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#### Research papers

## Hydrological characterization and prediction of flood levels of acidic pit lakes in the Tharsis mines, Iberian Pyrite Belt



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#### ABSTRACT

Opencast mining operations frequently lead to the creation of large voids that become anthropogenic lakes when the water table recovers. In the case of sulfide mining the stored water is of an acidic nature with significant concentrations of toxic metals and, therefore, a high pollutant potential. The main goal of the present work is to characterize the hydrological functioning and evolution of four acidic mine pit lakes in the abandoned mines of Tharsis, which is the second most important mine district in the Iberian Pyrite Belt (IPB). We present a simple methodology based on the use of the available orthophotographs and a Digital Terrain Model (DTM) together with the water balance of the pit lakes, which could be applied to other abandoned mining sites, where there is often a lack of hydrogeological information that prevents the application of more complex models. The accumulation of large volumes  $(5.2 \times 10^6 \, \text{m}^3)$  of acidic and metal-rich waters in these pit lakes poses a serious environmental concern, with dissolved concentrations up to 2000 mg/L of Fe, 223 mg/L of Al, etc. Sierra Bullones and Filón Norte are connected underground and present the same evolution, with water transfers from Sierra Bullones to Filón Norte. The water level in both pit lakes is increasing, with an average rise of 2.8 m/yr since the beginning of flooding. However, the increase in the evaporation rate, as a result of the larger flooded area as the water level rise, would induce a hydrological equilibrium before reaching the overflow level, leading to the formation of a terminal lake. On the other hand, the water level in Filón Centro and Filón Sur pit lakes remain approximately stable. The first behaves as a flow-through or terminal lake, depending on the annual rainfall, while the second acts permanently as a flow-through lake.

#### 1. Introduction

Mine closure constitutes one of the biggest environmental problems worldwide (Younger et al., 2002; Sarmiento et al., 2009; Robles-Arenas and Candela, 2010). In surface mining, large open pits are created. The cease of pumping once mining activities end generally leads to the progressive flooding of the open pits, generating pit lakes. The hydrological equilibrium of the pit lake is achieved when water inputs are equal to losses by evaporation or when overflows are produced on the surface and in subterranean areas, generating a discharge of polluted water to rivers or aquifers. The release of these waters can pose substantial environmental concerns (Davis and Ashenberg, 1989; Savage et al., 2000; Castendyk, 2011; Castendyk et al., 2015a,b; Boehrer et al., 2016). In the case of sulfide mining, the problem is worse because pit lakes usually store acidic waters with extreme concentrations of toxic

metals (Sánchez España et al., 2008; Cánovas et al., 2015) that are of a high pollutant potential (Nordstrom et al., 2015).

The Iberian Pyrite Belt (IPB) is one of the biggest polymetallic sulfide deposits in the world. The intense mining activity mainly developed during the second half of the nineteenth and early twentieth centuries has left an impressive environmental legacy, with large amounts of widespread mine wastes, which have an important impact on the Tinto and Odiel Rivers due to acid mine drainage (AMD) processes (see, for example, Sánchez España et al., 2005; Cánovas et al., 2016; Olías et al., 2016). As a consequence of the ceased mining at the end of the twentieth century, there are 22 flooded open pits in the Spanish part of the IPB (Sánchez España et al., 2008). Most of these pit lakes contain acidic water and lack management or control plans. Although the hydrogeochemical and limnological properties of some of these pit lakes have been previously studied (Sánchez España et al.,

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2008; Santofimia and López-Pamo, 2013; Cánovas et al., 2015), some of them have not achieved equilibrium, and their hydrological connections are not well understood (Sánchez España et al., 2014). This information is of paramount importance to foresee their evolution, to plan potential remediation measures, and to avoid potential environmental risks.

This issue is especially relevant for the Tharsis mine complex, which constitutes one of the most important exploitations of the IPB. This derelict district hosts five open pits, four of which are partially flooded by acidic waters (Sánchez España et al., 2008). In addition, there is a surface of around 3.6 km<sup>2</sup> covered by mine wastes that generate acidic leachates. As a consequence, main water bodies in the drainage basin are deeply polluted (Cánovas et al., 2017). Some of the acidic leachates generated in the Tharsis mine complex join the Meca River, which feeds the Sancho Reservoir (58  $\times$  10<sup>6</sup> m<sup>3</sup>). The Sancho Reservoir has suffered a progressive acidification in recent years, and it is considered to be one of the more extreme cases of surface water pollution worldwide (Cánovas et al., 2016). A large reservoir (246  $\times$  10<sup>6</sup> m<sup>3</sup>) is currently under construction to receive the acidic leachates from the Tharsis mine complex (Olías et al., 2011). If additional leachates were generated from existing pit lakes, the water quality of local streams and reservoirs would worsen.

The water quality of the pit lake depends on the balance between the acidity and alkalinity inputs into the lake together with the neutralization processes in the water column (Blodau, 2006; Castendyk et al., 2015b). There are currently several models that use information from these processes to predict the final water quality after mining cessation (Oldham et al., 2009; Vandenberg et al., 2011; Geller et al., 2013; Castendyk et al., 2015b). However, to implement these models a detailed knowledge of the hydrogeological characteristics of pit lakes and their surroundings is needed. While active mines can be well characterized, there is a lack of information on abandoned mine sites, as Tharsis, that prevents the application of such models.

Based on the concerns described above, the main goals of the present work are: 1) to analyze the flooding evolution of the pit lakes of the Tharsis mine complex from the available orthophotographs and a digital terrain model (DTM); 2) to preliminarily assess the hydrogeochemical characteristics of the pit lakes and the amount of stored pollutants; 3) to obtain a conceptual model of the hydrological behavior of the pit lakes and perform a water balance of each system and; 4) to predict the evolution of these pit lakes in the long term. This information is critical to identifying and adopting cost-effective measures to mitigate the impact of acidic waters in local receiving water bodies.

#### 2. Site description

The Tharsis mining complex has a Mediterranean climate with an average yearly precipitation close to  $600 \, \text{mm}$ , but it exhibits high intra- and inter-annual variability (Galván et al., 2009). Average temperatures in the area are close to  $16.5\,^{\circ}\text{C}$ . Summers are hot and dry, with maximum temperatures close to  $40\,^{\circ}\text{C}$ , while winters are humid and cold (minimum temperatures lower than  $0\,^{\circ}\text{C}$ ). The most important water courses are the Aguas Agrias Creek, which joins the Oraque River (feeding the Alcolea projected reservoir) to the east, and the Meca River, which is located in the south and is regulated by the Sancho Reservoir (Fig. 1).

The IPB is a part of the South Portuguese Zone of the Hercynian Iberian Massif, which extends from the western part of Spain to the Portuguese Atlantic coast (approximately 200 km long and 40 km wide) and consists of three lithologic units: the Phyllite-Quartzite Group (PQ), the Volcano-Sedimentary Complex (VSC) and the Culm Group (CG). The PQ group consists of a thick sequence of slates and sandstones of upper Devonian age. The materials of the VSC (upper Devonian – lower Carboniferous age) are composed of a volcanic sequence, with a variable thickness up to 1300 m (Tornos, 2006) and alternating episodes of felsic rocks (dacites and, in lower proportion, rhyolites) and mafic rocks

(occurring as basaltic sills or small stocks) intercalated in a sedimentary sequence (mainly phyllites). This sequence presents abrupt changes due to the intrusive character of some igneous rocks, the abundance of thrust defining the major contacts and the existence of several palaeogeographic domains (Tornos, 2006). The VSC hosts numerous sulphides deposits. Finally, the CG is a detrital unit of synorogenic turbidites of up to 3000 m thick, which basically consists of shales and conglomerates of Carboniferous age. The materials of the IPB has been considered of low permeability and do not constitute aquifer units of importance. Nevertheless, the thrust limits and fractured zones must be locally more permeable.

In the Tharsis area, the structure is defined by four major southdipping tectonic units limited by thrusts with different lithological and hydrothermal features (Tornos et al., 2009): 1) Slates and sandstones of the PQ Group, interpreted as a para-autochthonous group overlied by the other units, 2) The Lower Unit including the massive sulphides and slates, 3) The intermediate Unit, made by slates, intruded by basalts with bodies of hydrothermal breccias and 4) The Upper Unit, composed of rhyodacite sills intruding slates. There is no available data about the hydrogeological characteristics (hydraulic conductivity, storage coefficient, etc.) of these materials. Paleozoic rocks from the IPB are characterized by a low permeability. The hydraulic conductivities are around  $10^{-9}$  m/s (Gómez de las Heras et al., 2001). Nevertheless, these values can increase due to the existence of faults, diaclases, etc. Thus, at the Aznalcóllar mine zone values between  $9 \times 10^{-9}$  and  $10^{-5}$  m/s are found (Gómez de las Heras et al., 2001). Also, mining labors in the vicinity of pits promote an increase the permeability of such materials by fracturing (Cánovas et al., 2018).

Original sulfide reserves in the Tharsis mine complex are estimated to be around 133 Mt (Tornos et al., 2009). Mining in the IPB started around 5,000 years ago during the Chalcolithic (Nocete et al., 2005). After a period of inactivity, mining restarted during the Tartessian and Roman periods (Gonzalo y Tarín, 1888). Afterwards, a long period of low mining activity continued until 1856 when the mine was rediscovered (Deligny 1863) and underground mining was performed, mainly by room and pillars in Filón Norte, Sierra Bullones, Filón Centro, and Filón Sur (Fig. 1). Opencast mining started in 1866 in Filón Norte, and some years later in Sierra Bullones and Filón Centro (Gonzalo y Tarín, 1888). Mining in Filón Centro and Filón Norte ceased in 1884 and 1890, respectively, and focused on mineral exploitation in Sierra Bullones by both opencast and underground mining.

Since the beginning of the twentieth century, sulfur from pyrite was obtained to produce sulfuric acid. However, a new processing method was developed in Filón Sur from 1937 to 1964 to extract Au and Ag from the gossan (Checkland, 1967). In the mid-twentieth century, mining was restarted in Filón Centro, leading to an increase in the open pit. Around 1960, a new period of exploitation in the Filón Norte open pit also began. Mining ceased in Sierra Bullones in 1966 (after 100 years of intense exploitation), while Filón Norte mining finished at the end of the 90s. The last mining activities developed in the Tharsis mine complex were performed in Filón Sur from 1990 to 2001 to obtain Au and Ag by cyanide leaching of the gossan.

From 1856 to 2001, approximately 40 Mt of sulfide were obtained (Tornos et al., 2009). Approximately 17 Mt were obtained up to 1960, mainly from Sierra Bullones (Pinedo Vara, 1963), while the rest were obtained mainly from Filón Norte during the last 40 years of exploitation. About 5 Mt of mineral must be added to these figures from mining developed during the Roman and pre-Roman periods (Gonzalo y Tarín, 1888).

#### 3. Methodology

#### 3.1. Water level and total volume stored in the pit lakes

Due to the absence of water-level records, in order to estimate the flooding evolution of the pit lakes, digital terrain models (DTM) for

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