PROVENANCE AND ENVIRONMENTAL RISK OF WINDBLOWN MATERIALS FROM MINE TAILING PONDS, MURCIA, SPAIN

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Abstract

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.

Keywords: Atmospheric particulates, Heavy metals, Mine tailings, Environmental risk, Source identification

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 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-Roz 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 at al., 2014), plants (e.g. Conesa et al., 2006; Conesa et al., 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 and 2011c; García-Lorenzo et al., 2012; García and Muñoz-Vera, 2015), water (e.g. Alcolea et al., 2012 and 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 and 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.

2.2. Sampling

Wind-eroded materials were monthly collected at 3 different heights (20, 50, and 80 cm from the ground surface) for a period of a full year starting from June 2011 and ending in May 2012. A dust collector designed, tested, and used by Brotons et al. (2010) was employed. This multicollector (Fig. 1) consisted of a supporting central PVC tube to which a set of 8 collectors

were connected at 8 major geographical directions (N, S, W, E, NW, NE, SW and SE) at each of the 3 different heights including 20, 50, and 80 cm above the ground surface. Each collector was 6.5 cm wide and 12.5 cm long. Although this collector was not aerodynamic constructed, it was selected for this study because Brotons et al. (2010), after tested conventional wind deposition collector (Fryrear, 1986) as well as various wind-drive particle collectors built in others studies (García et al., 2004 and Moral et al., 2005) in the same area of this study, concluded that this was the best configuration for collecting dust particles from tailing ponds and under semiarid climate.

In order to collect the particles eroded from the tailing pond, four multicollectors were installed following a symmetric square distribution (100x100 meters) where the each collector was located in the vertexes of this square, covering the surface of the pond (Fig. 1). At the beginning of each month, plastic bags were connected to each collector and there were taken at the end of the month. Eight dust samples belonging to different geographic direction at each height were mixed to make a composite dust sample. Monthly sampling was continued for a full year from June 2011 to May 2012. Therefore, a total of 144 composite dust samples were taken.

One sample was taken from each of the four terraces of the tailing pond and four natural surface soil samples (0-10 cm) were taken from the northern and southern hills of the tailing pond where natural pine trees have made a forest canopy (Fig. 1). Each sample was the results of mix and homogenized three subsamples random distributed from each sampling location. Soils/tailing waste samples were analyzed for their geochemical characterization in order to evaluate from where the particles collected in the sampler came from.

Dust, soil, and tailing samples were transferred to the lab, air-dried at 40ºC for 48 h, passed through a 2 mm sieve and stored in plastic bags for analysis. A split of each sample was ground using an agate mortar (RetchRM 100).

2.3. Laboratory analysis

The pH of dust, soil, and tailing materials was measured in a ratio of 1:2.5 sample to deionized water suspension after 1 h of mechanical agitation and leaving overnight using a pH meter model GLP 21, CRISON. Their EC values were also determined and corrected for 25 ºC in the same suspension using an electrical conductivity meter model GLP 31, CRISON. Calcium carbonate equivalent (CCE) was measured in all the samples by the titration method (Soil Survey Staff, 2004).

Dust, soil, and tailing samples were analyzed for their particle size distribution. About 500 mg of each sample was dispersed in 1 ml of 10% sodium hexametaphosphate solution and 9 ml of deionized water prior to particle size distribution analysis (McTainsh et al., 1997) and then introduced to a Malvern Hydro 2000g laser particle size analyzer (MASTERSIZER 2000LF, Malvern Instruments). Each sample's particle size distribution was determined 3 times and the average of 3 readings was recorded.

Pseudototal metals concentration from the dust, soil, and tailing samples was determined using USEPA method 3051A (Environmental Protection Agency, 2007). About 500 milligrams (dry weight) of ground sample was placed into teflon vessels that contained 10 ml of concentrated HNO_3 (65% pure, Panreac), and digested using a microwave digester (Mars 6, 240/50 CEM company). Furthermore, the digested samples were diluted with deionized water to a final volume of 50 ml.

Certified reference materials (BAM-U110), available from the Federal Institute for Materials Research and Testing, as well as reagent blanks were run to test the quality of analyses. The concentration of heavy metals in dust, soil, and tailing extracts including Cd, Co, Cu, Ni, Mn, Pb, and Zn as well as that of Zr and Ti were measured by an Inductively Coupled Plasma Mass Spectrometer (ICP-MS) model Agilent 7500CE. The recoveries of metals ranged from 85.3% for Co to 105.8% for Cu.

2.4. Statistical analysis

To ensure the fitting of the data to a normal distribution the Kolmogorov-Smirnov test was applied. The relationship among metals concentrations in collected particles were studied by Spearman correlations analysis. All statistical analysis was performed using the statistics software SPSS 23 (IBM).

3. Results and Discussion

3.1. Rate of aeolian particles collected

The rate of aeolian dust trapped by the collectors at different heights from the tailing ground for the 12 months of sampling is depicted in Fig. 2. There is a great temporal variability in dust deposition rate ranging from the minimum of 2.13 g $month^{-1}$ in June at the height of 80 cm to the maximum of about 670 g $month^{-1}$ in both January and March at the height of 20 cm.

The amount of deposited dust is negatively well correlated with the height of collectors. The total quantity of dust collected at the heights of 20, 50, and 80 cm from the ground is 2891, 784, and 413 g $year^{-1}$, which corresponds to an average of 241, 65, and 34 g $month^{-1}$, respectively. These values are a bit higher than the rates measured by Brotons et al. (2010) who reported an average monthly deposition of 134, 42, and 34 g $month^{-1}$ at the heights of 21, 43, and 63 cm, respectively, for other tailing ponds in the Cartagena-La Union mining district. It has long been proven that the amount of particulate matters in air decreases as the distance from the ground surface increases (e.g. Hojati et al., 2012; Menéndez et al., 2014).

Such factors as the distance from the origin, climatic conditions at the depositional area, surface characteristics and orographic features of the depositional site have been reported to influence the dust deposition rate and its properties (Lawrence and Neff, 2009; Hojati et al., 2012; Menéndez et al., 2014). Contrary to our expectation, the deposition rate of dust in January and March in the study site is much higher than expected indicating that landscape position as physical factor controlled this phenomenon, rather than the climatic conditions. The results of the studies reviewed by Lawrence and Neff (2009) indicated that dust deposition rates are highly variable at the regional and local scales.

3.2. Dust physical and chemical properties

Mean values of pH, electrical conductivity (EC) and carbonates content in the dust collected at different heights, tailing sediments and natural surface soils in the pine tree forest surrounding the tailing pond are given in Fig. 3. All of the dust samples are slightly saline (4.83, 5.18 and 5.34 ds m^{-1} for 20, 50, 80 cm height respectively) much more than the soils of the surrounding area (1.11 ds m^{-1}) and a bit more saline than the tailing sediments (3.58 ds m^{-1}). Forest soils occur on the south and north facing slopes of the hills while the tailing occupies the low lying valley. Therefore, in addition to the soluble salts derived from the long lasting mining activities in the past, tailing sediments still receive the water soluble materials from the surrounding high elevation areas including the natural forest. Although statistically insignificant, the higher height of the collector, the higher the EC values of the dust collected. This seems to be mainly due to the greater association of different salts with finer particles trapped as the height of dust collection increases. Carmona et al. (2009) reported the formation of salt efflorescence on

the tailing pond surface during dry months, which can be easily suspended in the air after wind event due to very small size (1-10 □m) of minerals forming the efflorescence.

The pH value of the tailing sediments is acidic as also previously reported by other researchers (Conesa et al., 2008a; Acosta et al., 2011; González-Corrochano et al., 2014). In contrast, both dust samples from different heights and the soils are almost neutral in pH. All the dust samples as well as the soils and tailings contain a very negligible quantity of carbonates (less than 4 %) which is in agreement with the pH values. This is particularly interesting since the study area is among the driest ones in Europe and the natural soils in other areas of the Murcia Province with no sulphide-rich parent materials contain appreciable amount of carbonates (Gabarrón, 2017).

Particle size distribution of aeolian deposits monthly collected for a period of a whole year is shown in Fig. 4 and compared with average values in soils and tailings in Fig. 5. Aeolian deposits have more sand but less silt and clay content than both the soils and the tailings. Mean sand content of dust samples is more than 85%, while soils and tailings have, in average, less than 70% and less than 80% sand, respectively. In contrast, dust samples contain only about 11% silt in average, whereas the mean values of silt content in soils and tailings are 27% and 20%, respectively. Besides, dust samples have less than 1% clay in average as compared to about 2% clay content of both soils and tailings. Not only do these confirm that dust mostly originates from the local tailing and the surrounding soils, but further indicates that finer particles (less than 20 micrometer) of eroded materials from the tailing and soils are partly transferred by wind to other areas. Since finer particles pose much greater environmental risk to the depositional areas as compared to the coarse ones (USEPA, 2009), care should be taken to stabilize the tailings and the surrounding soils which are extremely contaminated with hazardous materials which would be further explained in the next subsection.

Coarse particles have a tendency to be deposited much closer to their source area due to their greater settling velocities and mass. In contrast, fine-sized particles can be transported by wind thousands of kilometers (Lawrence and Neff, \2009). Therefore, very coarse sized dust deposits produced inside the tailing is most likely of local origin.

As the height of dust collection increases, the amount of coarse particles (sand) decreases and the quantity of fine-textured materials (silt and clay) increases in dust sample (Figs 4 and 5). This is consistent throughout the year in all the 12 months of sampling with no exception. It has

long been proven that due to gravitational force, coarser particles would only be either creeping on the surface, move by saltation, or just enter the atmosphere and move forward by wind very close to the ground (Rehis and Kihl, 1995; Csavina et al., 2012; Hojati et al., 2012).

Monthly changes in the particle size distribution of dust samples (Fig. 4) also clearly indicate that coarse-sized fraction (sand) increases during the colder and moister period (Sep., Oct., Nov., Dec., Jan., and Feb.) as compared to the warmer and drier period of the year (Mar., Apr., May, June, July, and Aug.). The opposite trend occurs for the fine-sized fractions, particularly the silt fraction. This seems to be due to the stronger saltation activities of sand particles in the moister period (Xiaolan and Hongsheng, 2014). Environmentally speaking, this means that local inhabitants are exposed to more hazardous windblown materials having finer texture during spring and summer.

Monthly variability of more environmentally important particulates in dust samples including PM_{10}, $PM_{5.0}$, and $PM_{2.5}$ (USEPA, 2009) and the mean PM_{10} values for soils and tailings have been shown in Fig. 6. Fortunately, dust samples collected contain very negligible quantity of $PM_{2.5}$, less than 5% $PM_{5.0}$, and a maximum of 8% PM_{10}. Both soils and tailings contain a bit more PM_{10} than the dust samples. The mean values of PM_{10} content of soils, tailings, and dust samples are 10.5, 8.0, and 6.0%, respectively. If we assume that dust only comes from either soils of the surrounding hills or the barren tailings, this may indicate that very fine particles are mostly transferred to other areas further away.

3.3. Dust heavy metals geochemistry and Ti/Zr ratio

The mean values of the pseudototal concentration of Cd, Co, Cu, Ni, Mn, Pb, and Zn in dust samples collected at 20, 50, and 80 cm from the tailing ground are given in Table 1. Besides, the means of the same values for soils and tailings are also depicted in Table 1. Based on the data obtained, heavy metals concentration in the dust samples can be grouped into two categories. Category I encompasses the heavy metals with an extremely high pseudototal concentration including Cd, Zn, Mn, and Pb. Whereas, category II is composed of heavy metals having concentration into the range of background concentrations found in the Earth's crust which includes Co, Cu, and Ni. Regardless of the category, the concentration of all the heavy metals in the dust samples is higher than that in tailing and less than that of the soils. This

appears to be a good evidence confirming that the barren surface of tailings and also the surface soils of the surrounding area are the major contributors to the dust collected.

In the first category, the mean concentration of Cd, Mn, Pb, and Zn in dust samples ranges from 13 to 21, 827 to 1503, 1167 to 2730, and 4691 to 6289 mg kg^{-1} with the global mean concentrations of 18, 1254, 1831, and 5747 mg kg^{-1}, respectively. These values well fall between the average values in tailings and soils which are, respectively, 8.3 and 20.2 mg kg^{-1} for Cd, 626 and 3552 mg kg^{-1} for Mn, 1239 and 4393 mg kg^{-1} for Pb, and 2579 and 7590 mg kg^{-1} for Zn. High concentrations of Zn and Pb are expected because of they are the main metals of sulphide minerals (sphalerite and galena, respectively) from Cartagena-la Unión mining district. High Cd concentrations are due to its association to the exploitation of some zinc sulphide minerals such as sphalerite (Alvarez-Ayuso et al., 2013) where Cd replaces Zn due to their analogous nature (Alvarez-Ayuso and Garcia-Sanchez, 2003). Finally, manganese has been found as an impurity of sulphide minerals such as spharelite and pyrite (Graig et al., 1984; Wrigth, 2009). In Cartagena-La Unión mining district, García et al. (2008) found high amount of Mn carbonates associated to sulphide deposits, and other Mn minerals as pyrolusite, romanechite and psilomelane.

The extraordinarily high concentration of the above-mentioned heavy elements is comparable to those already reported for soils and sediments in the highly polluted mining areas of southern Spain (Conesa et al., 2006, 2007, 2008b; Lambrechts et al., 2011; Martínez-Pagán et al., 2011; García-Lorenzo et al., 2012; Acosta et al., 2014; Bes et al., 2014; Rodríguez Martín et al., 2014; Moreno-Barriga, 2017a, 2017b). However, such high levels of metals have rarely been reported in dust samples studied in either agricultural, urban, or industrial areas (Ordonez et al., 2003; Lawrence and Neff, 2009; Al-Khashman, 2014; Norouzi et al., 2017) or even in soil, sediment, and dust samples from mining areas in other parts of the world (Djebbia et al., 2017; Lüa et al., 2018).

In the 2nd category, the mean concentration of Co, Ni, and Cu in dust samples changes from 2.4-4.6, 9.1-15.2, and 42-71 mg kg^{-1} with the overall average of 3.8, 12, and 60 mg kg^{-1}, respectively. These values also well fall between those of average in tailings and soils which are, respectively, 2.1 and 8.3 mg kg^{-1} for Co, 9.5 and 22 mg kg^{-1} for Ni and 55 and 79 mg kg^{-1} for Cu. In contrary to the first category, the concentration of the second category metals in soil, tailing, and dust samples is very much similar to or even less than the values reported for southern Spain

(Conesa et al., 2006, 2007) and other areas in the world (Al-Khashman, 2014; Norouzi et al., 2017). Therefore, it appears that Co, Ni, and Cu in windblown materials are not of environmental concern, whereas tailing ponds should be well protected to minimize the transfer of tremendously enriched particles in Cd, Mn, Pb, and Zn to other areas.

As Fig. 6 shows, no clear and consistent relationship was found between the concentration of the heavy metals in dust samples and the height of dust collection. Neither were there any consistent temporal changes in the concentration of heavy metals in dust collected throughout the year of sampling.

The correlation matrix for the pseudototal concentration of metals in all the dust samples is presented in Table 2. Highly significant correlations among most of the metals further indicate that they likely originate from the same source.

The ratio of Ti/Zr in the dust samples taken from different heights is compared with that of the soils and tailings in Fig. 7. This ratio is almost the same for the dust samples from different heights, which is, in turn, more than the Ti/Zr value for soils and less than the Ti/Zr value in tailings. This supports the results obtained by other measurements (particle size distribution and heavy metals concentration) suggesting the contribution of both soils and tailings to the dust production in the pond studied.

4. Conclusions

The present work indicates that there is a great temporal variability in dust deposition rate in the tailing pond ranging from a very negligible quantity to the maximum of about 670 g $month^{-1}$ in both January and March at the height of 20 cm. As expected, amount of deposited dust decreases as the height of collectors increases. The total quantity of dust collected at the height of 20 cm from the ground is about 3.7 and 7.0 times more than that collected at the heights of 50 and 80 cm, respectively. Due to the location of the tailing pond studied, morphometry and the position of tailing pond control the rate of dust production, rather than the climatic conditions.

Dust deposits produced in the tailing ponds are slightly saline and neutral in pH with a very low quantity of carbonates. Regardless of the height of collection, they are all very coarse in texture with more than 80% sand, 5-18% silt, and less than 2% clay, very much comparable with both the soils and the tailings. This indicates that the dust mostly originates from the local tailing

and the surrounding soils. Deposited dust contains almost no $PM_{2.5}$ but up to 6% and 8% of $PM_{5.0}$ and PM_{10}, while tailings and surrounding soils have in average 8% and 10.5% PM_{10}. All these indicate that fine sized particles, which have much greater environmental risk are mostly transferred by wind to areas further away.

Regarding the concentration of heavy metals in deposited dust, two groups of metals were identified. The concentration of the first group, which includes Cd, Zn, Mn, and Pb, is extremely high, whereas the second group of heavy metals including Co, Cu, and Ni has normal concentration with respect to background levels in the Earth's crust. Besides, the concentration of all the heavy metals in the dust samples falls between that of tailings and soils. This also clearly indicates that the barren surface of tailings and also the surface soils of the surrounding area are the two major sources contributing to the dust production in the ponds. This is further corroborated by the Ti/Zr ratio in the deposited dust. Highly significant correlations among the concentrations of most of the heavy metals further indicate that they likely originate from the same source(s) discussed above.

In order to reduce the spread of particles eroded from the abandon tailing ponds, some remediation actions must be carried out, such as phytoremediation and promoting the development of vegetation in the tailing ponds.

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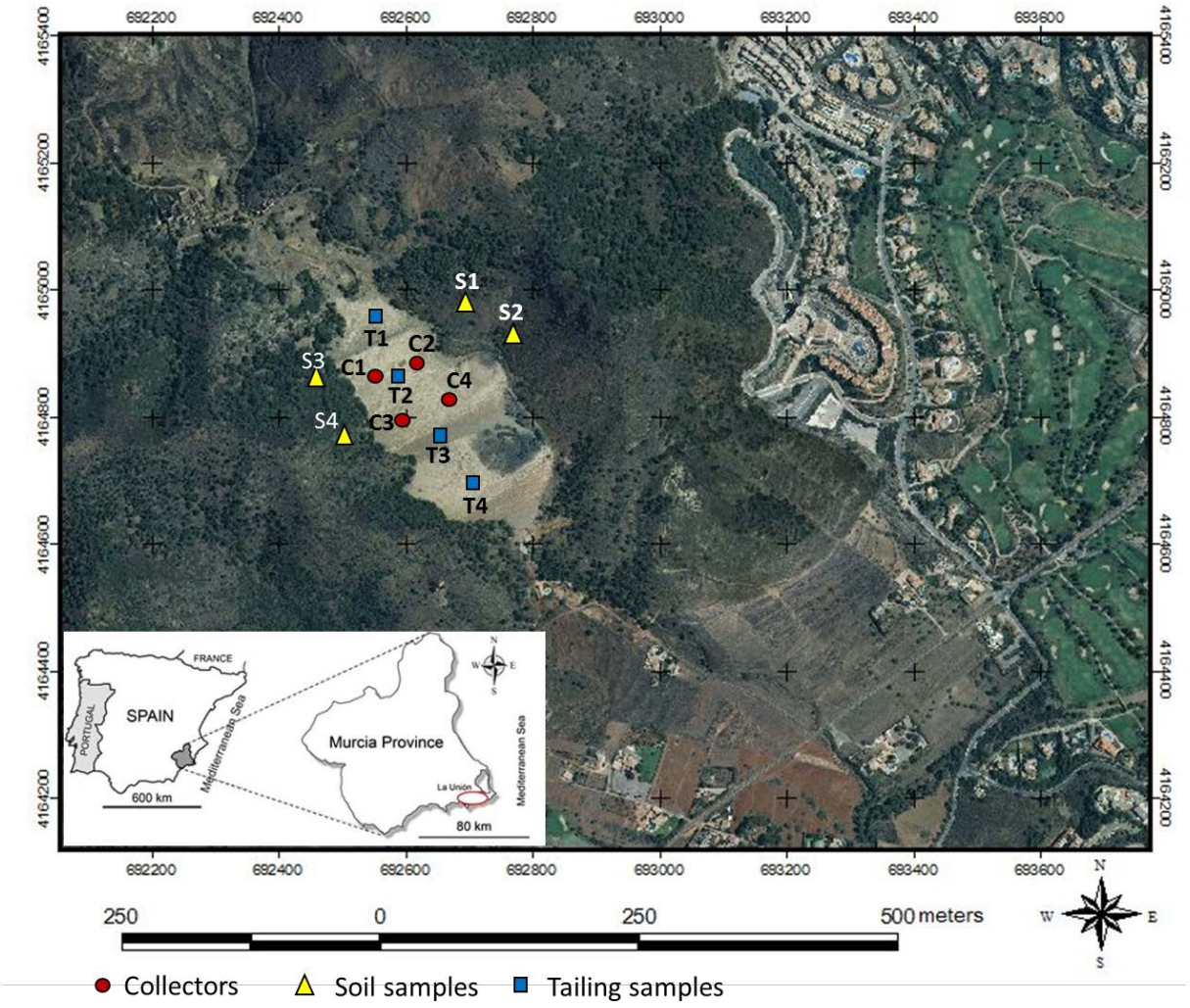

Fig. 1. The location of El Lirio tailing pond in southeastern Spain where dust collectors were installed (C_1, C_2, C_3, and C_4) as well as the sites of soil (S_1, S_2, S_3, and S_4) and tailing (T_1, T_2, T_3, and T_4) sampling. An image of a dust collector is also shown in the bottom left side.

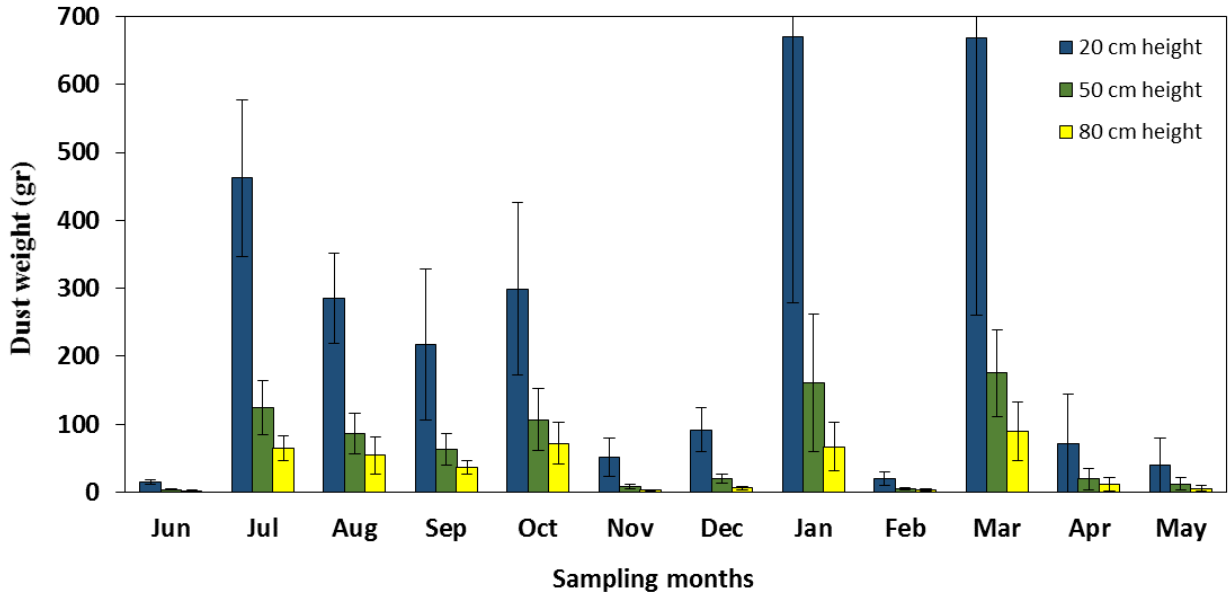

Fig. 2. The rate of windblown deposits collected at three different heights from the tailing pond surface during the 12-month sampling period. Bars indicate standard deviations, n=4.

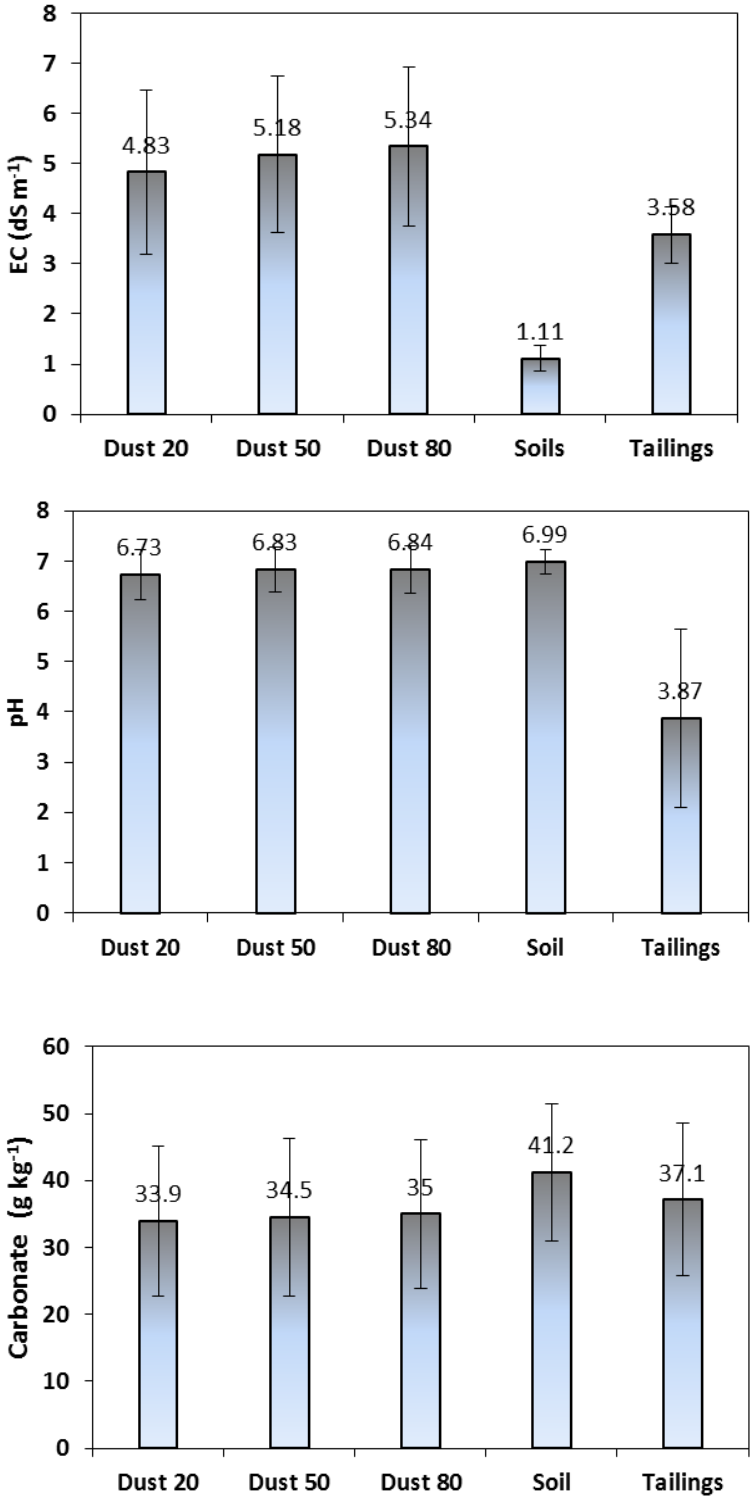

Fig. 3. Mean values of EC, pH, and carbonate content in dust samples collected at 20 (Dust 20), 50 (Dust 50) and 80 (Dust 80) cm from the tailing surface as compared to those of soils and tailings. Bars indicate standard deviations, n=48 for dusts and 4 for soils and tailings.

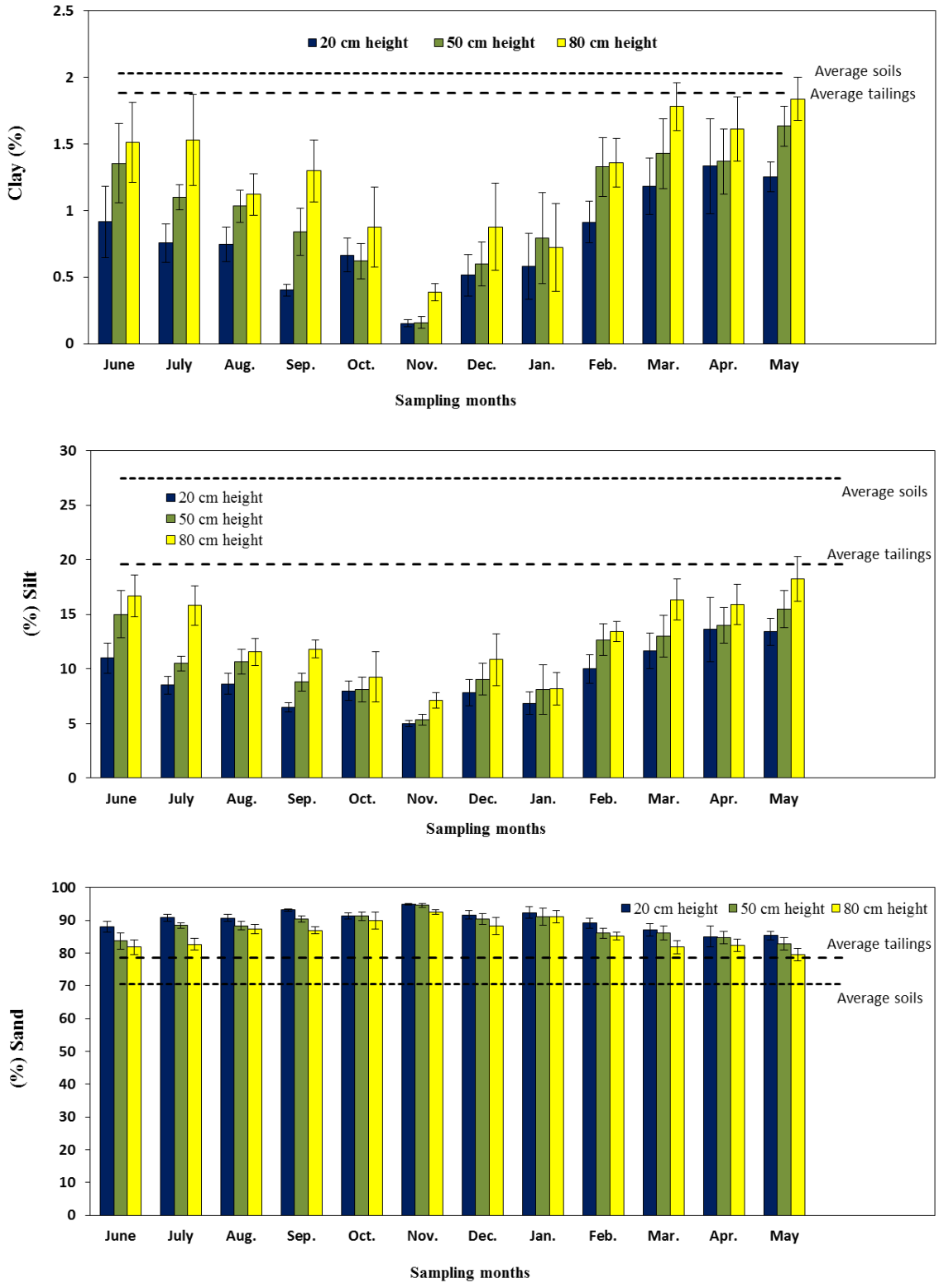

Fig. 4. Comparison of mean sand, silt, and clay contents of dust samples collected at 3 different heights from the tailing ground surface with reference to the average values in soils and tailings.

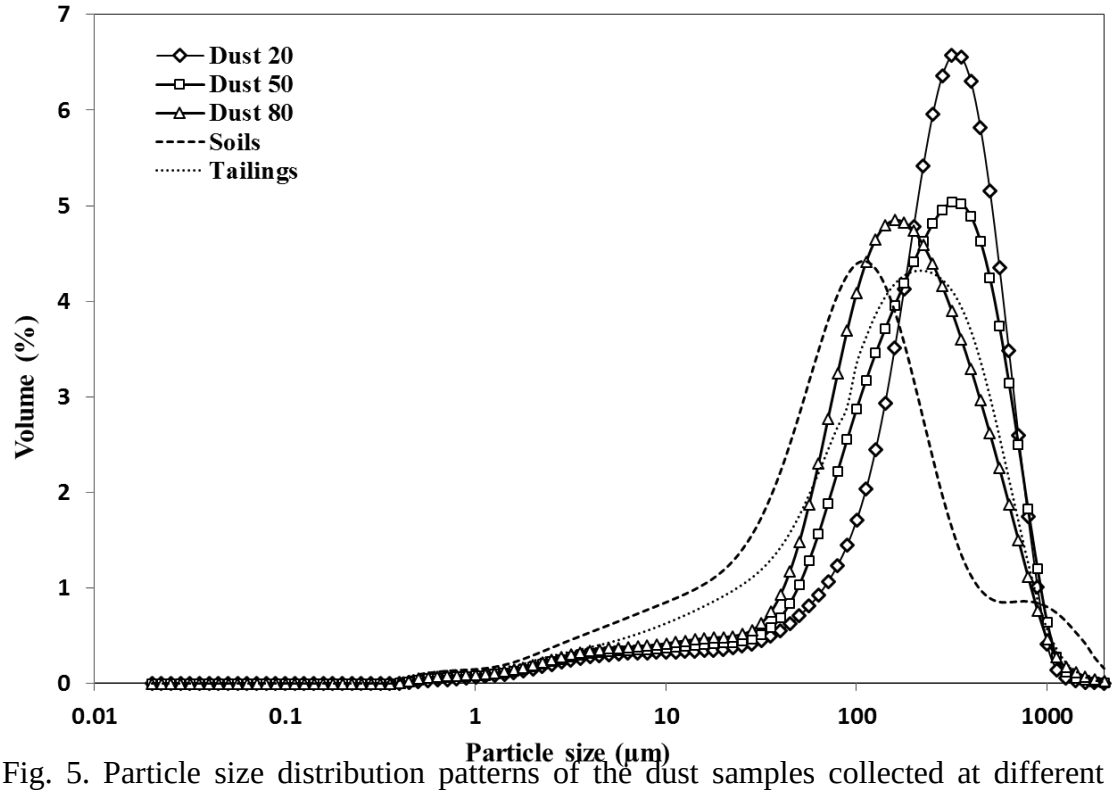

Fig. 5. Particle size distribution patterns of the dust samples collected at different heights as compared to those of soils and tailings. Dust 20, Dust 50, and Dust 80 denote the dust samples collected at the height of 20, 50, and 80 cm, respectively. For each case, the average of all the samples is shown.

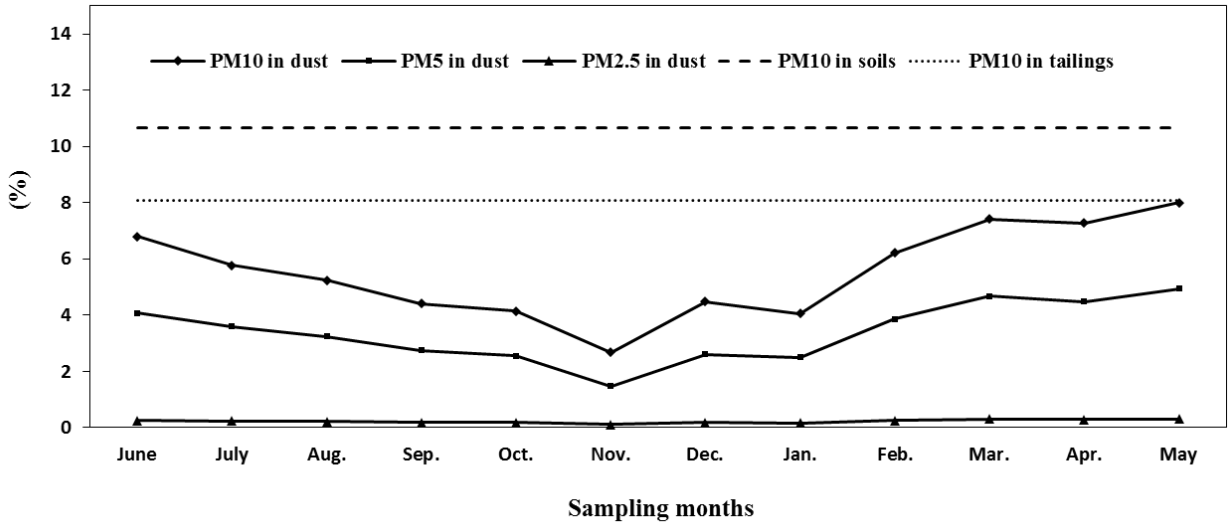

Fig. 6 Monthly changes in the PM_{10}, PM_5 and $PM_{2.5}$ values of dust and the level of PM_{10} in soils and tailings.

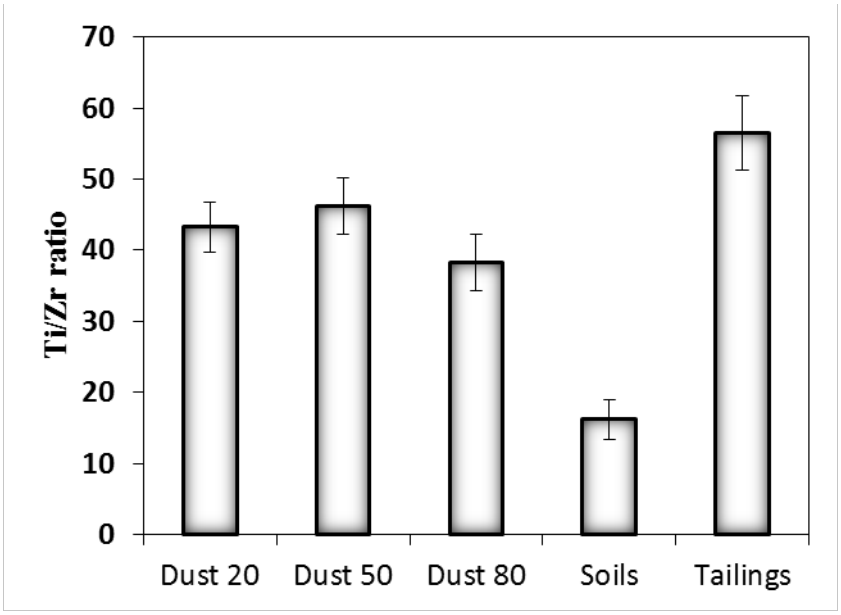

Fig. 7 Mean value of Ti/Zr ratios in the dust samples from different heights compared to that in soils and tailings.

Table 1. Metals concentration of dust collected at three different heights for all sampling periods (mean from four collectors)

Metal	Height	June	July	August	September	October	November	Dececember	January	February	March	April	May	Soil	Tailing
	20 cm	2263	2221	2119	1339	2205	1264	1167	1924	1340	1581	1456	2407		
Pb (mg kg^{-1})	**50 cm**	1791	2293	2384	1520	1993	1301	1196	2264	1411	1810	1588	2455	4393	1239
	80 cm	1404	2730	2209	1641	2337	1410	1350	2315	1478	1538	1551	2644		
	20 cm	6109	6099	6215	4913	5516	5518	5989	6072	5949	6389	5196	5825		
Zn (mg kg^{-1})	**50 cm**	5725	6078	5917	4815	5326	5628	5989	6174	5698	5808	5348	6098	7590	2578
	80 cm	5689	5819	5558	4691	5446	5706	6289	6270	5495	6171	5377	5974		
	20 cm	20	20	21	15	15	16	20	19	20	23	18	20		
Cd (mg kg^{-1})	**50 cm**	18	18	19	14	14	16	19	19	19	18	18	19	20	8
	80 cm	18	18	17	13	15	16	20	19	18	20	19	21		
	20 cm	15	15	13	9	13	9	9	14	10	10	10	14		
Ni (mg kg^{-1})	**50 cm**	12	15	14	9	13	9	10	15	9	11	11	15	22	9
	80 cm	10	15	13	10	14	9	10	15	10	10	10	15		
	20 cm	71	71	70	54	69	55	56	49	57	47	53	68		
Cu (mg kg^{-1})	**50 cm**	61	71	71	54	66	56	55	68	54	45	54	70	79	55
	80 cm	53	70	66	56	67	57	57	70	54	42	53	51		
	20 cm	1439	1417	1344	1069	1372	1163	1209	1086	1218	1018	1309	1434		
Mn (mg kg^{-1})	**50 cm**	1225	1385	1330	1124	1303	1233	1214	1503	1200	906	1398	1449	3524	626
	80 cm	1227	1370	1256	1151	1343	1184	1288	1499	1204	827	1349	1110		
	20 cm	4.6	4.5	3.9	3.0	3.8	3.3	3.6	3.2	3.5	3.2	3.9	4.4		
Co (mg kg^{-1})	**50 cm**	4.2	4.4	4.0	3.2	3.8	3.4	3.7	4.5	3.6	2.9	4.4	4.6	8.3	2.1
	80 cm	4.2	4.6	3.7	3.4	4.0	3.4	3.8	4.6	3.6	2.4	4.0	3.8		

Table 2. The Pearson's correlation coefficients among the total concentrations of heavy metals (n=139).

Co	0.922**					
Ni	0.753**	0.762**				
Cu	0.345*	0.309*	0.710**			
Zn	0.628**	0.560**	0.387*	0.377**		
Cd	0.761**	0.705**	0.390*	0.148^{ns}	0.802**	
Pb	0.273*	0.364*	0.696**	0.734**	-0.054^{ns}	-0.046^{ns}
	Mn	**Co**	**Ni**	**Cu**	**Zn**	**Cd**

* Statistically significant correlation at 0.05 levels; ** statistically significant correlation at 0.01 levels; ns: non-significant correlation.