

Trace element accumulation in plants from an aridic area affected by mining activities

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ABSTRACT

Trace element contamination has been a serious problem in the vicinity of abandoned mine sites. In the studied area, mining activities have produced great amounts of wastes, characterized by high trace elements content, acidic pH and minerals from supergene alteration. Trace elements have been dispersed, both downstream and downslope from the mine mainly due, mainly, to surface run-off. Trace elements hydric mobilization usually takes place during the rainy season, an important pollution route in the studied area. A wide range of techniques have been proposed to remediate soils polluted by trace elements. Among them, phytoremediation-based technologies could provide a long-lasting solution. The aim of this work was to determine trace element concentration in roots and leaves of five plant species (*Limonium carthaginiens*, *Arthrocnemum macrostachyum*, *Dittrichia viscosa*, *Glaucium flavum* and *Zygophyllum fabago*) growing in soils polluted by mining activities in order to determine which part of the plants accumulate heavy metals to the greatest extent. The metal concentrations in plants varied with plant species. Plant accumulation results showed that *Z. fabago* could act as an accumulator for Fe, and *A. macrostachyum* could accumulate in Fe, As and Mn. The other plant species collected from the mining sites were tolerant to trace element contamination.

Keywords: Trace Elements

Accumulator

Phytoremediation Translocation factor Soil contamination

1. Introduction

Mining activities generate a large amount of tailings that are generally deposited upon the soil surface. Tailings usually provide an unfavorable substrate for plant growth because of their low pH, high concentrations of trace elements and low nutrient content (Ha et al., 2011). Specifically, metalliferous mining and processing, including the dumping of wastes, often produce severe trace element pollution (Baker et al., 1994). Soils contaminated with trace elements usually lack established vegetation cover, either due to the toxic effects of pollutants or due to physical disturbance (Rotkittikhun et al., 2006). The contamination of soils due to the presence of toxic elements could result in negative consequences, such as damage of ecosystems and of agricultural productivity, deterioration of food chain, contamination of water resources, economic damage and serious human and animal health problems.

A wide range of techniques have been proposed to remediate soils polluted by trace elements. Soil clean-up technologies are often expensive, energy consuming and usually soil can rarely be used after the treatment. Some methods, however, may be considered a cost-effective solution for treating soils polluted by trace elements, and among them, phytoremediation-based technologies, may well provide a long-lasting solution, since they use plants to stabilize pollutants

from soils (Gardea-Torresdey et al., 1996; Hernández-Allica et al., 2006; Zhuang et al., 2005). Phytoremediation can be defined as the use of plants for containment, degradation or extraction of xenobiotics from water or soil substrates (USEPA, 2000).

The presence of vegetation reduces water and wind erosion, which may decrease the downward migration of contaminants into the groundwater. Furthermore, tolerant plants can help in the physical and chemical immobilisation of contaminants through root sorption or by inducing chemical retention in the soil (Prasad and Freitas, 2003). The best plants for phytoremediation purposes are native plants because of their greater chance of survival, growth and reproduction in environmental stress conditions than plants introduced from other environments (Adriano, 2001; Antonsiewicz et al., 2008).

There are several distinct groups of plant species according to their trace element accumulation capability (Baker, 1981): (a) excluders, which metal concentrations in the shoots are maintained at a low level up to a critical value across a wide range of soil concentrations; (b) accumulators, in which metals are concentrated in above-ground plant parts from low to great soil concentrations; and (c) indicators, in which the internal concentration reflects external levels. A particular sub-group within the accumulators is represented by the hyperaccumulators (Adriano, 2001; Brooks et al., 1977). Hyperaccumulators are plants commonly grown on metalliferous soils and able to complete their life cycle without any symptoms of metal phytotoxicity (Baker et al., 2000). The criteria to define hyperaccumulation for arsenic, lead, cobalt, copper and selenium are a

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concentration of 1000 mg kg⁻¹ or greater, on a dry leaf basis, where- as the threshold value for zinc and manganese hyperaccumulation is 10 000 mg kg⁻¹ (Baker and Brooks, 1989; Kabata-Pendias and Mukherjee, 2007). Cadmium hyperaccumulators are defined as values larger or equal to 100 mg kg⁻¹. Hyperaccumulators could be characterized by a shoot-to-root metal concentration ratio (transloca- tion factor, TF) of more than 1, whereas non-hyperaccumulator plants usually have greater metal concentrations in the roots than in the shoots. Several authors (Baker, 1981; Baker et al., 1994; Bech et al., 2012; McGrath and Zhao, 2003; Sun et al., 2008) included the bioac- cumulation factor (BCF) as an element for classification as a hyperac- cumulator species. The BCF refers to the plant metal concentration and the soil metal concentration ratio. A BCF value higher than 1 may be taken to indicate that the plant could act as a hyperaccumula- tor of trace elements.

The aim of this work was to determine the trace element concen- tration in roots and leaves of plant species growing in soils polluted by mining activities, in order to determine which part of the plants accumulate trace elements to the greatest extent. In addition, the study was carried out to compare the trace element accumulation capacity of selected plant species growing on contaminated soils and to assess changes in the concentration of soil-available metals. Five plant species (*Limonium carthaginiens*, *Arthrocnemum macrosta- chyum*, *Dittrichia viscosa*, *Glaucium flavum* and *Zygophyllum fabago*) were studied, and the transfer factor (TF) and the bioconcentration factor (BCF) were calculated.

2. Materials and methods

2.1. Study area and sample collection

The studied area is located in the southeast of Murcia Region (Spain). It is a coastal mountain chain with an approximate extension of 50 km². This zone was one of the main mining districts in the southeast of Spain (Martínez-Sánchez et al., 2008).

The annual average temperature is 17 °C and precipitation does not exceed 300 mm, with occasional torrential rainfall which normal- ly occurs between the end of summer and autumn. Perennial streams do not exist, and surface water consists of ephemeral streams, which are only operational during rainfall events. The stream flow is flashy and a significant amount of sediments are transported after long- lasting rainfalls.

As a result of mining activities, the studied area shows a large number of contaminations sources, formed by mining steriles, waste piles and foundry residues. The extractive activity has resulted in var- iations in the drainage network with modification of natural slopes, and favouring erosion processes, increased by the scarce vegetation present (García-Lorenzo et al., 2012).

For this study, 15 sediment samples were collected. Sampling was carried out in an area where the dispersion of trace elements and their zone of influence are being studied and where it had been ob- served that spontaneous revegetation had occurred in zones that had been affected by mining activity.

In addition to sediments, samples of five plant species, which nat- urally grow in this area, (*L. carthaginiens*, *A. macrostachyum*, *D. viscosa*, *G. flavum* and *Z. fabago*) were collected. In some of the sampling points *Arthrocnemum macrostachyum* and *L. carthaginiens* were not studied because of their specific requirements. In total, 15 samples of *D. viscosa*, *G. flavum* and *Z. fabago*, 10 of *L. carthaginiens* and 5 of *Arthrocnemum macrostachyum* were collected. *D. viscosa*, *Z. fabago* and *L. carthaginiens* usually grow in semiarid areas because of their low nutritional needs. They are resistant and perennial. *Arthrocnemum macrostachyum* shows similar behaviour to the above species but usually grows in wet areas. It is salt-resistant, grows slowly and sometimes forms groups. The main characteristic of *G. flavum* is its short life cycle.

2.2. Sample preparation and chemical analysis

Superficial sediment samples (0–10 cm) were collected from four subsampling points in a circle about 50 cm around the plant species and homogenised. Homogenized samples were air-dried at room temperature and sieved through a 2 mm screen for general analytical determinations.

The pH was measured in a 1:5 suspension of soil in high quality water obtained by means of a Milli-Q system. Electrical conductivity (EC) (dS m⁻¹ at 25 °C) was measured in the extracts obtained by filter- ing the 1:5 suspensions through a 0.45 µm cellulose acetate disk filter. Equivalent calcium carbonate (%) was determined by the volumetric method using a Bernard calcimeter. Organic carbon was determined by sulfochromic oxidation (Nelson and Sommers, 1982), according to the NFX 31-109 standard (AFNOR, 1993).

A semiquantitative estimation of the mineralogical composition of the samples was made by X-ray Diffraction (XRD) analysis using Cu-Kα radiation with a PW3040 Philips Diffractometer. X-powder software (Martín, 2004) was used to analyse the X-ray diffraction diagrams obtained by the crystalline powder method. The powder diffraction file (PDF2) database was used for peak identification, taking into account that the determination of minerals from soils by XRD analysis is not accu- rate below a limit of 5% of the total weight in a sample (depending on the crystallinity of individual minerals).

To determine the total trace element content, the sediments were first ground to a fine powder using a zirconium ball mill. Aliquots (0.1 g) of sediment samples were placed in Teflon vessels and 5 ml of concentrated HF acid solution, 200 µl of concentrated HNO₃ acid solution and 5 ml of water were added. When digestion in the

Table 1
Analytical characteristics in sediment samples.

Sample	pH	EC (dS m ⁻¹)	CaCO ₃ (%)	OM (%)	Zn (mg kg ⁻¹)	Pb (mg kg ⁻¹)	Cd (mg kg ⁻¹)	Fe (%)	As (mg kg ⁻¹)	Cu (mg kg ⁻¹)	Mn (mg kg ⁻¹)
S1	8.6	1.4	15.3	1.6	5455	2130	42.1	6.49	1235	299	1992
S2	5.0	7.4	bdl	bdl	8984	3989	88.3	25.14	752	142	4053
S3	7.4	2.8	10.1	bdl	3153	1241	123.1	26.11	669	123	3498
S4	7.8	2.9	1.0	bdl	7958	3621	34.3	19.23	454	124	3537
S5	6.9	1.4	1.0	1.0	7062	4741	19.1	24.53	561	127	3767
S6	6.6	1.7	1.3	bdl	7239	6757	20.4	24.23	530	145	3378
S7	5.3	0.5	bdl	1.3	4000	13500	15.1	19.25	644	185	1390
S8	4.2	2.5	bdl	1.0	2388	554	10.7	4.29	3115	110	6686
S9	8.1	0.5	13.3	1.2	1210	1936	11.2	7.30	304	131	1447
S10	7.5	0.2	bdl	bdl	6398	709	65.5	38.59	554	68	8350
S11	7.4	3.5	bdl	bdl	6600	1003	74.3	50.58	868	147	10806
S12	7.5	0.2	1.2	bl d	135	63	6.3	16.11	40	171	864
S13	7.9	0.4	28.7	0.5	1494	538	12.1	4.63	327	129	1584
S14	7.7	0.7	bl d	1.3	3670	2041	14.8	11.83	563	119	1528
S15	7.5	0.5	4.9	0.7	3049	3002	14.5	18.99	664	150	1735

Table 2
Average mineralogical composition (%) of the sediment samples in the study area.

Sample	Phy	Fsp	Qtz	Gp	Jar	Hem	Gt	Cal	Dol	Aka	Sid	Gol	Am
S1	27	44	6		18	5							1
S2	35	2	28	9	10	6				11			1
S3	28	2	29		6	7		14		12			2
S4	13	3	34	13	8	3		6		14		4	2
S5	12	4	30	14	11	2	4			14		6	3
S6	15	2	33	9	11	3	6			12		7	2
S7	15	2	32	7	14	4	8			11		6	2
S8	3	5	34	32	11	2				13			1
S9	30	13	39			9				7			2
S10	31		15	7	9	5				8	22		3
S11	20		9	4	6	9				1	42		1
S12	3		3					2	89				3
S13	28	7	40					16	7				3
S14	53	7	22			7						8	2
S15	30	5	47			4		11					4

Phy: Phyllosilicates; Fsp: Feldspars; Qtz: Quartz; Jar: Jarosite; Gt: Goethite; Aka: Akaganeite; Cal: Calcite; Dol: Dolomite; Gol: Goldichite; Gp: Gypsum; Hem: Hematite; Sid: Siderite; Am: Amorphous.

microwave oven was complete, the solutions were transferred to a volumetric flask and brought to 50 ml.

Fresh vegetable samples were separated into root and aboveground biomass and then lyophilized. 200 mg of lyophilized vegetal tissue was placed in Teflon vessels with 3 ml of water, 2 ml of concentrated H₂O₂ and 5 ml of concentrated HNO₃ acid solution. When digestion in the microwave oven was complete, the solutions were transferred to a volumetric flask and brought to 50 ml.

The zinc, iron and manganese content was determined by flame atomic absorption spectrometry (FAAS) using a Perkin-Elmer 1100B Atomic Absorption Spectrophotometer. The lead, cadmium and copper content was determined by electrothermal atomization atomic absorption spectrometry (ETAAS) using a Unicam 929 AAS spectrometer. The arsenic content was measured by atomic fluorescence spectrometry using an automated continuous flow hydride generation spectrometer (PSA Millenium Merlin 10055 for arsenic). The reliability of the results was verified by analysing a standard reference material (SRM 2711 Montana Soil, NCS DC 73320 and NCS DC 73323 for soil samples and SRM 1515 Apple leaves and SRM 1572 Citrus leaves for plant samples). Spikes, duplicates and reagent blanks were also used as part of quality assurance/quality control.

Bioconcentration factor (BCF) and translocation factor (TF) are important parameters in heavy metal uptake studies (Marchiol et al., 2004; Bose et al., 2008). In order to evaluate the phytoextraction potential of the selected plants, the bioconcentration factor ($BCF = C_{\text{roots}}/C_{\text{soil}}$)

and the translocation factor ($TF = C_{\text{leaves}}/C_{\text{roots}}$), where C represents the metal concentration, were calculated.

3. Results and discussion

3.1. Soil properties and metal concentrations

The general characteristics of the soil samples are given in Table 1. Most samples showed neutral pH values, with an average value of 7.1, although S2, S7 and S8 showed acidic pH values. All the samples presented a very low organic matter percentage (mean value of 0.8%), low soluble salt content and low calcium carbonate percentage. The average value for calcium carbonate was 9.7% and the highest value was 28.7%, in S13. The average value for Zn was 4586 mg kg⁻¹, 3073 mg kg⁻¹ for lead, 40 mg kg⁻¹ for cadmium, 19% of iron, 806 mg kg⁻¹ for arsenic, 129 mg kg⁻¹ for copper and 3566 mg kg⁻¹ for manganese.

The plant species selected grew in poorly developed soils formed by materials from surrounding mining areas, carbonate materials and phyllites. The mineralogical analysis showed that the main minerals were quartz and phyllosilicates, while the minority minerals were products of mining activities (iron oxides and hydroxides, siderite, jarosite and gypsum), calcite and feldspars (Table 2).

Fig. 1 summarises the conceptual model of main processes affecting materials in areas with mining influence in the studied area. The most important weathering processes in the studied area are oxidation–reduction, hydration, hydrolysis and dissolution–precipitation. In the studied sediments, mining activities have produced intense morphological changes, modifying drainage network and natural slopes, taking place remnant erosion processes. In addition, torrential rainfall and the scant vegetation present in the studied area amplify the erosive process (Fig. 2). During the transport phase, dissolution and hydration processes acquire more importance and weathering products are mobilized as dissolved or particulate phases. When kinetic energy of the system diminishes, sedimentation processes happen. Dissolved and particulate materials are deposited, taking place heterogeneous chemical reactions (García-Lorenzo et al., 2012).

3.2. Metal concentration in plants and bioconcentration and translocation factor determination

Table 3 summarises the average trace element content in roots and leaves of selected species and Table 4 showed the bioconcentration (BCF) and translocation (TF) factor results for collected samples. In *L. carthagenis* and *Z. fabago* leave concentration was lower than root concentration. Since all samples showed BCF and TF lower than unity, these species could be considered as tolerant.

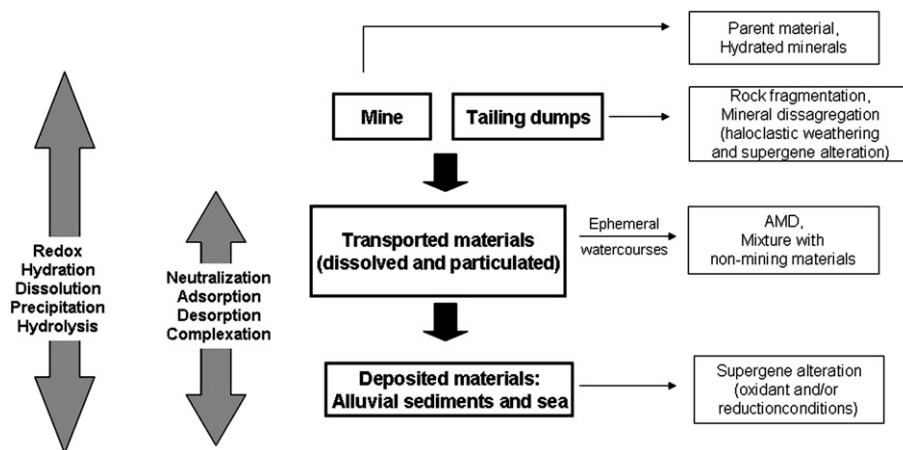


Fig. 1. Main processes affecting materials in areas with mining influence.



Fig. 2. Pictures of the studied area.

Iron content in leaves of *G. flavum* was higher than those found in roots of this species. For the other elements, trace element content was higher in roots. BCF was lower than unity for all the studied elements in all samples, while TF was higher than unity for iron. *G. flavum* seems to have a higher capacity for iron translocation from roots to shoots.

In *Artrocnemum macrostachyum*, BCF was higher than unity for zinc and cadmium, showing that *A. macrostachyum* seems to have higher capacity for zinc and cadmium absorption from soil to roots. In addition, arsenic, manganese and iron have been translocated to the aerial biomass (TF value higher than unity).

Finally, *D. viscosa* showed higher concentrations in leaves than in roots for Pb and iron. For the other elements root concentration was higher than leaves concentration. BCF in this species was lower than unity for all the studied elements, and this species could be considered as a tolerant plant for all the studied elements.

4. Conclusions

The studied area is strongly affected by mining activities, and superficial materials have suffered weathering processes, releasing a great amount of trace elements and sulphates, which are transported

Table 3
Mean trace element content in roots and leaves (mg kg^{-1}) of selected species (\pm standard deviation).

<i>Limonium carthaginiens</i>							
	Zn	Pb	Cd	Fe	As	Cu	Mn
Roots	392 \pm 27	138 \pm 5	2.9 \pm 0.8	909 \pm 51	8.4 \pm 1.1	11.1 \pm 0.3	75.1 \pm 1.7
Leaves	297 \pm 22	107 \pm 7	2.4 \pm 0.4	807 \pm 32	2.7 \pm 0.5	8.1 \pm 0.6	28.6 \pm 0.4
<i>Glaucium flavum</i>							
	Zn	Pb	Cd	Fe	As	Cu	Mn
Roots	1406 \pm 38	1232 \pm 21	7.6 \pm 2.2	241 \pm 24	42.8 \pm 1.6	25.1 \pm 2.2	296.9 \pm 41.8
Leaves	289 \pm 16	147 \pm 33	7.3 \pm 2.1	275 \pm 14	2.8 \pm 0.4	18.0 \pm 1.5	83.8 \pm 12.3
<i>Artrocnemum macrostachyum</i>							
	Zn	Pb	Cd	Fe	As	Cu	Mn
Roots	4893 \pm 45	743 \pm 24	24.4 \pm 2.4	9499 \pm 44	97.4 \pm 8.3	70.2 \pm 1.3	597.4 \pm 1.6
Leaves	1649 \pm 13	724 \pm 9	3.4 \pm 0.7	18822 \pm 21	101.0 \pm 3.3	19.9 \pm 0.8	1893.4 \pm 1.4
<i>Zygophyllum fabago</i>							
	Zn	Pb	Cd	Fe	As	Cu	Mn
Roots	114 \pm 2.3	3.8 \pm 0.4	18.2 \pm 3.3	1600 \pm 29.6	15.1 \pm 2.3	24.7 \pm 4.6	113.8 \pm 1.3
Leaves	47 \pm 1.1	3.1 \pm 0.6	32.4 \pm 2.1	2000 \pm 50.7	11.8 \pm 1.3	35.8 \pm 0.7	47.4 \pm 2.2
<i>Dittrichia viscosa</i>							
	Zn	Pb	Cd	Fe	As	Cu	Mn
Roots	446 \pm 2.2	25 \pm 1.4	33.7 \pm 0.8	37 \pm 2.2	21.7 \pm 0.6	53.9 \pm 0.2	445.6 \pm 0.9
Leaves	187 \pm 1.2	187 \pm 1.1	3.9 \pm 0.6	1119 \pm 15.7	13.6 \pm 0.3	14.8 \pm 0.3	65.2 \pm 0.3

Table 4
Bioconcentration factor (BCF) and translocation factor (TF) average value in selected plant species (\pm standard deviation).

	<i>Limonium carthaginens</i>		<i>Glaucium flavum</i>		<i>Artrocnemum macrostachyum</i>		<i>Zygophyllum fabago</i>		<i>Dittrichia viscosa</i>	
	BCF	TF	BCF	TF	BCF	TF	BCF	TF	BCF	TF
Zn	0.05 \pm 0.02	0.71 \pm 0.11	0.48 \pm 0.02	0.53 \pm 0.02	1.01 \pm 0.24	0.22 \pm 0.08	0.23 \pm 0.16	0.62 \pm 0.09	0.33 \pm 0.19	0.45 \pm 0.11
Pb	0.05 \pm 0.03	0.78 \pm 0.02	0.35 \pm 0.09	0.50 \pm 0.13	0.47 \pm 0.16	0.88 \pm 0.17	0.05 \pm 0.02	0.50 \pm 0.12	0.48 \pm 0.22	0.65 \pm 0.13
Cd	0.10 \pm 0.05	0.71 \pm 0.09	0.65 \pm 0.21	0.90 \pm 0.25	2.34 \pm 0.36	0.03 \pm 0.02	0.18 \pm 0.06	0.85 \pm 0.24	0.38 \pm 0.11	0.47 \pm 0.09
Fe	0.01 \pm 0.02	0.80 \pm 0.13	0.28 \pm 0.09	1.14 \pm 0.02	0.03 \pm 0.02	1.44 \pm 0.25	0.01 \pm 0.03	0.63 \pm 0.15	0.43 \pm 0.18	0.20 \pm 0.12
As	0.01 \pm 0.02	0.56 \pm 0.15	0.28 \pm 0.02	0.06 \pm 0.02	0.08 \pm 0.02	1.07 \pm 0.22	0.01 \pm 0.02	0.40 \pm 0.14	0.15 \pm 0.08	0.50 \pm 0.21
Cu	0.12 \pm 0.04	0.62 \pm 0.02	0.60 \pm 0.11	0.77 \pm 0.05	0.70 \pm 0.18	0.26 \pm 0.09	0.11 \pm 0.04	0.72 \pm 0.25	0.17 \pm 0.07	0.77 \pm 0.17
Mn	0.01 \pm 0.02	0.58 \pm 0.13	0.42 \pm 0.02	0.46 \pm 0.02	0.05 \pm 0.02	4.76 \pm 0.39	0.03 \pm 0.01	0.55 \pm 0.13	0.15 \pm 0.08	0.35 \pm 0.09

by torrential rainfall, a usual phenomenon in the studied area, as dissolved and particulate forms. In this area a natural attenuation process is taking place when the parent material is capable of buffering drainage, being acid neutralization a primary control on the process of metal attenuation. In addition, revegetation processes are taking place in natural attenuated areas.

Obtained results suggested that *Z. fabago* could accumulate iron and *A. macrostachyum* iron, arsenic and manganese. The other plant species collected from the mining sites were tolerant, because their BCF and TF values were lower than unity. Although these plant species do not accumulate high concentrations of trace elements, they may be a good tool for reducing erosion effects and also for reducing the leaching of metals.

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