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Research article

Change in metals and arsenic distribution in soil and their bioavailability beside old tailing ponds

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ABSTRACT

The objectives of this study were to determine the metals and arsenic transfer from mining ponds to agricultural and forest soils, and identify the dynamic of metal(loid)s in the soil-plant system for a native plant species (*Ballota hirsuta)* in two old mining districts: La Unión and Mazarrón (Spain). Soils and plants from mining ponds and natural and agricultural areas were collected and analyzed for soil properties, and chemical partitioning of Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn and As. Results showed that mine, forest and agricultural soils were contaminated by As, Cd, Cu, Pb, and Zn. Chemical partitioning revealed higher mobility of metals in mining ponds than natural and agricultural soils except for Fe and As which were mostly bound to soil matrix due to the mineralogical compositions of soils. The accumulation of metal(loid)s in *B. hirsuta* in La Unión decreased as Fe > As > Cr > Ni > Cu > Zn > Cd > Mn > Co > Pb while in Mazarrón was As > Fe > Cr > Pb > Cu > Ni > Co > Mn > Zn > Cd, showing that *B. hirsuta* has high ability to bio-accumulate Fe, As, Cr, Cu and Ni; and Pb (in Mazarrón), transferring a significant concentration of theses metal(loid)s, except Pb, to edible parts without exceeding the toxicity limits for animals. Therefore, *B. hirsuta* could be useful as phytoextractor species for Cr, Cu, As and Ni, while it can be used as phytostabilizer species for Zn, Co, Pb and Cd.

1. Introduction

Mining activities have a negative impact on the environment by causing changes in landscapes, destruction of habitats, contamination of soil and water, and degradation of land resources (European Environmental Bureau, 2000). Even after mine closure, huge unrestored tailing ponds remain in the mining areas with the consequent environmental risks, such as metals aerial dispersion, leaching of metals and groundwater contamination, runoff of tailing, runoff of acid mine drainage etc.

In semiarid regions, lack of vegetation and scarce but torrential precipitation favour erosion of abandoned and unreclaimed mining ponds, promoting dust spread and acid mining drainage generation, which affects adjacent natural and agricultural lands (Li et al., 2006; Liao et al., 2016). Once the metal(loid)s reach these areas, native and crop plants can uptake metal(loid)s and accumulate them on their tissues (Chopra and Pathak, 2012; Gitet et al., 2016; Moreno-Jimenez et al., 2016) which causes a high environmental, ecological and human health risk. The accumulation of metal(loid)s by consumables plants can cause toxicity for wild fauna, cattle and human. In order to reduce the risk of toxicity for human, some countries have developed regulations which establish allowed concentrations of metals in foods (McGrath and Zhao, 2015). However, the metal exposure for wild fauna by ingestion mainly come from accumulated metals in edible parts of plants used as feed (Martínez- López et al., 2014; Gall et al., 2015). For humans the metals exposure occurs mainly through directly plants consumption, such as cereals and vegetables which can accumulate metals in grains and leaves (Li et al., 2006; Alvarenga et al., 2014). In addition, contamination of drinking water sources by mining operations is other important route of human exposure (Northey et al., 2016; Mhlongo et al., 2018; Rakotondrabe et al., 2018).

La Unión and Mazarrón municipalities (Murcia Region, southeast Spain) have been highly exposed to the negative effect of mining and ore operation during decades. Although the mining activity was abandoned twenty five years ago, more than eighty tailing ponds remain in the area, most of them unrestored. These mining ponds are composed by high amounts of Fe-oxyhydroxides, sulphates, and potentially leachable metal(loid)s due to extreme

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acidic conditions. For a long time, these mining residues have been transported downstream during periods of high rainfall and atmospherically dispersed, negatively affecting natural, agricultural and populated areas (Zornoza et al., 2012; Alcolea et al., 2015; Sánchez et al., 2017). However, the dynamic of metal(loid)s in soil-plant systems from natural lands has been little investigated. In fact, some common plant species growing in natural soils around mine areas has not yet been studied, such is the case of *Ballota hirsuta,* a native species which grows in natural soils from both mining districts. This plant protects the soil against wind and water erosion, being adapted to both semiarid climatic condition and high metal(loid)s content in soils. Consequently, we expect that *B. hirsuta* would be able to good species for using in phytoremediation techniques.

Therefore, the objectives of this study were: (1) to determine the metals and arsenic transfer from mining ponds to near agricultural and forest soils; (2) to evaluate the behaviour of metals and arsenic in the soil-plant system for a predominant endemic species (*B. hirsuta)*; and (3) to study the ability to accumulate and transfer metal(loid)s of *B. hirsuta* in order to determine its potential use as phytoremediation plant.

The results of this study can be used by the public administration and private companies to design low-cost restoration strategies, using native plants species, which will prevent the transfer of metal(loind)s from these areas to nearby agricultural areas with the consequent benefits for farmers.

2. Materials and methods

2.1. Study area and sampling collection

Sampling areas were located in two different mining districts: Cartagena-La Unión and Mazarrón (southeast of Spain) (Fig. 1). Wastes from both mining districts comes from the extractive activity of Pb, Zn and Fe sulphides which started in the times of the Romans and has continued until 1991 and 1960 in La Unión and Mazarrón, respectively. The climate is semiarid Mediterranean characterized by an annual average temperature of 18 °C and an annual precipitation of 290 mm (AEMET, 2016). The mining districts of Cartagena-La Unión and Mazarrón, and other mining districts around the world, are composed by a succession of areas which include a mine, tailing ponds, natural areas, and sometimes agricultural or urban areas. For this study, one of these areas in each mining district was selected, including a mining pond, a natural area, and an agricultural area (Fig. 1). La Unión pond (30S X684915, Y4165525) has an extension approximately of 15500 m^2 and Mazarrón pond (30S X647316, Y4162776) of 6100 m^2. The sampling in the surface of each tailing pond was carried out following a

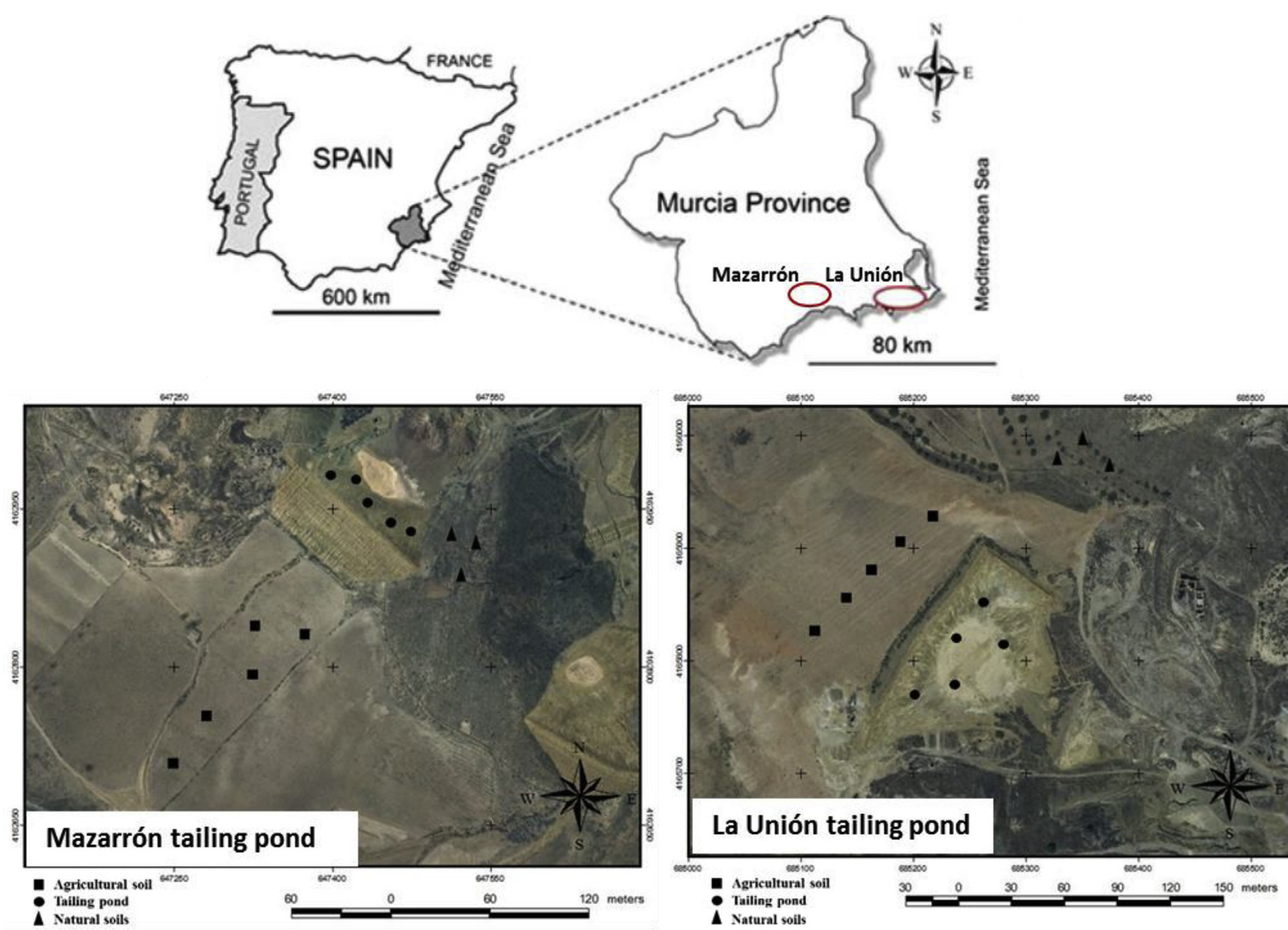

Fig. 1. Sampling points in La Unión (right) and Mazarrón (left).