



A multidisciplinary characterization of a tailings pond in the Linares-La Carolina mining district, Spain



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ABSTRACT

Geochemical and geophysical techniques have been applied to investigate the potential environmental impact of the abandoned La Aquisgrana mine tailings, one of the most important sulfide-bearing tailings ponds of the Linares-La Carolina mining district (Spain). The geometry of the pile has been defined through geological field work and electrical resistivity imaging (ERI) surveys. The ERI profiles revealed the position of the bedrock surface below the tailings central area (more than 40 m in thickness) and helped in placing drill core sampling for geochemical analysis and a piezometer installation. In the 21 extracted samples, the total content of Pb, Zn, Cu, Fe, Mn, Sr, Rb and As were determined, with significant values being found for Pb and Zn ($>2000 \text{ mg kg}^{-1}$), Mn ($>700 \text{ mg kg}^{-1}$) and As ($>170 \text{ mg kg}^{-1}$). Although AMD has not been identified, water samples from the saturated zone of the tailings are characterized by elevated dissolved sulfate (2495 mg L^{-1}), Fe (20 mg L^{-1}), Mn (16 mg L^{-1}) and Zn (7 mg L^{-1}) contents, with an E.C. value of 3.3 mS cm^{-1} and pH from 5.6 to 6.9, suggesting a process of metal mobilization. Physical and chemical data obtained from drill core and groundwater samples were combined with the electrical resistivity profile of the tailings to characterize areas with higher metal concentrations. A central low resistivity area ($<30 \Omega \cdot \text{m}$) was identified in the vadose zone of the pile, correlated with higher Fe, Zn, Pb, As, and sulfide concentrations, and with a significant increase in silt and clay content. The lowest resistivity values ($<5 \Omega \cdot \text{m}$) were measured in the saturated zone of the tailings, related with the highest metal(oid) contents in both solid phase and water. A lower resistivity area close to the fractured zone of the bedrock suggests a preferential flow path for subsurface water infiltration. The generated geophysical–geochemical model has allowed to depict areas of the tailings characterized by high metallic and water contents that present the greater risk for contamination.

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1. Introduction

The Linares-La Carolina metallic mining district (southern Spain, Jaén province, Fig. 1) is characterized by vein deposits that are primarily composed of lead and copper sulfides (Castelló and Orviz 1976). These mineralizations have been the subject of intense historical exploitation, but were abandoned during the late 20th century. The mineralogical industry was developed in parallel, with the creation of numerous gravimetric and flotation plants. Until the mid-1950s, only gravimetric techniques were used to determine mineral concentrations. The waste products were typically deposited in heaps located near the mining facilities, sometimes

possessing high mineral grades due to the technical limitations of these processes (Gutiérrez-Guzmán 1999; Contreras and Dueñas 2010).

During the second half of the 20th century, a flotation process was introduced to separate and concentrate metal-bearing mineralin the Linares-La Carolina mining district. The flotation technique resulted in significant advances in the mineral processing industry, allowing nearly 100% of all metal content to be recovered via mining, a percentage that was virtually unthinkable using traditional gravimetric methods. In fact, ancient mine wastes with higher metal grade contents were reworked using flotation to obtain their metallic contents. The final waste products were deposited in tailings ponds, often with no prior land preparation. This created considerable environmental risks. At least 32 large tailings impoundments have been identified in this mining district (Gutiérrez-Guzmán 1999; Martínez 2002; Contreras and Dueñas 2010). Significant attention is

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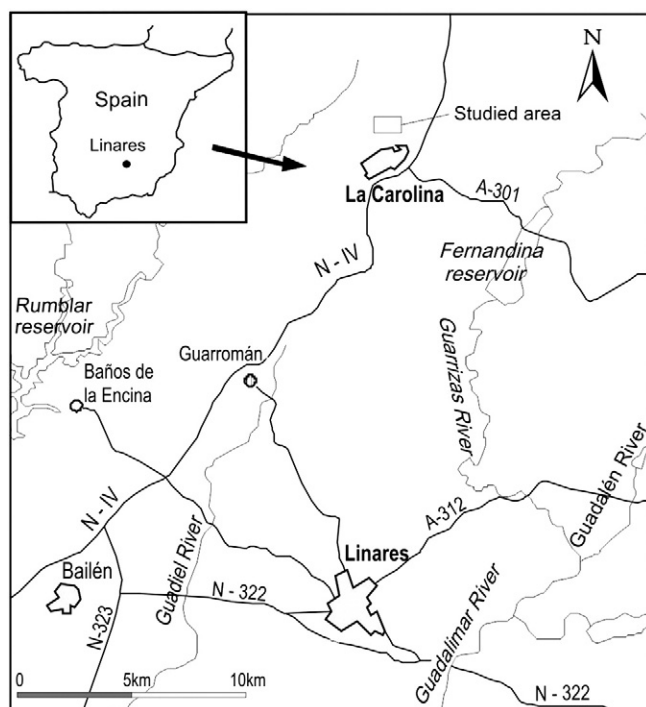


Fig. 1. Location of the study region.

currently given to the management of these potentially hazardous materials.

Thus, the main objectives of this study are to:

- Identify the deposited residue levels that may present a contamination risk through the combined use of geophysical and geochemical techniques.
- Characterize the tailings and provide data for planning of remediation actions.

The La Aquisgrana mine tailings, one of the most important tailings impoundments in the district, were selected, based on its size (with a total volume of around one million m³) and its location. It is situated in an agricultural area with olive groves and close to the La Campana River floodplain, suggesting potential soil and water environmental risks. These studies have a special interest for the control of tailings from other mining regions, where the exploitation, mineral recovery and metallurgy processes represent major heavy metal sources (Davis 1983; Li and Thornton 1993, 2001; Chopin and Alloway 2007; Chae Jung 2008; Bundschuh et al. 2012; Gómez-Ros et al. 2013; Domínguez et al. 2016).

2. Description of the study area

The La Aquisgrana mine tailings pond is one of the largest (200 × 180 × 35 m) and most representative of the La Carolina mining district. It is situated in the La Carolina municipality, 2 km to the north of the town, close to the remains of the La Aquisgrana abandoned washery (Fig. 2, Fig. 3A).

The La Aquisgrana mine was operated from 1899 to 1983. The exploited ore consists of hydrothermal Pb–Ag veins and Cu–Fe sulfides, which is hosted by metamorphic rocks, such as phyllites with sporadic quartzite layers (Lillo 1992). Specifically, the mineral paragenesis consists of:

- Pb–Ag sulfantimonites, with galena being the main ore, along with cerussite and anglesite. Silver sulfantimonites occur as minor ore minerals, and ankerite is the predominant gangue.

- Cu–Fe sulfides, of which chalcopyrite, pyrite and marcasite are the main ores, sphalerite and galena are the minor ores, and quartz, calcite and chlorite are the main gangue minerals.

In 1973, a flotation plant was constructed to rewash the former gravimetric heaps located in the vicinity of the mineral exploitation (Gutiérrez-Guzmán 2007). The flotation flow-sheet consisted of finely grinding the graded mixtures, with waste sizes of <16 mm. The pulp was taken to a closed-cycle Denver-type classifier to separate particles with Tyler mesh sizes of less than 48 (0.297 mm). Larger sizes were returned to the mill. Upon reaching the optimal size, pulp that contained 42% solids was transferred to the flotation process. Denver dual-cell, 80 × 80 cm sub-ventilation equipment was used to perform roughing, cleaning and scavenging processes. The most commonly used collectors were sodium and potassium oxalate. Long-chain aliphatic alcohols were typically used as foaming agents along with pine oil. Sodium cyanide was used as a depressor, which forms zinc cyanide with ZnSO₄. This depresses iron sulfide, arsenopyrite and sphalerite, and also has the beneficial effect of serving as a desilter. CaCO₃ and Ca(OH)₂ were used as regulating agents to attain a basic bath with a pH between 8 and 10, which is ideal for galena flotation. Finally, a fine concentration was obtained with lead concentrations approaching 75% (Contreras and Dueñas 2010).

The tailings were dumped on Palaeozoic phyllite substrate. The tailings impoundment was built from a small dike of spoiled materials. It is situated in a valley, with the tailings occupying the riverbeds of two tributary creeks of the La Campana River, which flows by the foot of the impoundment. The runoff from the creeks is channeled by a drainage pipe at the bottom of the tailings, which discharges into the La Campana River. The top of the tailings is positioned on a slight south-westward facing slope, which collects water during the rainy season. In addition, the slope features substantial surface erosion, cracks and local landslides. In addition, bank erosion occurs on the lower sections of the slope during large flood events, undermining the overall structure.

De la Torre et al. (2010) studied the mineralogy of tailings from the La Carolina district. Two samples were taken from the first 30–50 cm of La Aquisgrana tailings and studied using X-ray diffraction (XRD) and a scanning electron microscope (SEM). The results indicate that the tailings are primarily comprised of quartz and phyllosilicates, which are very abundant. Feldspars, calcite, ankerite, cerussite and galena were also identified as trace minerals. Precipitated salts from the surface of the tailings were also analyzed. Gypsum was the predominant mineral phase, but Fe sulfate with As and Zn was also present.

3. Methodology

3.1. Geophysical surveys

A geophysical survey was conducted using Electrical Resistivity Imaging (ERI). This technique has been previously used for tailings analyzes (Martínez-Pagán et al. 2009, 2011; Gómez-Ortiz et al. 2010; Martín-Crespo et al. 2010; Placencia-Gómez et al. 2010; Martínez et al. 2012, 2014; Martín-Crespo et al. 2012, 2015; Zarroca et al. 2015).

This is a non-destructive geoelectrical method that analyzes underground materials based on their electrical behavior, differentiating based on electrical resistivity (Telford et al. 1990; Store et al. 2000). The method consists of implanting electrodes over a profile, with a pre-determined separation based on the degree of sensitivity and depth to be analyzed. A higher sensitivity is obtained when the electrodes are placed closer, but wider spaced electrodes allow for scanning at greater depths (Loke and Barker 1996; Loke and Dahlin 2002; Dahlin and Zhou 2004). Technically, an electrical-resistivity imaging survey can be conducted using different electrode arrays (e.g., dipole–dipole, pole–pole, pole–dipole, Wenner, Schlumberger or Wenner–Schlumberger)

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