Article preview
Abstract
Introduction
Section snippets
References (67)
Cited by (45)

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# Atmospheric dust characterisation in the mining district of Cartagena-La Unión, Spain: Air quality and health risks assessment

Isalyne Blondet [a], Eva Schreck [a], Jérôme Viers [a], Sandra Casas [b], Irene Jubany [b], Neus Bahí [b], Cyril Zouiten [a], Grégory Dufréchou [a], Rémi Freydier [c], Corinne Galy-Lacaux [d], Silvia Martínez-Martínez [e], Angel Faz [e], Martin Soriano-Disla [e], Jose A. Acosta [e], José Darrozes [a]

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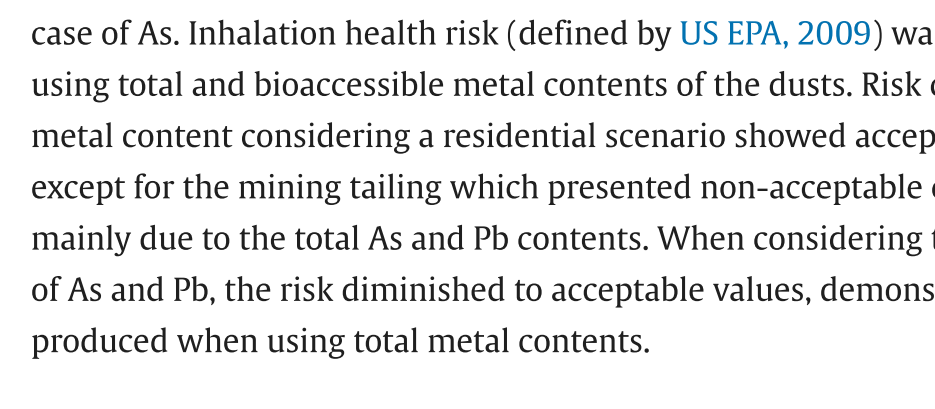
## Highlights

- Atmospheric dusts were recorded using Owen gauges in 4 sites of the mining area.
- High contents of Zn, Pb, As, Cd and Sb were measured in atmospheric fallouts.
- Atmospheric deposition in the urban area mainly contained sulphates and sulphides.
- Cancer and hazard risks above thresholds were detected in the main tailing area.
- The risk falls below the acceptable level when considering bioaccessibility factors.

## Abstract

Nowadays, air pollution has a major impact on the environment and human health. Owen gauges allow the sampling of atmospheric depositions in polluted sites for further characterisation. This paper shows the study of the air particles of an old mining zone in Cartagena-La Unión (South-east of Spain) in order to quantify their potential risk on human health. There were 4 strategic sites monitored: the main mining tailing (Avenque), the urban area (La Unión), an agricultural zone (formerly mining) and a site in the Mediterranean coast. Physico-chemical and mineralogical characterisation was applied to atmospheric fallouts. The granulometry revealed a dust particle size around 15 μm, with the coarsest particles in the urban area and the thinnest at the tailing site. XRD analyses showed the presence of quartz, carbonates, sulphides and sulphates. Observations with SEM-EDX confirmed chemical spectra and allowed us to classify the particles into well-crystallised minerals and heterogeneous dusts. Total metal content determination was carried out by ICP-MS analyses and results showed Zn, Pb, As and Cd fluxes (respectively 2549, 1275, 68 and 7 $\mu g \cdot m^{-2} \cdot d^{-1}$) exceeding the limit values set by European legislations in the mining area. The fluxes of Zn, Pb and As also exceed these standards in the urban area whereas the coastal zone only exceeds the thresholds in the case of As. Inhalation health risk (defined by US EPA, 2009) was quantified in the sites using total and bioaccessible metal contents of the dusts. Risk calculations using total metal content considering a residential scenario showed acceptable risk for all sites except for the mining tailing which presented non-acceptable cancer and hazard risk mainly due to the total As and Pb contents. When considering the bioaccessible fraction of As and Pb, the risk diminished to acceptable values, demonstrating the overestimation produced when using total metal contents.

## Graphical abstract


AGRICULTURAL AREA
Las Matildes mine
URBAN AREA
La Unión
MINING AREA
Avenque Tailing
COASTAL ZONE
Playa del Gorguel
Sulphates, sulphides, carbonates, silicates, chlorides
XRD; SEM-EDX
Accumulation of Zn, Pb, As, Cd, Sb
Non-significant risk by inhalation
15 μm
AIR QUALITY : Sampling by Owen gauges (6 months - May to November 2017)
Dust characterisation
Assessment of health risk by inhalation

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## Introduction

In Europe, >3 million sites are listed as polluted areas due to anthropogenic activities and 250,000 of them need to be urgently remedied (Jones et al., 2012). Among human activities, extractive and especially mining ones are important sources of metallic contamination in the environment, particularly due to highly toxic and persistent heavy metal emissions in the different environmental compartments (Boamponsem et al., 2010). The fate of stored mining residues remains a very high concern because the associated risk depends on metal contents but also on the mineral species in presence. Actually, according to abiotic parameters (such as pH, temperature, redox potential, *etc.*), metal speciation can change and enhances their bioavailability, and then their toxicity for the environment and the human populations (Goix et al., 2014; Navel et al., 2015).

The “Sierra Minera” of Cartagena-La Unión corresponds to a large zone of mining activities in the south-east of Spain whose first exploitation by the Carthaginians (Murcia county, n.d.) is the oldest in Europe and dates back to the 3rd century BCE (Gonzalez-Fernandez et al., 2011). Extractive processes were taken over by the Romans producing about 45,000 tons year$^{-1}$ of lead during the 1st and 2nd century CE (Moreno-Grau et al., 2002). Neglected until the 19th century, its exploitation was then extended to a larger scale. But now, this region includes nearly 40 abandoned mine tailings on an area of about 160 ha (Martínez-Orozco et al., 1993), in between the town of La Unión and the Mediterranean Sea.

Mining activity in Sierra Minera historically focused on the extraction of lead (Pb), zinc (Zn) and iron (Fe) as reported by Alcolea-Rubio (2015). Various studies have thus been carried out in this area for assessing the contamination of soils and plants, as well as surface and groundwater (Acosta et al., 2011; Alcolea-Rubio, 2015; Conesa et al., 2006, Conesa et al., 2008, Conesa et al., 2009; Conesa and Faz, 2010; Garcia et al., 2008; Garcia-Lorenzo et al., 2012; Gonzalez-Fernandez et al., 2010; Martínez-Sánchez et al., 2008; Moreno-Grau et al., 2002; Robles-Arenas et al., 2006). Conesa et al. (2008) estimated that the contents of Zn, Pb and As in the main tailing (Avenque Tailing), near the city of La Unión, are about 9100, 5200 and 350 mg kg$^{-1}$, respectively. Moreover, several investigations (Conesa et al., 2006, Conesa et al., 2009; 2010; Gonzalez-Fernandez et al., 2010) on plant species naturally growing on these contaminated soils have reported that Pb and Zn are bioaccumulated in plant shoots, highlighting a global contamination risk. However, except for Sánchez Bisquert et al. (2017) who studied metallic contaminations in soluble and insoluble fractions of atmospheric deposits in the large scale of 50 km$^2$ from the plain of Campo de Cartagena to the Mar Menor touristic area, the air quality in this mining district of Cartagena-La Unión remains poorly described (especially in terms of dust physico-chemical characterisation and health risk assessment), even if it becomes a major challenge for human health.

Actually, recent studies have reported that in the last 50 years, atmospheric pollution has become a growing public health concern worldwide and accounts for almost 10% of all deaths according to the Environmental Performance Index (EPI, 2016). Indeed, >3.5 billion people live in countries with unsafe levels of air pollution (Yang and Wang, 2017). Air quality and climate change from local to global scales are also influenced by airborne emissions from human activities in urban, industrial and mining areas (Baklanov et al., 2016).

Thus, as the mining wastes are stored near urban, touristic and agricultural areas in this “Sierra Minera” of Cartagena-La Unión, pollution dispersion could involve severe and growing damages for the environment and human health, especially due to these atmospheric fallouts (Csavina et al., 2012). Wind erosion can resuspend and transport metallic-rich particles over long distances from mine tailings to the surrounding urban and agricultural areas (Kulkarni et al., 2011). Resuspension of large quantities of particles from mine waste deposits, whether active or abandoned and mainly associated with wind events, result in “atmospheric dusts”, corresponding to dry and wet depositions (Castillo et al., 2013; Sánchez Bisquert et al., 2017). These depositions from mining sites present severe toxicity due to their high contents of heavy metals (Golomb et al., 1997) and especially the finest particulate matters (PM, with a diameter <10 μm, $PM_{10}$ or even 2.5 μm, $PM_{2.5}$) that can reach the pulmonary alveoli and potentially the blood system (Goix et al., 2011). Many health problems have been already identified (respiratory and cardiovascular diseases, intestinal disorders, keratoses, skin cancers, neurological effects and mutagenic pathologies) during inhalation, contact or ingestion of atmospheric particles (Boamponsem et al., 2010; Wise et al., 2017). Several studies have also reported positive correlations between the effects of atmospheric dusts on human health due to heavy metal contents and the distance between ore extraction and inhabited zones (Munksgaard and Parry, 1998; Navel et al., 2015). However, although there is a difficulty in establishing direct relations of the total amount of metals in the air particles with inhalation effects on human health, exposure assessment methods are commonly used to quantify the potential associated risks. Conventional exposure assessment methods can overestimate the risks associated with these locations as they are based on the total metal(loid) content of the air particles, which cannot be totally bioavailable. Bioavailability is defined as the potential of the metal(loid) to mobilise from the soil matrices to the blood stream; bioaccessibility is then defined as the potential to reach the human digestive or respiratory systems (Hernández-Pellón et al., 2018). Bioaccessibility by inhalation can then be estimated using*in-vitro* tests, simulating pulmonary fluids and inhalation conditions, and is now considered as a rapid, simple and low-cost method especially useful for rapidly predicting the health risk in these kinds of areas (Guney et al., 2017).

Moreover, in a global context, the current situation of contaminated sites is expected to worsen due to climate change. Actually Alcolea-Rubio (2015) states that, in this semi-arid region, global warming could lead to an increase of aridity indices and enhance atmospheric transport of heavy metals. As an example, the mass balance of metals on the Mar Menor (coastal salty lagoon in the north of the Sierra Minera) shows that metal transfer due to wind erosion represents 81%, while the groundwater contributes to 16% and the runoff to 3% (Alcolea-Rubio, 2015), suggesting a high impact of contaminant dispersion thought the air.

Taking in considerations all these statements, it becomes therefore essential to determine the air quality within this mining area of the Cartagena-La Unión district in order to accurately quantify the associated health risks for the local population. This study aims to sample atmospheric dusts in an area of 25 km$^2$ on 4 strategic sites (urban, agricultural, coastal and mining areas) in order to carry out a physico-chemical characterisation of the dust deposits, quantify the elemental contents (heavy metals especially) and finally, evaluate the potential health risk for the population considering total content and specific bioaccessibility factors of the metals of concern. For that, a combination of physical and chemical methods (X-ray diffraction, high-resolution scanning electron microscopy: SEM–EDX and laser-granulometry) was applied for the analysis of samples of atmospheric fallout dusts from the selected locations of the contaminated area, and bioaccessible *in vitro* tests were conducted for health risk assessment. This made it possible to determine the pollution distribution, the contaminant fluxes existing from the mining sources to the Mediterranean Sea and the health risks potentially induced in people living in this mining environment, and finally inform the population, as a part of the global SOIL TAKE CARE project (under the Interreg SUDOE Program) which combines several approaches, both scientific and societal.

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## Section snippets

### Study area

The study area of the mining district of La Unión is located in the Sierra Minera, at 10 km from the city of Cartagena in South-Eastern Spain. The Sierra Minera, with an area of approximately 50 km$^2$, is part of the betic chain set up in the Miocene (5.3 to 23 million years, (SIGES, 2017)). At the end of this period, the post-orogenic magmatic activity associated with a hydrothermal circulation allowed for the creation of polymetallic ore deposits (Garcia et al., 2008). The surrounding rocks of …

### Total dust deposition

The Owen gauges placed for 6 months at the 4 strategic sites raised an average of 39 g m$^{-2}$ year$^{-1}$ of dusts (corresponding to dry and wet deposits) for the first sampling campaign. The mining (Avenque tailing) and the urban (La Unión) sites had the highest sampled masses with respectively 60 and 51 g m$^{-2}$ year$^{-1}$. The coastal zone near the Mediterranean Sea showed a deposition of 31 g m$^{-2}$ year$^{-1}$. Finally, the agricultural (and formerly mining) area of Las Matildes had the lowest atmospheric …

### Physico-chemical description of dusts

The atmospheric dusts collected near the mining tailing were those with the finest grain size, suggesting a potential impact on the environment near the Avenque tailing. Indeed, two characteristic peaks were found at 0.5 and 10 μm, but the finest particles, with diameters <10 μm ($PM_{10}$) or even thinner $PM_{2.5}$, are reported to be the most dangerous because they could reach, after inhalation, the pulmonary alveoli and potentially the blood system (Goix et al., 2011). The fine grain size found at …

### Conclusions

The atmospheric dusts collected in the Owen gauges had a mean particle size around 15 μm with, not surprisingly, the finest particles present at the mine tailing area—probably due to ore treatments of the long past mining activity. The urban area showed coarser grain size (with a peak at 50 μm), possibly related to human activity in the city. These atmospheric dusts included, among others, sulphides (iron, zinc and lead), sulphates and carbonates, found in particular in urban and agricultural …

### Acknowledgments

The European Union is acknowledged for co-funding SOIL TAKE CARE SOE1/P4/F0023 through the European Regional Development Fund (ERDF), under the Interreg SUDOE Program. We thank Ludovic Menjot and Priscia Oliva from the GET Laboratory for their technical and scientific help in XRD data acquisition and interpretation. We want to also express our gratitude to our colleagues Frédéric Julien from EcoLab and Stéphane Le Blond du Plouy from the Centre de MicroCaractérisation Raimond Castaing for their …

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Revealing the release and migration mechanism of heavy metals in typical carbonate tailings, East China
2024, Journal of Hazardous Materials
Hide abstract

Refining the occurrence characteristics of tailings hazardous materials at source is of great importance for pollution management and ecological reclamation. However, the release and transport of heavy metals (HMs) from tailings under rainfall drenching in simulated real-world environments is less well portrayed, particularly highlighting the inherent neutralisation in tailings wastes under superimposed dynamic conditions. In this study, dynamic leaching columns simulating actual conditions were used to observe the release and transport of HMs from tailings under acid rainfall infiltration at spatial and temporal scales. The release rate of trace elements (e.g., As, Cr, Ni, Pb, Cd) is high. Neutralisation in the presence of carbonate rocks in the gangue reduces HMs release intensity from tailings with high heavy metal content, along with the precipitation of iron oxides and chromium-bearing minerals, etc. In addition, the vertical differentiation of HMs is more relevant to physical processes. In the absence of carbonate rocks in gangue, the lowest pH value is reached within 1.2 h after acid rain infiltrates the tailings. At the same time, Cu, Zn and Cd are released significantly from the minerals at the superficial level. The release of As(III) is mainly concentrated in the early and late stages of water-rock contact.

Environmental hazards posed by mine dust, and monitoring method of mine dust pollution using remote sensing technologies: An overview
2023, Science of the Total Environment
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The over-exploitation of mineral resources has led to increasingly serious dust pollution in mines, resulting in a series of negative impacts on the environment, mine workers (occupational health) and nearby residents (public health). For the environment, mine dust pollution is considered a major threat on surface vegetation, landscapes, weather conditions and air quality, leading to serious environmental damage such as vegetation reduction and air pollution; for occupational health, mine dust from the mining process is also regarded as a major threat to mine workers' health, leading to occupational diseases such as pneumoconiosis and silicosis; for public health, the pollutants contained in mine dust may pollute surrounding rivers, farmlands and crops, which poses a serious risk to the domestic water and food security of nearby residents who are also susceptible to respiratory diseases from exposure to mine dust. Therefore, the second section of this paper combines literature research, statistical studies, and meta analysis to introduce the public mainly to the severity of mine dust pollution and its hazards to the environment, mine workers (occupational health), and residents (public health), as well as to present an outlook on the management of mine dust pollution. At the same time, in order to propose a method for monitoring mine dust pollution on a regional scale, based on the Dense Dark Vegetation (DDV) algorithm, the third section of this paper analysed the aerosol optical depth (AOD) change in Dexing City of China using the data of 2010, 2014, 2018 and 2021 from the NASA MCD19A2 Dataset to explore the mine dust pollution situation and the progress of pollution treatment in Dexing City from 2010 to 2021. As a discussion article, this paper aims to review the environmental and health risks caused by mine dust pollution, to remind the public to take mine dust pollution seriously, and to propose the use of remote sensing technologies to monitor mine dust pollution, providing suggestions for local governments as well as mines on mine dust monitoring measures.

Evidence supporting the value of spontaneous vegetation for phytomanagement of soil ecosystem functions in abandoned metal(loid) mine tailings
2021, Catena
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This work studies a set of soil indicators (physical, physico-chemical and biological), evaluated under field and laboratory conditions, in different stages of spontaneous vegetation colonization in abandoned metal(loid) mine tailings from Mediterranean semiarid areas. The results provide evidence about the interest of spontaneous colonization by native vegetation for the phytomanagement of abandoned metal(loid) mine tailings in terms of providing ecosystem functions. Bare soils (B), small groups of pine trees (P), scattered pine trees with shrubs and herbs under the canopy (P+S), and dense patches of pine trees with shrubs and herbs under the canopy (DP+S) were studied inside mine tailings abandoned ≈40 years ago. Besides, pine forests next (FN) and away (FA) from the tailings were also studied. Pioneer and nurse plants were mainly found inside the tailings, although ecological indexes in P+S and DP+S were similar to FN and FA. Pedogenesis evidences such as structure development and increase in cation exchange capacity, organic C and N were found in tailing soils from B to DP+S. However, soil metal(loid)s did not follow the same variation pattern. For example (in mg kg$^{-1}$): P showed the maximum total Cu (≈277) and Zn (≈17,860), while P+S of As (≈1250) and Pb (≈14,570). B had the maximum water extractable Pb (≈4) and Zn (≈207), while FA of As (≈0.192) and Cu (≈0.149). Soil microbial biomass C, enzyme activity, $CO_2$ emission, organic matter decomposition and feeding activity of soil dwelling organisms indicated similar, or even higher, biological activity in P+S and DP+S than in FN and FA. In fact, FA showed the highest soil ecotoxicity risk (reduced enchytraeid reproduction). Therefore, mine tailing soils can be effectively modified following spontaneous vegetation colonization, achieving conditions with capacity to provide certain ecosystem functions. Hence, phytomanagement of these tailings should be preceded by a detailed knowledge of the existing spontaneously colonized sites, which should be preserved to take advantage of their potentiality.

A review of dust accumulation and cleaning methods for solar photovoltaic systems
2020, Journal of Cleaner Production
Hide abstract

Utilizing solar energy to generate electricity on large scale photovoltaic (PV) power plants became a trend as a new option adopted by many countries. The optimum installation of PV power plants depends on the geographical location, which specifies irradiation, latitude, longitude, tilt angle, orientation, etc. However, the PV panel affected by many environmental parameters, which has a significant impact on the power productivity, conversion efficiency, and cost of energy. Dust is one of the essential parameters that affect PV panel performance, yield, and profitability. However, the dust characteristics (type, size, shape, meteorology, etc.) is geographical site specified. Many researchers investigated PV panel dust cleaning and mitigation methods. This paper put into perspective the recent investigations of dust impact on PV systems and decent cleaning methods. It is found that daily PV power losses and monthly efficiency reduction due to dust in some locations is more than 1% and 80%, respectively, which is relatively high. The present paper aims to provide an appraisal of dust problem and cleaning methods status, challenges, and prospects. Also, a critical review is developed for researchers interested in this field. It is found the main dust elements which have impact are six dust pollutants. Also, the study conclude that selection of the dust cleaning method depends on many parameters in term of technical and economic aspects. Finally, this paper contains a comprehensive review of dust problem and cleaning methods for engineers, designers, and researchers dealing with PV systems.

Accumulation of heavy metals in phytoliths from reeds growing on mining environments in Southern Europe
2020, Science of the Total Environment

> *Citation Excerpt :*
> …These factors facilitate metal lixiviation, which is further accelerated by the overly acidic character of soils. High concentrations of As, Cd, Cu, Pb and Zn were observed in Cartagena soils, consistent with results of Conesa et al. (2006) and Blondet et al. (2019). Besides, elevated concentrations of Mn, Sb and Sn were measured in these soils.…

Hide abstract

In Southern Europe, soil contamination by heavy metals (HM) due to mining and industrial activities is a long-known problem. Yet, despite soils being widely contaminated through decades, some plants are still able to grow. Some of these plants, like giant reed (*Arundo donax*) or common reed (*Phragmites australis*) are capable of accumulating substantial amount of HM. These reeds also contain small silica structures in their shoots, called phytoliths. However, the role of phytoliths in reducing stress caused by these HM remains unknown. The aim of this work is then to determine if phytoliths represent a preferential structure for the bioaccumulation of HM in plants. Therefore, plants from mining-contaminated sites in Spain and France were sampled and HM concentrations in total plant shoots were compared to those in phytoliths for eight metal(oid)s: As, Cd, Cu, Mn, Pb, Sb, Sn and Zn. Results show that *Arundo donax* and *Phragmites australis* tend to accumulate Cd, Sb and Sn but limit the uptake of As, Cu, Mn, Pb and Zn in plant shoots despite that the concentration of these HM in soil is quite high. Therefore, reeds appear as tolerant to high metal concentrations in soils, and phytoliths are identified as preferential structures for encapsulation of As, Cu, Mn, Pb and Zn, while Cd, Sb and Sn were mainly found to be accumulated in organic tissues rather than in phytoliths.

Tillandsia usneoides as biomonitors of trace elements contents in the atmosphere of the mining district of Cartagena-La Unión (Spain): New insights for element transfer and pollution source tracing
2020, Chemosphere
Hide abstract

Nowadays, atmospheric pollution has a major impact on the human health and the environment, encouraging the development of biomonitors of the air quality over a wide zone.

In this study, the relevance of the epiphyte plants *Tillandsia usneoides* is studied to estimate the transfer of metal(loid)s from a former Zn and Pb mining zone in the Southeast of Spain (Cartagena-La Unión) to the local atmosphere. Biomonitoring was performed by installing plants in 5 sites along a transect from the main mining area to the urban and the coastal zones. An aliquot of plants was collected in every site every 2 months over 1 year. The *Tillandsia usneoides* have been observed with SEM-EDX, and analysed by ICP-MS to determine trace element concentrations, magnetic susceptibility signals and Zn and Pb isotopes ratios.

Results show that atmospheric particles are distributed homogeneously at the plant surface. By comparing elemental contents in *Tillandsia usneoides* with regard to the values of the geochemical background of the region of Murcia, significant enrichments are observed in the epiphyte plants for Sb, As, Cd, Zn and Pb.

The statistical analyses (decentred PCA and PLS) also suggest that the kinetics of dust deposition is slower for the urban and coastal sites compared to the mining sites and highlight an influence of agricultural activities in Cu deposition. The similarity of isotopic compositions (Zn and Pb) between *Tillandsia usneoides*, soils and atmospheric particles also put in evidence that these plants could be a powerful tool to trace the source of matter in the atmosphere. Finally, this experiment provides new insight to better understand the foliar absorption mechanisms.

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