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Impact of abandoned mine waste on atmospheric respirable particulate matter in the historic mining district of Rio Tinto (Iberian Pyrite Belt)

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1. Introduction

Mineral deposits record the presence of geochemical anomalies and so exhibit unusual element concentrations and mineral associations, in both ores and mine waste, which can be potential hazards to the environment and human health (Lottermoser, 2007). Traditionally, mining activities have been considered a major pollutant to the environment, mainly in soils and natural waters, although the environmental impact around mining centres varies depending on whether the mine is active or abandoned, the mining method used, and the meteorological and geological conditions (Bell et al., 2001).

The atmospheric dispersion of particles from mine sites is also an important mechanism for human exposure to metallic pollutants, especially during the active mining period (Ghose and Majee, 2000). High winds can resuspend and carry contaminated dust from mining areas to residential areas, where it can be inhaled and deposited (Zota et al., 2009).

The effects of respirable particles on the health of the population in communities close to mining areas have been shown in a number of papers using mineralogical, geochemical and source apportionment approaches (Zota et al., 2009). The application of complex statistical methods of source apportionment can be helpful in quantifying the contribution of the mine waste source to airborne particulate matter.

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ABSTRACT

This work documents for the first time the levels and composition of atmospheric particulate matter in the historic mining district of Rio Tinto (Spain) to estimate the contribution and impact of resuspended particles from hazardous mine waste on air quality. The resuspended mine waste dust contributes notably (32%) to the total concentrations of toxic trace metals (Bi, As, Cu, Pb, Cd, Zn and Sb) into the atmosphere, with the consequent impact on public health.

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We present here the composition and concentration of respirable atmospheric particulate matter (PM_{10}) in the Rio Tinto mining district of Spain. We also assess the impact of windmobilised material from mine wastes.

2. Study area

The Rio Tinto mining district is one of the oldest mining districts in the Iberian Pyrite Belt (Fig. 1A). This belt is a vast mineralised zone that extends along the southwestern margin of Iberia, hosting a large number (over 80) of volcanogenic massive sulphide deposits (Leistel et al., 1998). The Rio Tinto mines have been exploited since pre-Roman times for their valuable copper, gold and silver deposits, although the most intensive period of mining operations occurred between the end of the 19th century and the middle of the 20th century, when pyrite was the main raw material used for sulphuric acid production by the European chemical industry.

Past mining, mineral processing and smelting activities have generated huge amounts of hazardous mine waste (ore stockpiles, slag deposits, spoil heaps, mine tailings and waste rock piles) deposited over extensive areas (Fig. 1B), which are affected not only by water run-off and leaching but also by wind erosion. The most widespread waste rocks are composed mainly of primary minerals (quartz, feldspars and micas), but they also contain metal-bearing phases (pyrite with minor chalcopyrite, sphalerite, galena and arsenopyrite) and their secondary weathering products (haematite, goethite, jarosite and sulphate efflorescences). Therefore, these materials have the potential for releasing

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Fig. 1. (A) Generalized map of the Rio Tinto mining district and location of the PM_{10} sampler. The wind rose diagram for the monitoring period (from March 2009 to March 2010) is shown as an inset. (B) Partial panoramic view of the Rio Tinto mining area showing extensive tailings and waste-rock dumps. (C) Location of the high-volume PM_{10} sampler on the roof of the town hall building. The mining waste can be seen in the background.

potentially toxic and environmentally harmful trace elements, such as As, Cu, Pb and Zn, into the environment (Lottermoser, 2007; Romero et al., 2006; Chopin and Alloway, 2007; López et al., 2008; Fernández-Caliani et al., 2009).

The climate of the study area can be classified as Mediterranean, which is characterised by long, dry summers and short, mild winters, although it is modified somewhat by Atlantic influences. The annual average temperature is 15–20 °C, the annual average rainfall is 400–700 mm, and the predominant wind directions are northwest and south-southeast, as indicated by the wind rose diagram in Fig. 1 (inset) (study period March 2009–March 2010). The pronounced evaporation of the acid sulphate waters during the dry season results in the precipitation of efflorescent iron sulphate minerals on the surface of the waste piles and along the banks of streams (Lottermoser, 2007).

3. Materials and methods

A sampling site was established in the municipality of Nerva, which has approximately 6000 inhabitants (UTM coordinates: X=715,936 and

Y=4,174,804), to assess the levels and composition of PM₁₀. The PM₁₀ sampling was performed using a high volume sampler (CAV-A-PM1025, 30 m³ h⁻¹), designed and manufactured by MCV, S.A, Barcelona, Spain (http://www.mcvsa. com) equipped with PM₁₀ inlets on the roof of Nerva's town hall building (Fig. 1C). Munktell quartz filters were used in the collection of atmospheric particles. One 24-hour sample was collected every four days during the sampling period from 10 March 2009 to 25 March 2010. Accordingly, a total of 90 samples were collected for this study.

Once the mass levels of PM_{10} were obtained, acid digestion was performed following the method proposed by Querol et al. (2001) for the analysis of major and trace elements by means of inductively coupled plasma-optical emission spectrometry and inductively coupled plasma-mass spectrometry.

The soluble fraction of 1/4 of each filter was extracted with MiliQ-grade deionised water at 60 °C, and the major-ion content $(SO_4^{2-}, NO_3^{-}, Cl^{-} \text{ and } NH_4^{+})$ in the leachates was determined by means of ion chromatography. Finally, the remaining 1/4 of each filter was used to determine the total carbon (C_{total}) content using a LECO SC-144 DR instrument. The average precision and accuracy fell within the normal analytical error (in the range of 5–10%) and were controlled by repeated analysis of NBS-1633a (fly ash) reference material. The lower detection limit (LDL) for most of the elements in the solution was 0.01 ppb.

To identify different source contributions to ambient PM_{10} levels at the sampling site, a principal component analysis was performed using STATISTICA v. 4.2 on the complete chemical data series of concentrations of PM_{10} as independent variables according to the methodology described by Thurston and Spengler (1985). A factor analysis was performed to identify the possible sources, and a multi-linear

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