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Abandoned mine sites as a source of contamination by heavy metals: A case study in a semi-arid zone

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Abstract

Mining and milling operations, including grinding, concentrating ores and disposal of tailings, along with mine and mill waste water, provide obvious sources of contamination in the surface environment. Climatic effects such as heavy rainfall events, have a great impact in the dispersion of metals in semi-arid areas, since soils are typically scarcely vegetated. The dispersion and influence of soluble and particulate metals present in the materials from an abandoned mine, Cabezo Rajao, in SE Spain, was evaluated. Tailings and soils were sampled and analysed for pH, EC, CaCO₃, grainsize, mineralogical composition and heavy metal content, while water samples were collected and analysed for pH, EC, soluble metals and salts. The mean concentrations of Pb, Zn, Cd, Cu and As in solid samples were 8.3 g kg⁻¹, 12.5 g kg⁻¹, 40.9 mg kg⁻¹, 332.1 mg kg⁻¹, 314.7 mg kg⁻¹ respectively, and 1.5 mg l⁻¹, 50.3 mg l⁻¹, 13.6 g l⁻¹, 17.2 mg l⁻¹, 1.7 mg l⁻¹ in water samples respectively. These metals can be dispersed downstream and downslope from the tailings by water after rainfall. Soil samples collected in the surroundings of Mar Menor Lagoon were analysed, reflecting the influence of the transport of soluble and particulate materials from Cabezo Rajao, especially of Pb and Zn. However, the presence of high amounts of carbonate in the soils around the mine area revealed the stabilization of all the metals studied. © 2007 Elsevier B.V. All rights reserved.

Keywords: Heavy metals; Abandoned mines; Metal transport; Calcareous soils; Water runoff

1. Introduction

Elevated levels of heavy metals can be found in and around disused metalliferous mines due to discharge and dispersion of mine wastes into nearby agricultural soils, food crops and stream systems (Moore and Luoma, 1990; Alloway, 1995; Jung, 2001). Heavy metals contained in the residues from mining and metallurgical operations are often dispersed by wind and/or water after their disposal (Johnson et al., 1994; Adriano, 2001). These areas have severe erosion problems caused by wind and water runoff (van Geen et al., 1999; Querol et al., 2000; Chopin et al., 2003; Razo et al., 2004) in which soil and mine spoil texture, landscape topography and regional and microclimate play an important role.

The pollution of water by dissolved metals in mining areas has mainly been associated with the oxidation of sulfide-bearing minerals exposed to weathering conditions, resulting in low quality effluents of acidic pH and

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containing a high level of dissolved metals, such as cadmium, copper and zinc, and anions, such as sulfates and carbonates, plus suspended materials (Pentreath, 1994; Salomons, 1995).

During erosion and overland flow, runoff is enriched by pollutants (Novotny, 1995), a process that occurs in two phases. Firstly, the detachment of sediments and pollutants from the parent material is selective in the case of the dissolved fraction of pollutants (ionised metals and dissolved complexes in the pore water of the soil) and for the fine soil fractions. Secondly, when rainwater reaches the soil, some metals are dissolved and enter into solution, while others remain adsorbed and/or precipitated and move with the soil particles.

The extent and degree of heavy metal contamination around mines vary depending upon geochemical characteristics and degree of mineralization of the tailings (Johnson et al., 2000). The metals released by sulfide oxidation are attenuated by precipitation, coprecipitation and sorption reactions (McGregor et al., 1998; Berger et al., 2000) in and around the mines. Such dispersion of metals and their inputs into receiving systems such as soils, sediments and waters have been the subject of numerous studies (Kwong et al., 1997; Lee et al., 2001; Kim et al., 2002).

There are several former mining sites in SE Spain (La Unión, Mazarrón amongst others), in the vicinity of which the calcareous soils can present high levels of heavy metals with a strong spatial dispersion of values (Pérez-Sirvent et al., 1999; García Rizo et al., 1999). The fate and transfer of these metals are complex and depend on the soils mineralogy and the physical transport process involved.

The present study was carried out in Cabezo Rajao (La Unión, in the northern part of the Sierra Minera, Cartagena, Spain) and its surroundings. The zone of Cabezo Rajao has an area of approximately 270,000 ha. The morphology is simple, consisting mainly of a predominantly flat area with slopes under 10%. Cabezo Rajao is an elevated promontory placed in the centre of the zone with a maximum height of 198 m, showing slopes higher than 15% that increase to 30% towards its top (Fig. 1).

Cabezo Rajao is included in one of the main mining districts of SE of Spain, La Unión, stratobound sulphide deposits in carbonate sequences in the Nevado– Filábride and Alpujárride complexes (Lunar et al., 2002) and it is included in the so called Betic. zone. This Pb–Zn–(Ag–Sn) district is an excellent example of the interplay between basin formation, normal faulting, volcanism, hydrothermal activity and mineral deposition (Manteca and Ovejero, 1992), all synergistically

combining to create exceptional concentrations of metals. Hydrothermal activity related to middle Miocene subvolcanic magmatism was focused along normal faults bounding a graben. The mineralization occurs (i) within a strongly altered zone above the Miocene footwall; (ii) in pebbly mudstone beds where the hydrothermal activity led to dissolution, void formation and mineral deposition; and (iii) in fault breccias along the normal faults that bound the Miocene sediments (Lunar et al., 2002). It is a good illustration of a PSG (pyrite, sphalerite, galena) mineralometallic outcrop associated with an intrusion of volcanic rock of intermediate basicity and hydrothermal alteration with deposited metallic ores (Manteca and Ovejero, 1992), which has transformed the original rock mineralogy to produce a complex paragenesis. These mines have been exploited since antiquity (Phoenician, Carthagenian and Roman remains have been found), although much more intensively in the 50 years preceding the 1980's, after which a crisis in the siderometallurgic sector put an end to such activity. The Sierra Minera from La Union, produced important amounts of lead, silver, zinc and pyrites until 1991. The production in the last few years was 130 t/year pyrite, contributing 40%, 60% and 12% to the Spanish production of lead, silver and zinc respectively (Dirección General del Medio Natural, 1998).

The soils of Cabezo Rajao have suffered serious human intervention in the form of mining, but natural soils developed on the volcanic rocks, resulting from the last volcanic action to occur in SE Spain, still remain. These are little evolved due to climatic conditions, the lack of vegetation and the influence of erosion phenomena (Dirección General del Medio Natural, 1998). The soils around Cabezo Rajao are calcareous (Pérez-Sirvent et al., 2002) and may occasionally be flooded after rain, when they receive substantial quantities of materials from the mining area.

The annual average temperature at the site is $17 \,^{\circ}$ C, and precipitation does not exceed 300 mm with occasional torrential rainfall which usually occur in the period between the end of summer and autumn. The superficial drainage network is defined by watercourses which are the result of the effects of the most intense rainfalls and that flow to the Mar Menor Lagoon to the north.

The materials present in Cabezo Rajao are especially prone to erosion due to a general lack of vegetation cover, causing displacement of spoil and soil particulates — a cause of some concern to nearby inhabited communities (Navarro, 2004).

The objective of this study was to evaluate the dispersion and influence of the soluble and particulate

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