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Hydrogeochemical behavior of an anthropogenic mine aquifer: Implications for potential remediation measures

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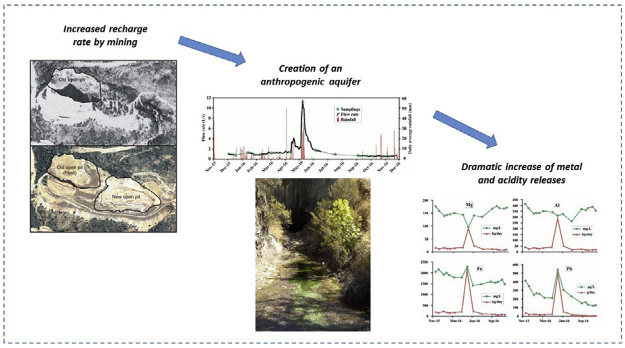
HIGHLIGHTS

- An anthropogenic mine aquifer with a similar behavior than karstic systems
- Pollutant concentration shows a low variability except during rainy episodes.
- A water balance identified the open pit surface as the aquifer recharge area.
- Dramatically increased recharge rate of the anthropogenic aquifer by recent mining
- Potential measures are proposed to mitigate the pollution of this mine aquifer.

GRAPHICAL ABSTRACT



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ABSTRACT

This study characterizes the hydrogeochemical behavior of one of the most pollutant sources in the Iberian Pyrite Belt, namely, the Poderosa adit outflow. This artificial spring arises from an anthropogenic mine aquifer with a similar hydrogeological behavior to karstic systems, where the infiltration area is an endorheic zone and the aquifer shows allogenic recharge. Recent mining has markedly increased the contaminant levels. The pollutant load released from the adit to the receiving water body is very high, with average loads of 280 kg/day of Fe, 47 kg/day of Al, 17 kg/day of Cu and so on. However, a high variability is observed related to hydrological and geochemical factors, especially during intense rainy episodes. Thus, the pollutant load during these events suffers a dramatic increase, i.e., from ~100–200 kg/day of Fe during base flow to almost 2200 kg/day during the flow peak. These data highlight the importance of short but intense rainy events on metal fluxes from mining areas, which has been previously reported in surface waters but scarcely reported in mine adits, with expected lower response times to rainfall. The pollutant load released by non-point sources, i.e., spoil heaps, is lower than that released from the adit most of the year, although it increased noticeably during intense rainy events. Some remediation measures were adopted during the 1990s without a suitable hydrogeological characterization and were shown to be ineffective. On the basis of the obtained results, potential restoration measures are discussed.

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1. Introduction

The formation of anthropogenic aquifers in mining zones is common after mine closures (e.g., Younger et al., 2002; Jacobs et al., 2016). During mining labor, a large system of galleries and pits is created and fracturing causes a sharp increase in rock porosity. In addition, enormous

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coarse grain spoil heaps are usually dumped in the vicinity of the mines. All these geologic materials can behave as aquifers, generating previously inexistent outflows that can be of an acidic nature in the case of sulfide mining due to the process commonly known as acid mine drainage (AMD) (Wolkersdorfer, 2008; Nordstrom et al., 2015). These anthropogenic springs often have permanent discharges and present extreme levels of contaminants (Jacobs et al., 2016), which may cause serious damage to the receiving water courses (e.g., Alpers et al., 1992; Banks et al., 1997; Cidu et al., 2011; Cravotta et al., 2014; Frau et al., 2015; Cánovas et al., 2016a).

This environmental problem strongly affects the Odiel River Basin (SW Spain), which drains the materials of the Iberian Pyrite Belt (IPB), an area well known for hosting many polymetallic massive sulfide deposits, which have been exploited intensely since the second part of the 19th century. The low permeability of the IPB materials provokes the inexistence of natural aquifers in the watershed. The groundwater flow is only limited to shallower layers (a thickness of a few meters) and is related to secondary porosity by fracturing (Galván, 2011). The only natural springs in the area correspond to ephemeral outflows of minor importance (<0.1 L/s). However, mining anthropogenic aquifers usually generates permanent AMD discharges that may play a key role in the pollution pathways at the IPB (Caraballo et al., 2016; Cánovas et al., 2016b).

The European Union Water Framework Directive demanded that member states maintain a high quality status of surface and groundwater and prevent any quality deterioration by 2015 (Heinz et al., 2007). The longevity of AMD processes (Younger, 1997) has forced this to be delayed to 2027 for the Odiel River Basin because improvements within the timescale would be disproportionately expensive. Despite the poor quality of surface waters, a large reservoir (Alcolea, 247 hm^3) is currently under construction in this basin, although reasonable doubts about the final quality of the reservoir water have been reported (Olias et al., 2011). Urged by the effective recovery of water quality in the basin in the short term, regional authorities have recommend in the River Basin Management Plan, among other measures, the use of passive technologies to reduce the mining pollution. However, the implementation of restoration measures must be based on a thorough understanding of these hydrogeochemical systems (Macías et al., 2017).

Alternatively, diffuse water pollution is considered as a major threat for water quality and is the largest remaining problem of water pollution in many countries (Orr et al., 2007). Compared to point pollution, diffuse pollution is more complex and difficult to control due to its numerous and dispersed sources, and the difficulties in tracing its pathways (Yang and Wang, 2010). This is especially significant in derelict mines where wastes are usually widespread in the surroundings of the mine sites. The occurrence of such diffuse sources in AMD affected systems has been reported in detail (e.g., Mayes et al., 2008; Kimball and Runkel, 2009; Gozzard et al., 2011), however the contribution of point sources, such as mine adit outflows, to the total pollutant load may be preponderant (Walton-Day and Mills, 2015; Beanes et al., 2016). Nevertheless, the contribution of diffuse and point sources to water pollution must also be quantified in order to adopt cost-effective restoration measures.

On this basis, the main goals of this work are: 1) the hydrogeological characterization of an anthropogenic mine aquifer in the IPB, 2) to estimate the contribution of point and diffuse sources to the pollutant load delivered by this mine system and 3) to assess the most sustainable remedial actions. The information provided by this study could be of significant use in the restoration of other derelict anthropogenic mine aquifers worldwide, especially those originating from sulfide mining.

2. Study area

2.1. Site description

The Odiel River Basin (SW Spain) is one of the most AMD impacted regions in the world. The absence of carbonate materials in the IPB, which could neutralize the acidity released, together with the large scale mining developed in the area, have led to extreme pollutant levels in the watershed, causing the deterioration of most of the Odiel watercourses and transporting a huge amount of contaminants to the Ría of Huelva estuary (e.g., Sánchez España et al., 2005; Olías et al., 2006; Cánovas et al., 2007).

A significant pollutant source of the Odiel River is the Poderosa mine, located in the northern part of the IPB. This can be considered a small mine (23 ha of affected surface; Grande et al., 2014) with respect to others located in the IPB. However, there is a derelict mine gallery (known as the main adit, Fig. 1) that constitutes one of the most pollutant outflows of the IPB. The permanent release of pollutants throughout the year has a dramatic impact on the Odiel River waters (Sánchez España et al., 2005, 2006; Sarmiento et al., 2009).

The climate in this region is of the Mediterranean type, with rainy winters and dry, warm summers. The average temperature is around 16 °C, with minimum values in winter (~0 °C) and maximum values in summer (up to 40 °C). The average pluviometry in the catchment is close to 750 mm, although a high inter- and intra-annual variability is observed. Most rainfall (~60% of the total) is collected between October and January, while rainfalls between June and August are almost negligible (Galván, 2011).

The materials outcropping in the study area belong to the IPB, which extends from the western side of Seville (Spain) to the Portuguese Atlantic coast (~200 km long and 40 km wide). The IPB is formed by three lithologic units, namely, the Phyllite-Quartzite Group (PQ), the Volcano-Sedimentary Complex (VSC) and the Culm Group. The PQ group consists of a thick sequence of shales and quartzites of the upper Devonian age. The materials of the VSC (upper Devonian-lower Carboniferous age) are composed of a volcanic sequence, with alternating episodes of felsic (dacites and, to a lesser extent, rhyolites) and mafic rocks intercalated in a sedimentary sequence mainly composed of shales. The Culm group is a detrital unit of synorogenic turbidites, basically composed of shales and conglomerates of the Carboniferous age. All these materials were folded and subjected to a low-grade metamorphism during the Variscan orogeny.

Two different massive sulfide deposits are found in the Poderosa mine (Fig. 1), which are enclosed in acid epiclastic rocks (Pinedo Vara, 1963): a deposit of 175 m long and an average thickness of 7 m (North Lode), and a smaller one of ~150 m in length and 2 m thick (South Lode). Over the sulfide deposits, a gossan layer with a thickness ranging from 20 to 60 m is found. Sulfides found in Poderosa mine are mainly composed of pyrite and, to a lesser extent, chalcopyrite, chalcocite and covellite.

2.2. Mining history in the area

Mining activity in Poderosa dates back, at least, to the Roman times. There are still remnants of this activity, such as derelict galleries (Fig. 1). Modern mining, developed at a higher scale, began around 1864 (Gonzalo y Tarín, 1888; Pinedo Vara, 1963) initially using the already existing Roman galleries in South Lode and a new gallery built at level 325 m for mineral transport and mine dewatering. Copper grades of up to 7% were found during exploitation, one of the higher values in the IPB (Gonzalo y Tarín, 1888). The mineral was processed by open-air calcination, deposited in heaps of ~50 tones and roasted until the sulfur was almost depleted. After calcination, the roasted mineral was leached with AMD to enhance Cu dissolution, which was subsequently recovered in cementation channels by Fe scrap addition.

Around 1870, a new sulfide body of higher thickness was discovered (North Lode). This resulted in a great increase in sulfide production coinciding with the railway construction to transport the mineral. This railway ended near a new gallery at 298 m. Around 1880, open pit mining in North Lode began (Fig. 2A). Later, another new gallery (main adit at 228 m, Fig. 1) was drilled around 1915 to the west of the open pit to

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