



## Mobility indices for the assessment of metal contamination in soils affected by old mining activities



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### ABSTRACT

This work presents a methodology to assess the environmental potential risk of soils when submitted to different conditions. The potentially toxic element (PTE) behaviour was evaluated by single chemical extractions in order to simulate four conditions: PTEs leaching under actual rainfall conditions; acid mine drainage; and an anoxic and an oxic environment. Soil pollution assessment was carried out using the contamination factor (CF) and the pollution load index (PLI) for total contents and indicators of mobility were established for each extraction: natural mobility indicator (NMI), acid mine drainage mobility indicator (AMI), oxic mobility indicator (OMI) and anoxic mobility indicator (ANMI). The total PTE content in the samples was high, and this content decreased when the distance of the sampling site to the source of contamination increased. The results obtained after the extractions suggested that the highest PTE content was extracted in the acidic medium. The mineralogical composition is an important factor that should be taken into account in the evaluation of PTE mobility, firstly because the mineral phases react differently in the proposed situations depending on their chemical nature, and secondly, because the presence of a particular phase (with different degree of reactivity) depends on the degree of weathering. The most reactive materials under oxic conditions are, in the study area, the most modern. The proposed indices may constitute a first attempt to define areas with high potential environmental risk, where urgent action is needed. In addition, the proposed methodology could be a valuable tool to prioritise these interventions.

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### 1. Introduction

Mining and smelting activities are important sources of potentially toxic elements (PTEs) in the environment, resulting in considerable soil contamination (Adriano, 2001; Alloway, 1995). Modern day mineral extraction and treatment processes are very efficient and resulting wastes contain lower concentrations of PTEs than their historical counterparts. However, inefficient mineral processing operations meant that wastes from historical mining operations contained higher PTE concentrations (Lottermoser, 2007). In these cases, tailings were heterogeneous and strongly affected by weathering, showing high PTE content and reactivity. These areas present erosion problems caused by wind and water run-off, where soil and mine spoil texture, landscape topography and regional and local meteorological conditions play an important role (Navarro et al., 2008).

The characterisation of soil PTE contamination is commonly based on the determination of total contents. However, the total concentration

is not sufficient, as it only provides limited information on the chemical behaviour and the mobility of PTEs.

PTEs are present in the soil in different fractions (water-soluble, exchangeable, carbonate-associated, Fe–Mn oxide-associated, organic-associated and residual forms), which may strongly affect the way the element behaves in the soil, depending on its bioavailability, toxicity, chemical interactions and its mobility within the soil (Arenas-Lago et al., 2014). Then, the assessment of the potential effect of soils polluted by PTEs should be based on the identification of mineral phases in which the metals are bound and the evaluation of the metal retention and mobility (Adamo et al., 2003).

A variety of selective reagents have been proposed for studying mobility, but none have been fully recognised as a universal procedure applicable to all types of soils or sediments (Favas et al., 2011; Martínez-Sánchez et al., 2008). Sequential chemical extractions use a series of selective reagents chosen to successively dissolve the mineralogical fractions thought to be responsible for retaining the larger part of the PTEs (Rodríguez et al., 2009). However, these approaches had fundamental limitations, such as the lack of specificity in element removal, readsorption and subsequent redistribution amongst phases, lack of comparability between studies as experimental parameters vary widely and the potential for contamination increases with number

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of steps and the length of time required to process successive steps (Favas et al., 2011; Sutherland, 2010).

On the other hand, single-step extraction methods enable a rapid evaluation of the PTEs associated with a specific fraction in soils and sediments. These methods may give useful information about PTE behaviour, when different environmental conditions were simulated by the selected single extractant or a mixture of them (Alvárez et al., 2011).

The aims of this study were to investigate the mobility and the geochemical behaviour of potentially toxic elements (Pb, As and Cd) under different environmental situations in soils affected by mining activities by applying four selective extraction procedures. Additionally, the application of mobility indicators allows to explore the degree of PTE contamination.

## 2. Material and methods

### 2.1. Study site

Sierra Minera is a belt of low rising hills (maximum height, 431 m.a.s.l) in southeast of the province of Murcia (Spain), in the Cartagena – La Unión area (Fig. 1).

The average annual temperature in the area is 17 °C, and total annual precipitations do not exceed 300 mm, with occasional torrential rainfall, which frequently occurs between the end of summer and autumn. Perennial streams do not exist, and surface water consists of ephemeral streams (gullies), which only flow during rainfall events.

This coastal mountain range, with an approximate area of 50 km<sup>2</sup>, is bound by the Mediterranean Sea to the south, and by a large closed basin (known as Campo de Cartagena) to the north. In this endorheic basin, an area with high influence of Sierra Minera could be delimited, with a high degree of local and diffuse contamination, limited by the Miranda and Atamaría gullies and the Mar Menor (García-Lorenzo, 2009; Mantilla, 2010). The predominant soils in this area are luvic Calcisols and relic Luvisols (red soils) followed by calcic and petrocalcic Xerosols. The abundance of limestone causes in most cases, carbonated

soils. In addition, Campo de Cartagena shows vestiges of postorogenic volcanic activity, including numerous small hills mainly composed of andesites. The soils developed over these materials mainly are Leptosols.

Sierra Minera consists of Paleozoic (mostly micaschists) and overthrust Triassic materials (metamorphic and sedimentary carbonates), emplaced during the Alpine orogeny as part of the Betic chain of southern and southeastern Spain. Miocene volcanism triggered hydrothermal activity, which resulted in the formation of one of the most important polymetallic mining districts of the SE Iberian Peninsula, already known to ancient Mediterranean cultures some 3000 years ago. The mining district of La Unión used to be one of the most important mining sites in Spain and is a perfect example of the close relationship amongst the Miocene magmatism, tectonics, and metallogenic processes in SE Spain. Mining activities originally (from the VII century B.C.) focused on Ag and Pb.

The ore deposits of this zone have iron, lead and zinc, as the main metal components. Iron is present in oxides, hydroxides, sulphides, sulphates, carbonates, and silicates; lead and zinc occur in galena, sphalerite, carbonates, sulphates, and lead or zinc-bearing (manganese, iron) oxides (Oen et al., 1975).

Following many centuries of intermittent activity, mining resumed in the XIX century, with a renewed interest in Zn and Fe. From 1940 to 1957, mineral separation was made by froth flotation and prior to this, by gravimetric techniques. From the mid-XXth century, large open pits predominated, until 1990, when mining ceased altogether. From 1957 to 1991 the ores mined were transported to “Lavadero Roberto”, the beneficiation plant located in Portman Bay, which used seawater for flotation processes. Processed tailings were dumped directly to the bay, which was flooded by sediments, resulting in the coastline retracting 100 m (Martínez-Sánchez et al., 2008). The composition of the mineral waste is related to the original paragenesis that was mined: paragenesis 1 (chlorite, sulphides, siderite, and silicates) and paragenesis 2 (greenalite, magnetite, sulphides, siderite, and silicates) (Peña et al., 2013).

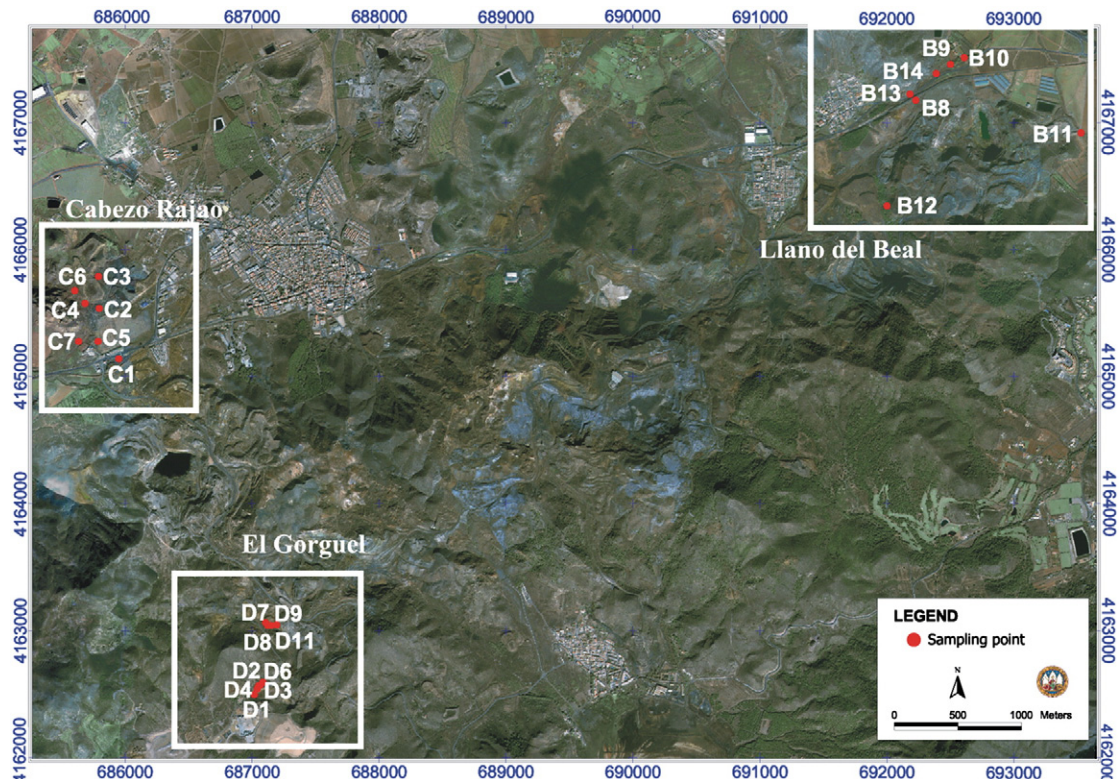


Fig. 1. Selected zones in the study area.

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