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Provenance and environmental risk of windblown materials from mine tailing ponds, Murcia, Spain☆

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Atmospheric particulates play a vital role in the transport of potentially toxic metals, being an important exposure pathways of people to toxic elements, which is faster and can occur in a much larger scale than water, soil and biota transport. Windblown materials in abandoned tailing ponds have not been well examined. The objectives of this investigation were: to study the major physical and geochemical properties of the materials eroded by wind inside the tailing ponds, and to understand the relative contribution of different sources to its heavy metals concentration. Study area is located in Cartagena-La Union mining district (SE Spain), where metallic mining of Fe, Pb and Zn has been developed for more than 2500 years. Wind-eroded particulates were monthly collected at 3 different heights (20, 50, and 80 cm) from the ground for a period of a full year using 4 dust collectors. Four tailing samples and 4 surface soil samples from the surrounding hills were also taken. Dust, soil, and tailing samples were examined for pH, particle size distribution, electrical conductivity, calcium carbonate content, Pb, Cu, Zn, Cd, Mn, Co, Ni, Ti and Zr concentrations. The results indicated that very coarse textured, slightly saline, and almost neutral wind-eroded deposits were generated with a very high temporal variability throughout the year. They also showed that the concentration of Cd, Mn, Pb and Zn, in the dust samples is extraordinarily high (18, 1254, 1831, and 5747 mg kg^{-1} respectively), whereas Co, Ni, and Cu had concentrations into the range of background concentrations found in the Earth's crust (3.8, 12, and 60 mg kg^{-1} respectively). Besides, the concentration of both categories of heavy metals in the dust samples was higher than that in tailing and less than that of the soils. The barren surfaces of tailing ponds and also the surface soils of the surrounding area seem to be the major contributors to the dust collected. Therefore, abandoned mines as well as their tailing ponds should be rehabilitated by proper technologies and then well stabilized and/or covered by appropriate plant vegetation to control the transfer, particularly by air, of environmentally hazardous materials to other areas.

1. Introduction

The fate and pathways of environmental pollutants have been the focus of numerous research projects, particularly in the last few decades (e.g. Ramsperger et al., 1998; Manoli et al., 2002; Tegen et al., 2002; Al-Khashman, 2004; Basha et al., 2010; Kim et al., 2014; Wang et al., 2014; Al-Harbi, 2015; Serbula et al., 2017). Despite the fact that both natural and anthropogenic sources contribute to the contamination of different ecosystems, there seems to be no major reason to very much worry about the natural environmental pollutions. Instead, different aspects of human induced contamination sources have been, and continue to be, the subject matter of numerous research papers (Csavina et al., 2012).

Significant quantities of pollutants are produced by anthropogenic activities such as industrial manufacturing (Gabarrón, 2017; Norouzi et al., 2017), farming practices (Lawrence and Neff, 2009; Csavina et al., 2012; Gabarrón, 2017), transportation and traffic (Hojati et al., 2012; Gabarrón, 2017; Norouzi et el., 2017), and

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mining operations (Csavina et al., 2011, 2012; Castillo et al., 2013; Kim et al., 2014; Wang and Liang, 2014; Karaca et al., 2017; Serbula et al., 2017). Water, air, biota, and, soil are the major transport pathways for pollutants in the environment (Csavina et al., 2012). Contaminants may be transported by air through the direct transfer of volatile materials or the indirect attachment to particulate matters. Atmospheric particulates play a vital role in the transport of pollutants in different environments, particularly those that have low solubility and volatility and remain attached to the soil particles. Transport of contaminants by atmospheric particulates appears to become more important as land use activities and projected climate change increase (Pelletier, 2006).

As compared to other transport pathways (soil, water, and biota), the transfer of pollutants by air is faster and also can occur in a much larger scale. While fine atmospheric particles can easily transfer the environmental contaminants at regional, continental, and even global scales, coarse-textured suspended particles carry the environmentally hazardous materials locally or at the landscape scale. The mechanism of contaminants transport by air is believed to be an understudied issue, especially when compared to the transport pathway by soil and water (Csavina et al., 2012).

Despite the higher importance of atmospheric transfer pathway, most studies that assess issues related to pollutant transport in the environment mainly focus on transport by water or soil. By 2012, nearly 10000 peer-reviewed papers published on the contaminant transport from mining activities by water; about half as many studies focused on that by soil; and only a few hundred studies focused on contaminant transport by atmospheric particulates. More specifically, during the same period, only a few papers were published on the transport of metal(loid)s in dust derived from mining activities (Csavina et al., 2012). In contrast, the health and environmental risks associated with the redistribution of mine-derived contaminants are potentially very high (Brotons et al., 2010).

Anthropogenic sources of atmospheric particulates are mainly associated with very high levels of chemical contaminants. They include construction operations, landfills, agricultural activities, dirt roads, pastures and feedlots, mining operations, and mine tailings (Csavina et al., 2011, 2012; Gabarrón, 2017). Among these sources, mining activities and mine tailings have been claimed as the most important anthropogenic sources of dust having the highest potential contaminant concentration as well as the highest particulate emissions (Csavina et al., 2011, 2012; Wang and Liang, 2014; Serbula et al., 2017).

Spain has a long history of mining, dating from pre-historic era up to the current time (Martín-Crespo et al., 2015). Located in the Murcia region in southeastern Spain, the "Sierra de Cartagena-La Union" constitutes the end of the Betic mountain chains where the largest Pb and Zn ore deposit in southern Europe (Gomez-Ros et al., 2013) was active for more than 2500 years (Oen et al., 1975). As a result of metals extraction during mining, particularly lead and zinc, large quantities of wastes were generated. Despite the fact that the mining activities were ceased in 1991 (Robles-Arenas et al., 2006), abandoned mines as well as the 85 tailing ponds as the legacy of the past mining operations are still part of the landscape of the area. Left behind tailing ponds are still carrying significant environmental risks. For a long time, these mining residues have been transported downstream during periods of high rainfall and atmospherically dispersed, negatively affecting natural, agricultural and populated areas (Zornoza et al., 2012a; Alcolea et al., 2015; Sánchez et al., 2017). Previous investigations in this mining area have indicated that the major components of the environment are highly contaminated with heavy metals, especially Zn, Cd and Pb (Robles-Arenas et al., 2006; Conesa et al., 2008a, 2008b; García and Muñoz-Vera, 2015).

Numerous investigations have been carried out on the impacts of abandoned mining and their tailing ponds in the Cartagena-La Union district on soils (e.g. Conesa et al., 2008a; Gonzalez-Fernandez et al., 2011b; Martínez-Martínez et al., 2013; Bes et al., 2014), plants (e.g. Conesa et al., 2006, 2007; Lambrechts et al., 2011; Párraga-Aguado et al., 2013), sediments (e.g. Robles-Arenas et al., 2006; Acosta et al., 2011; Gonzalez-Fernandez et al., 2011a, 2011c; García-Lorenzo et al., 2012; García and Muñoz-Vera, 2015), water (e.g. Alcolea et al., 2012, 2015; Trezzi et al., 2016) and even biodiversity of animals (Rodríguez Martín et al., 2014). Besides, different approaches to remediate the contaminated soils have recently been tested (e.g. Martínez-Pagán et al., 2011; Zornoza et al., 2012b; Gomez-Ros et al., 2013; Acosta et al., 2014; Martinez-Oró et al., 2017; Moreno-Barriga et al., 2017a, 2017b). However, despite the highest priority of contaminants transfer by air, as discussed above, the nature and provenance of windblown materials as well as their environmental risk have not yet been fully understood. Besides, the area has great economic opportunities in agricultural production and tourism industry (Conesa et al., 2008b). Therefore, the objectives of this research were: (i) to fully examine the major physical and geochemical properties of the materials eroded by wind inside the tailing ponds, and (ii) to investigate the origin of atmospheric dust and the relative contribution of different sources to its heavy metals concentration.

2. Material and methods

2.1. Description of the study area

The study area is located in the Cartagena-La Union mining district, southeastern Iberian Peninsula (Fig. 1). This mining district occupies an area of about 50 km^2 where there are about 20000 people living in five villages and a small size city (Bes et al., 2014). The natural vegetation in the area mainly consists of different species including more than 200 taxa from about 50 families of vascular plants, particularly xerophytic ones (Rodríguez Martín et al., 2014) and also evergreen forests dominated by *Pinus halepensis* (Párraga-Aguado et al., 2013).

Lead, zinc, and iron were the major metals being extracted from the ore deposits in this zone. Zinc and lead are present as sphalerite, galena, carbonates, sulfates and Zn- and Pb-bearing oxides. Iron occurs in sulfides, sulfates, carbonates, oxides, hydroxides, and silicates (Oen et al., 1975). Because of the long time continuous mining operations, large volumes of mining wastes, extremely contaminated with hazardous heavy metals, were generated during the concentration and smelting processes and accumulated in tailing ponds. These ponds are now mostly barren and subject to be moved and transferred to other areas by water and wind erosion posing a high environmental risk of contaminating urban, rural, agricultural, recreational, and stream and marine environments nearby.

The study area is characterized by a semiarid Mediterranean climate having about 250–300 mm of annual precipitation mostly occurring in fall and spring. The potential evapotranspiration rate in the study area is more than 900 mm $year^{-1}$ (Martínez-Martínez et al., 2013). Its annual mean temperature is 17.1 °C (Conesa et al., 2006). The landforms are low lying, with steep slopes due to its proximity to the Mar Menor coast.

There are 85 mine tailing ponds in the Cartagena-La Union mining district (Martínez-Martínez et al., 2013), one of the most extensive ones of which, called El Lirio, was selected for this study. This tailing pond presents an area of 73 000 m^2 and a volume of 750 000 m^3.