



Composition and risk assessment of roasted pyrite ash from fertiliser production

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HIGHLIGHTS

- Variation in metal accumulation depends on the raw material of pyrite ash.
- Roasting process bounds Zn, Cu, Cd and Ni to labile phases.
- Pb shows high mobility but low leachability.
- 60% of metals in pyrite ash are associated with respirable particles.
- Ecological risk index (RI) does not report risk for the environment by any particle size.

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ABSTRACT

Pyrite ash is a residue from the roasting of pyrite ores to obtain sulphuric acid used in the fertiliser industry and its production is widely extended worldwide. The mismanagement of this waste may result in environmental and health damages due to its physico-chemical characteristics. The main objective of this study was to examine the physico-chemical and mineralogical composition of roasted pyrite ash from an abandoned fertiliser company, and to evaluate the environmental risk caused by the wind and water dispersion of metals posed by this waste. In order to achieve these objectives, a sequential extraction procedure and a physical fractionation into six size fractions: >100, 100–50, 50–20, 20–10, 10–2.5 and < 2.5 μm were applied. Results showed that pyrite ash is composed mainly of iron-oxides such as hematite (46%) and secondary minerals as anglesite and shows high concentrations of Pb (7464 mg kg^{-1}), Zn (2663 mg kg^{-1}) and Cu (585 mg kg^{-1}). The highest Risk Assessment Code (RAC) values were found for Cd, Pb and Zn, bound to the more labile fractions. Conversely, Pb showed the lowest water solubility due to the covering effect provided by a coating of anglesite in the pyrite ash surface. Most of the metals were associated to both the coarsest (>100 μm) and the finest (2.5–10 μm) fractions, although none represented an environmental risk according to the ecological risk index results. However, 30% of the metals were bound to the respirable fraction ($\leq 100 \mu\text{m}$) posing a potential risk for human health and a high potential dispersion by wind to the surrounding areas.

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1. Introduction

The production of sulphuric acid by pyrite roasting has been an industrial activity widely extended in the mid-20th century in Spain. The worldwide production of sulphuric acid in 2012 exceeded 230 million tonnes being the production of phosphate fertiliser

materials, especially wet-process phosphoric acid, the major end-use market for sulphuric acid, accounting for over 58% of total world consumption in 2014 (Chemical Economics Handbook, 2014). Asia was the leading producer of sulphuric acid (45% of the overall production), and China, US, India, Russia and Morocco are the top five sulphuric acid producing countries, although Eastern and Western Europe were also large users, representing a big challenge worldwide (Merchant research and consulting, 2017).

The production process of sulphuric acid consists of the roasting of pyrite ores to produce solid oxides and gaseous sulphur dioxide

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at temperatures of 600–1000 °C. After cleaning and cooling, the sulphur dioxide in the roasting gas is further processed to sulphuric acid (Runkel and Sturm, 2009). The roasting process leaves a residue consisting of iron oxide (mainly hematite), a considerable amount of metals and a small amount of sulphur (Oliveira et al., 2012; Pérez-López et al., 2009). Unfortunately, the management of the roasted pyrite ashes in Spain in the latest mid-20th century was low or non-existent (Domenech et al., 2017; Soriano-Disla et al., 2018).

The mismanagement of residual pyrite ashes resulted in environmental impacts such as acid drainages and acidification, water erosion and runoff, loss of vegetation and wind erosion, thus promoting the spreading of particles containing metals. The dispersion of metals depends on the particle size and the surface properties of the material on which they are deposited (Kong et al., 2011).

The main risk for the human health is from the high prevalence of metals in the environment and their mobility. Studies have revealed that the presence of metals in the environment can cause damages to human health such as skin and lung cancer, kidney damage, cardiovascular diseases, dizziness and fatigue and anaemia (Wilson and Pyatt, 2007; Abbas and Abdelhafez, 2013). Metals can enter the human body through dietary intake, soil ingestion, dermal absorption or dust inhalation (Wilson and Pyatt, 2007). Therefore, the mobility of metals and the particle size of pyrite ashes are key factors to determine the environmental and human risks. Thus, to identify the main binding sites of each metal is necessary to understand the geochemical processes that govern metal mobilisation and potential risks (Yuan et al., 2004; Acosta et al., 2014). Sequential extraction procedures provide information about the main binding sites, the strength of metal binding to particles and the phase associations of trace elements in the residue (Rauret, 1998). Hence, the metal mobility and bioavailability determined by the sequential extraction procedure are critical for the environmental risk assessment of roasted pyrite ashes (Pérez-López et al., 2009).

The importance of analysing the particle size distribution and metals associated to each fraction is due to the ability of fine particles to retain large amounts of metals due to their high specific area (Kong et al., 2011). Fine particulate matter such as PM10 and PM2.5 are susceptible to wind erosion and hence they are quite mobile in the environment (Acosta et al., 2009). These particle sizes are also respirable, being the inhalation the main way by which these particles reach the human body. Thus, they pose a potentially hazardous risk threat to human health.

The main objectives of this study were (1) to examine the mineralogical and physico-chemical composition of pyrite ashes collected from a legacy fertiliser company and (2) to evaluate the environmental risk of pyrite ashes dispersion by water and wind.

2. Material and methods

The study area was located close to a population settlement, in a legacy industrial area of Cartagena (SE Spain) that was affected by the sulphuric acid and P– fertilisers production. The climate of the area is Mediterranean semiarid with an annual average temperature of 17 °C and annual average precipitation of 313 mm. The predominant wind directions are northeast and southwest (Capel-Molina, 1986) and the area surrounding the pyrite ash pond is geologically composed of carbonate rocks and the resulting materials of transported sediments on which soils have developed (Ortiz, 1986).

2.1. Sample collection and analytical methods

Ten surface pyrite ash samples (0–30 cm) were collected using a

soil spade and packed in plastic bags to be carried to the laboratory. Each sample was composed of five subsamples.

Samples were dried in a forced-air oven at 45 °C for 72 h and sieved through a 10 mm mesh sieve for the leaching test (UNE-EN 12457–4). In addition, one subsample was sieved through a 2 mm sieve for physicochemical analysis and another subsample was ground with an agate mortar (RetschRM 100) for the total metal analysis.

The pH was measured in a solution of 1:2.5 soil/water ratio (Soil Survey Staff, 2004) Electrical conductivity (EC) was measured in a 1:5 soil/water suspension (Soil Survey Staff, 2004).

The total concentration of metals (Cd, Cr, Cu, Ni, Pb and Zn) was analysed using 0.5 g of ground pyrite ash, by placing the pyrite ash into vessels with 10 ml of nitric acid (65%) and digested using microwave according to the US-EPA method 3051 (USEPA, 2007).

Chemical distribution of metals was determined by a modified Tessier sequential extraction procedure (Tessier et al., 1979; Li et al., 1995). The following five fractions were obtained: exchangeable (8 ml of 0.5 M MgCl₂; acid soluble (8 ml of 1 M NaOAc adjusted to pH 5.0); oxidizable (30 ml of 0.04 M hydroxylamine hydrochloride with 25% HOAc), reducible (3.3 ml of 0.02 M HNO₃ adjusted to pH 2 and 5 ml of H₂O₂) and residual (microwave digestion with 10 ml of nitric acid (65%) (USEPA method 3051). All the samples in each fraction were centrifuged at 2000 rpm for 20 min and filtered.

The total metal concentration in the bulk sample and in each fraction from the sequential extraction was measured using atomic absorption spectrometer (AAAnalyst 800, Perkin Elmer). Reference material (BAM-U110) from the Federal Institute for Material Research and Testing (F.I.M.R.T.2010) and reagent blanks were used as quality control samples during the analyses. Metal recoveries from the bulk samples were as follows: 92–109% for Cd, 91–98% for Cu, 94–105% for Pb and 91–99% for Zn. For the sequential extraction, the recoveries were as follows: 94–106% for Cd, 97–108% for Pb, 99–102% for Ni, 91–102% for Ni, 95–99% for Cu and 95–101% for Zn.

The mineral composition of pyrite ash was determined by X-ray diffraction (XRD) and scanning electron microscopy (SEM). X-ray diffraction analysis was conducted on randomly oriented ground samples using a Bruker D8 X-ray diffractometer with a GADDS™ area detector system operated at 40 kV and 20 mA to generate a monochromatic Co-K α X-ray radiation. The GADDS™ detector was programmed to collect the diffracted beam for 3 min in four locations to cover a 2 θ from 3° to 90°.

The morphology of pyrite ash samples was observed using a Scanning electron microscope (SEM) (Philips XLS-30) equipped with an energy-dispersive system (EDS) to determine the chemical composition of each observed particle. Sample for SEM analysis was mounted on an aluminium stub and coated with platinum for 3 min using a Denton™ vacuum system before submicroscopic observations. The SEM was operated at 15 keV and 1.94 A filament current. In situ chemical composition of particles was recorded in EDAX spectrum collected either in spot (~1 μ m) or full field of view mode for 400 s. Semi-quantitative (\pm 5%) elemental composition of particles was corrected for Z (atomic number), A (absorption) and F (fluorescence) factors.

2.2. Particle size separation

For the evaluation of environmental risks by wind dispersion, pyrite ash samples were separated into six different particle sizes: \leq 2.5 μ m, 2.5–10 μ m, 10–20 μ m, 20–50 μ m, 50–100 μ m and >100 μ m. Particle sizes of 50–100 μ m and >100 μ m were obtained by sieving the subsamples of 2-mm sieved samples. Sizes below 50 μ m were obtained by repeated sedimentation and decanting based on Stokes' law (Ljung et al., 2008; Boisa et al., 2014; Li et al.,

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