures, deficiency of nutrients, and salinity (Díaz and Honrubia 1993). Plant survival may be facilitated by the humidity of the air, which is high due to the proximity of the coast. This is important for the development of the local vegetation, mitigating the scarcity of rains with the formation of fogs and cryptoprecipitations (Martos-Miralles et al. 2001).

P. miliaceum has been cited in some mining areas from southeast Spain (Conesa et al. 2006; Melendo et al. 2002) and may have potential interest as pasture. Although the concentrations of metals taken up through roots and accumulated in plant tissues in this area have been shown to be low, metal-rich dust particles adhering to the surfaces of plants or direct soil ingestion must also be taken into account in order to prevent cattle to graze in these areas.

4 Conclusions

High heavy metal concentrations were found in a mine tailing from the Cartagena–La Unión Mining District. The decisions about potential land uses should consider the high metal pollution which surpasses several official thresholds.

Three plant species, *H. decumbens*, *L. spartum*, and *P. miliaceum*, spontaneously colonize parts of the tailings in spite of the high DTPA metal-extractable concentrations and low pH. These plant species do not uptake high concentrations of metals ($<60 \text{ mg kg}^{-1}$ in shoots for Cu, Pb, and Zn), providing a good tool to effect the surface stabilization of the tailing with low risk of affecting the food chain.

The plant metal communities which spontaneously grew on acid tailings should be taken into account in order to carry out low-cost alternatives for the decreasing of the environmental risks associated to mining soils. Lime amendments to increase pH and organic nutrients to increase nutrient levels should be evaluated too.

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