

Original Paper

# Mineralogy and Geochemistry of Rural Road Dust and Nearby Mine Tailings: A Case of Ignored Pollution Hazard from an Abandoned Mining Site in Semi-arid Zone

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Abandoned mine tailings are considered as one of the main sources of potentially toxic elements. Because of the lack of supervision, particularly from small-scale or artisanal mining, abandoned tailings have become part of the natural landscape, especially in rural areas from developing countries. Abandoned mine tailings represent a latent problem in terms of the possible affectations to human health and the environment. An example of this is the small-sized ( $\sim 200 \times \sim 300$  m) abandoned mine tailings located  $\sim 500$  m south of San Felipe de Jesús town, Sonora, in northwestern Mexico. The mineralogy determined in mine tailings samples consists of divalent hydrated metal sulfates (rozenite, starkeyite, kieserite, szomolnokite and epsomite), trivalent hydrated metal sulfates (coquimbite) and divalent-trivalent hydrated sulfates (copiapite), which are highly soluble efflorescent minerals associated with acid mine drainage. Rozenite was detected in road dust samples, evidencing that dust is dispersed and transported from abandoned residues. In order to assess the possible impact of the tailings (un-oxidized, oxidized, efflorescent minerals), concentrations of potentially toxic elements (total and soluble fractions) in samples from mine tailings, unpaved road soils and road dust from San Felipe de Jesús were determined. Average concentrations (ppm) of potential toxic elements in mine tailings samples ranged from 16,756–1306 (As), 665–98 (Cd), 5691–338 (Cu), 14,162–832 (Pb), 492–82 (Sb), 176,219–8285 (Zn). Enrichment factors determined in mine tailings, agricultural soils and road dust exhibit similar patterns, differing only in level of enrichment, which also confirms the dispersion of potentially toxic elements toward surroundings. Contamination Index (CI) and Hazard Average Quotient (HAQ) were calculated in mine tailings to assess potential contamination associated with potentially toxic elements dispersed by aeolian and/or hydric processes, respectively. The CI values suggest that mine tailing materials have a high potential for polluting soils and sediments. Semi-arid conditions of the region favor the suspension and transport of contaminants, potentially affecting surrounding agricultural fields and population. The HAQ values from efflorescence minerals and mine tailings indicate that potential of toxicity is very high, and might affect the quality of water (groundwater and surficial) in the region. CI and HAQ can provide a good estimation of pollution hazards associated with the abandoned mine tailings in the San Felipe de Jesús area.

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## INTRODUCTION

Mining is an essential activity to obtain chemical elements indispensable to cover a wide range of needs, from basic housing construction to the development of state-of-the-art technology. However, it is common knowledge that mining has produced large quantities of hazardous wastes that may contain high concentrations of potential toxic elements (PTE), e.g., Cu, Pb, Cd, Zn, Hg, Ag, Sn, As, Sb, among others (Rodríguez et al. 2009; Rashed 2010; Anawar 2015; Martín-Crespo et al. 2018).

Mine tailings, which are defined as a composite pile of a solid–liquid mixture, include fine-sand to silt-size particles of minerals from ore, gangue, rocks and sediments, which are separated from main ore and deposited in impoundments (Hudson-Edwards et al. 2011; Quispe et al. 2013). These are commonly found around or near mining sites, and often contain high concentrations of PTE. These pollutants can be dispersed into the environment, either by aeolian and/or hydric processes into terrestrial and aquatic systems (Adriano 2001; Mendez and Maier 2008; Panagos et al. 2013; Anawar 2015). The high environmental impact and health risk that mining residues represent is a fact that has been extensively recognized (e.g., Plumlee and Morman 2011; García-Giménez and Jiménez-Ballesta 2017).

Regulations and acts regarding the confinement and maintenance of mine tailings are very strict during mining operation to prevent failures (e.g., Kossoff et al. 2014), and to avoid the dispersal of PTE. However, a problem arises with mine tailings

generated during historical mining, because it was most likely performed without environmental regulations and lax mining laws, and under less efficient metallurgical techniques. For instance, the extractive technology was not completely effective in the past, or in some cases, artisan techniques were used, leading to high concentrations of PTE in the resultant tailings (Hudson-Edwards et al. 2011).

Mine wastes generated by past mining operations, whether with or without owner or legal guardian, represent an environmental problem because of the lack of remediation during or after closure of the activity. These wastes are commonly called abandoned mine tailings, and are in fact considered important sources of PTE, impacting significantly the environment, since they may remain un-vegetated for tens to hundred years and disperse over tens of hectares (Mendez and Maier 2008, and references therein). Therefore, polluted abandoned tailings not only affect at the local or regional scale, but also can act as source of pollutants in national or even cross-border contexts. Moreover, due to the lack of supervision and attention over the years, abandoned mine tailings are subject to deterioration and erosion, and eventually become part of the natural landscape. This is especially true in rural areas in developing countries, where they represent an ignored but latent problem in terms of the possible impacts to human health and the environment (Obiora et al. 2016).

Climatic conditions are determining factors regarding the dispersion of pollutants into the environment. On the one hand, an important process for the release of PTE in humid environments is through the generation of acid mine drainage (AMD). This consists of the oxidation of sulfide minerals exposed to weathering processes, resulting in the generation of acidic sulfate waters with high levels of dissolved metals (Navarro et al. 2008; Sánchez-Bisquert et al. 2017). The development of AMD commonly occurs in humid environments during most of the year (Akcil and Koldas 2006); however, in arid and semi-arid regions, it tends to occur through intermittent episodes, mainly controlled by intense precipitation during short periods of time, which can potentially contribute soluble

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metals to hydrological systems (Romero et al. 2008; Santisteban et al. 2015). Moreover, intense precipitation episodes, followed by rapid evaporation and low infiltration, favor the formation of efflorescent salts on mine tailings surfaces (Mendez and Maier 2008). These salts can easily dissolve in water, which also implies the mobilization of contaminants into hydrological systems (Gieré et al. 2003; Hammarstrom et al. 2005). On the other hand, dust plays an important role in terms of rapidity, long range and effectiveness of transporting contaminants in arid and semi-arid regions (Csavina et al. 2012). Therefore, aeolian transport is the main dispersion process in these environments. Furthermore, in the case of mine tailings, the dispersal of particles is efficient due to their nature (e.g., fine-grained, semi-consolidated, un-vegetated). Moreover, temperature contrasts will favor decrepitation of efflorescent salts formed during dry seasons, decreasing cohesion, which raises the susceptibility to be dispersed. Abandoned mine tailings therefore pose a considerable risk to human health and environment, particularly in arid and semi-arid regions where aeolian erosion is highly effective (Csavina et al. 2012).

In general, concentration of metals in dust from large urban population centers, as well as the environmental and health consequences, is an issue that has been extensively studied (Huang et al. 2018; Najmeddin et al. 2018; Ramirez et al. 2018). In the case of rural areas, the knowledge is limited compared to large cities, most likely due to the scarce anthropogenic contaminant sources in rural areas (e.g., vehicular traffic, industrial zones, construction, municipal waste incineration). However, small settlements in rural zones are not excluded from environmental impacts, especially when located around abandoned mining sites (Marrugo-Negrete et al. 2014; Sánchez-Bisquert et al. 2017).

Developing countries, as in the case of Mexico, usually lack a database with information about the location, mapping and geochemical features of abandoned mine tailings, as well as their proximity to human settlements and hydrological networks. In the semi-arid zones of northwestern Mexico, abandoned tailings have been ignored by authorities for decades. Particularly in Sonora, only a few academic studies are available dealing with the dispersion, geochemical features, risk characterization and phytostabilization potential of the abandoned mine tailings from Nacoziari, Sonora (e.g., Romero et al. 2008; Meza-Figueroa et al. 2009; de la O-Villanueva

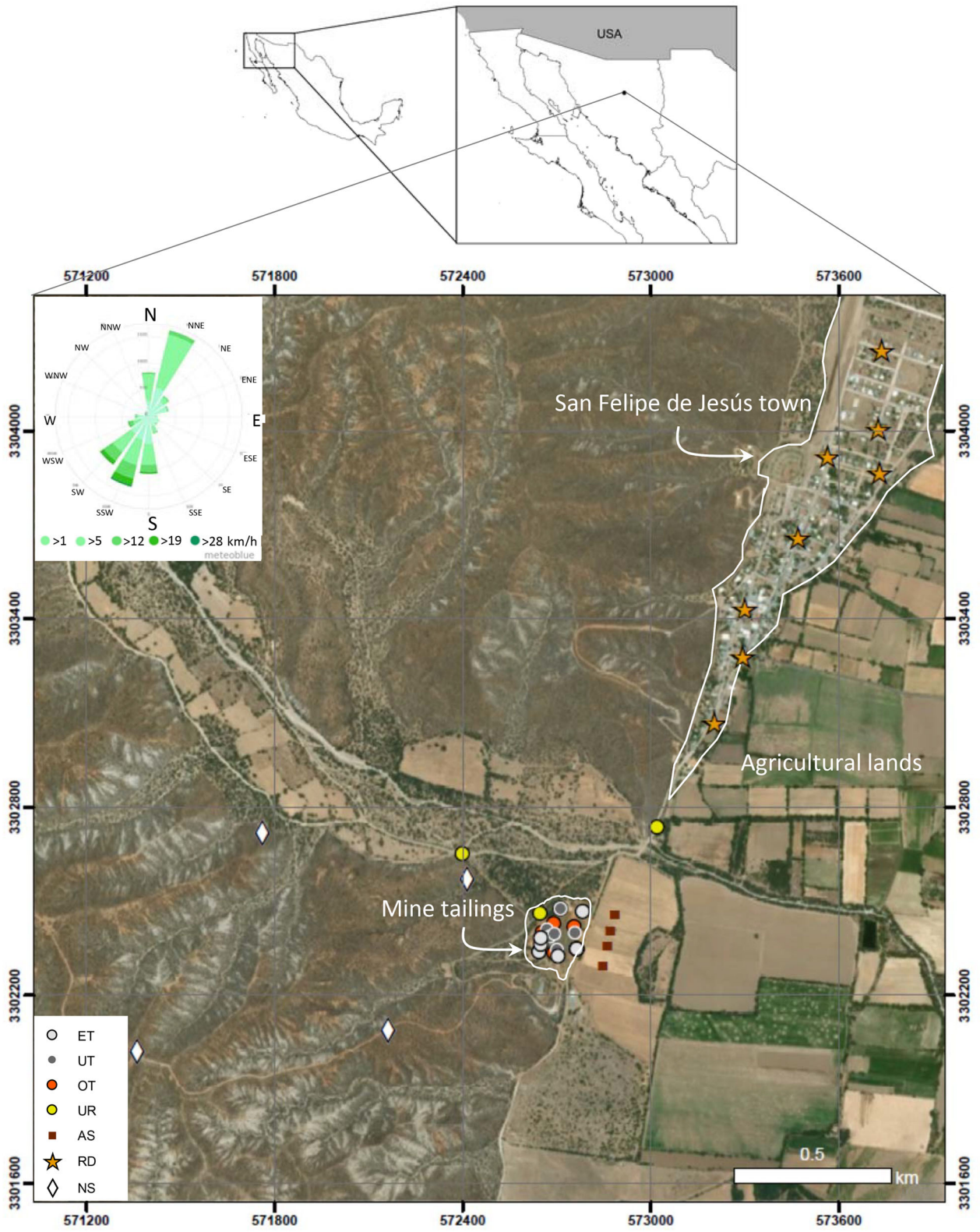
et al. 2013; Santos et al. 2017). Thus, our knowledge about the PTE content in rural dust near abandoned tailings associated with small-scale mining in Sonora is limited. The aim of the present study is to examine the PTE concentrations (total and soluble) in road dust from San Felipe de Jesús in central Sonora, as well as in a small-scale abandoned mine tailings, located 0.5 km away from town. The idea is to perform mineralogical and geochemical characterizations of rural dust and mine residue to assess dispersion, potential pollution and toxicity of an abandoned mine tailings deposit.

## MATERIALS AND METHODS

### Study Site

The study area is located within the Sonoran Desert, in the central portion of the state of Sonora, in northwestern Mexico, and comprises the rural area of the San Felipe de Jesús town and the small-scale abandoned mine tailings, situated ~ 500 m apart (Fig. 1). Climate in the region is classified as semi-arid. Meteorological data for the period between 1981 and 2010 indicate that the region has average annual precipitation of 524 mm, concentrated mainly during July–August, and has annual average temperature of 19.6 °C. The last reported population is 396 inhabitants (INEGI 2010); the main economic activities in the region are agriculture and cattle raising, although San Felipe de Jesús was founded in 1657 for mining purposes. The study area is located along the Sonora River, next to agricultural fields (Fig. 1).

The dimensions of the abandoned tailings are 140 × ~ 160 m, with a maximum and minimum heights of ~ 5 and ~ 2 m, respectively, and comprise a surface of ~ 16,300 m<sup>2</sup> (Espinoza-Madero 2012). The accumulation of the mine tailings of San Felipe de Jesús started in 1920s (Technical Report 2012), developing gully erosion structures. The tailings are characterized by the absence of vegetation, and are reddish (oxidized) in the external zones and grayish (un-oxidized) in the internal zones (Fig. 2a and b); on tailing surfaces, either on un-oxidized and oxidized zones, it is common the accumulation of efflorescent minerals as white-colored crusts (Fig. 2c and d). The mined metals in the area included lead and zinc, and minor gold and silver (Technical Report 2012).



◀ **Fig. 1.** Location map showing the abandoned mine tailings and San Felipe de Jesús town, in central Sonora. ET: efflorescence crusts developed on un-oxidized and oxidized tailings; UT: un-oxidized tailings; OT: oxidized tailings; UR: unpaved roads surrounding mining residue; AS: agricultural soils; RD: road dust from San Felipe de Jesús town; NS: natural soils.

### Sampling and Analytical Methods

A total of 27 surficial (0–5 cm depth) samples were collected on the abandoned mine tailings, of which seven correspond to efflorescent salts; mine tailings samples were taken using a stainless steel shovel, whereas efflorescent salts were carefully taken using a stainless steel spatula. Four surficial (0–5 cm depth) samples of agricultural soils, located next to the tailings deposit, were collected using a stainless steel shovel. Three surficial (< 1 cm depth) unpaved road samples surrounding the abandoned residue, some of them, presumably affected by the tailings. Moreover, eight road dust samples were evenly collected from San Felipe de Jesús town (Fig. 1); all road samples were taken using a brush and a dustpan. Finally, five soil samples were collected on different types of soils of the studied area, in order to determine the geochemical background of the region; with the purpose to avoid any anthropogenic input, these samples were collected between 10 and 20 cm depth with a stainless steel shovel. All samples were placed in hermetic bags and stored for further preparation.

If needed, the samples were dried at 30 °C in an oven. Subsequently, the samples were weighted and sieved to obtain the fine-grained fraction from mesh 635 (20 µm) for further mineralogical and geochemical studies. In order to determine the pH and electric conductivity (E.C.) values, a proportion of 1:5 of total sample and water was shaken according to method 9045C (USEPA 1995), using a symphony™ Benchtop Meters model B30PCI.

The mineral species of mine tailings were determined by X-ray diffraction (XRD), using a Shimadzu XRD-6000 diffractometer with an accelerating voltage of 40 V and a filament current of 35 mA at the National Laboratory of Geochemistry and Mineralogy (LANGEM). The diffractometer is equipped with a vertical goniometer and can scan for  $\theta$  or  $2\theta$ . The measurement was made by an angular interval of  $2\theta$ , with a scan angle between 4° and 70°, at a speed of 2° per minute (Bish and Post 1989; Buhrke et al. 1998).

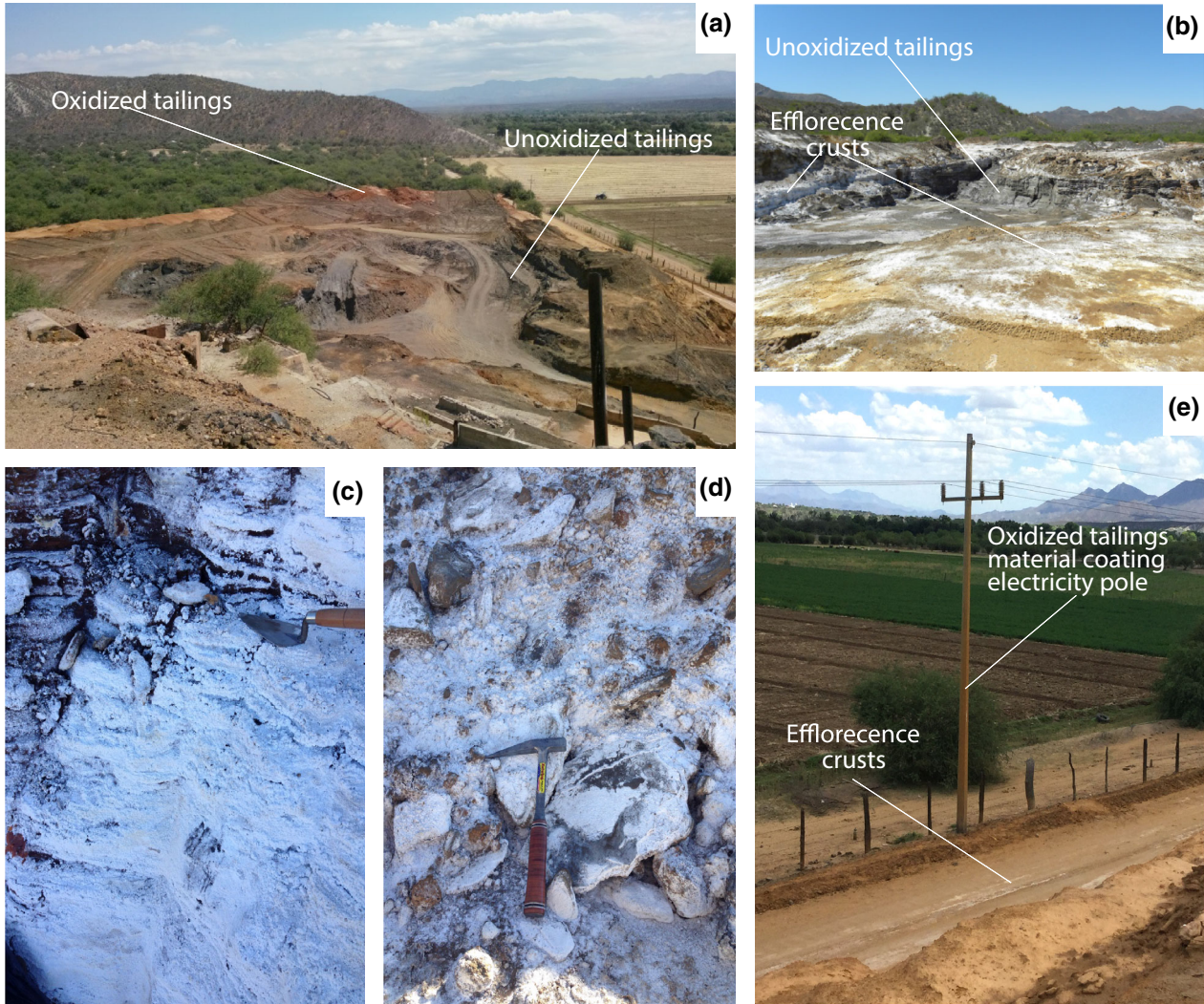
Elemental concentrations of As, Cd, Cu, Pb, Sb, Zn, Fe, Mn and Ti were determined using a portable X-ray fluorescence (pXRF) analyzer (Niton XLt3) from Thermo Scientific at LANGEM, according to method 6200 from USEPA (2007). Accuracy and precision were determined by using reference standards materials for soil (Montana 2711a, Montana 2710a, and TILL-4) and mine tailings (RST-4) samples. Concentrations were determined by triplicate measurements; in general, results were within acceptable ranges ( $\pm 20\%$ ). The concentrations of water-soluble elements (As, Cd, Cu, Fe, Mn, Pb, Zn and Sb) were determined by the De-ionized Water Leach (ME-MS03) method of ALS laboratories in Canada, which consists of leaching 1 g of sample in 25 ml of de-ionized water, and digested in a water bath at 60 °C for two hours. The final solution is separated from the solids by centrifuging and decanting the supernatant, and then elemental concentrations were measured by ICP-MS; accuracy and precision ( $\pm 10\%$ ) were determined by using the reference material OREAS-45d.

### Statistical Analysis

A *T* test was performed to compare elemental concentrations between samples of un-oxidized and oxidized tailings, and between samples of efflorescent salts formed in both types of tailings. Additionally, a *T* test was carried out to compare elemental concentrations in samples of natural soil with those from unpaved road, agricultural soil and road dust samples. Principal component analysis (PCA) was also performed to recognize associations of variables (mineralogy, total elemental and soluble concentrations) between different road dust samples and mine tailings. Statistical analyses were performed by using the XLSTAT (2014) software for Windows (version 8.1, Addinsoft).

### Environmental Indices

Enrichment factor (EF) has been widely used to distinguish chemical elements between natural geochemical background and anthropogenic sources (Zhang and Liu 2002). The mathematical expression to calculate EF is defined as (Feng et al. 2004; Ghrefat et al. 2011):



**Fig. 2.** Panoramic views showing un-oxidized and oxidized (a) portions of mine tailings located in San Felipe de Jesús area. Perspective of un-oxidized and oxidized tailings developing efflorescence crusts (b). Detailed view of efflorescence crusts formed on un-oxidized (c) and oxidized tailings (d). Unpaved road next to mine residue, containing oxidized mine tailings material and efflorescence crusts, and electricity pole coated with oxidized tailings material (e), demonstrating mobility of material from abandoned waste.

$$EF = \frac{[El]_{\text{sample}}/[X]_{\text{sample}}}{[El]_{\text{background}}/[X]_{\text{background}}}$$

where “El” is the concentration of studied element, whereas “X” is the concentration of a reference element. Sample refers to the studied material (e.g., soils, sediments, dust), whereas background refers to local geochemical background values that have been used as reference material to approach more realistic values (e.g., Loska et al. 2003; Abraham and Parker 2008; Kaushik et al. 2009; Wu et al. 2014; Zhang et al. 2014). Birth (2003) categorized EF as: minor enrichment (1.5 < EF < 3); moderate

enrichment (3 ≤ EF < 5); moderately severe enrichment (5 ≤ EF < 10); severe enrichment (10 ≤ EF < 25); very severe enrichment (25 ≤ EF < 50) and extremely severe enrichment (50 < EF).

The index of contamination (IC) has been proposed by Alberruche del Campo et al. (2014) to systematic ranking of abandoned mining waste facilities. It consists of the normalization of toxic elements to background levels, considering only the elemental ratios greater than one. The IC is expressed as:

$$IC = \frac{1}{n} \times \sum_{i=1}^n \frac{X_i}{BL_x}$$

where  $X_i$  is concentration of the  $i$ th element in the studied sample,  $BL_x$  represents regional background level of element  $X$  and  $n$  is number of ratios greater than one. According to this index, the greater the value of IC, the greater is the likelihood of pollutants emission to surrounding soils and sediments (Arranz-González et al. 2016). Additionally, these authors also proposed a hazard average quotient (HAQ), which is an index to evaluate the likelihood of water to be polluted with soluble toxic elements from abandoned mining waste. The HAQ is expressed as:

$$HAQ = \frac{1}{n} \times \sum_{i=1}^n \frac{X_{Li}}{WQL_x}$$

where “ $X_{Li}$ ” is the concentration of element  $i$ th in a water leachate (according to the EN-12457-2 test) from the studied sample, “ $WQL_x$ ” represents the water quality standard for element “ $X$ ” and “ $n$ ” is the number of ratios that yield values greater than one (Alberruche del Campo et al. 2014; Arranz-González et al. 2016).

## RESULTS AND DISCUSSION

### Mineralogical Species

The mineralogical species detected by XRD in samples from mine tailings, efflorescent crusts and road dust, are shown in Table 1. The minerals found in samples from un-oxidized and oxidized tailings consist of silicates, sulfates, carbonates, sulfides and oxides. Among the silicates found, in order from the most to the least abundant, quartz, microcline, kaolinite, muscovite, richterite, enstatite and spessartine were detected. The presence of these minerals and amphibole, pyroxene, garnet and magnesioferrite indicates a typical gangue mineral association observed in skarn mineralization, as observed previously in the San Felipe de Jesús area (e.g., Mead et al. 1988), while the ore minerals were found to consist of pyrite, sphalerite and arsenopyrite. Carbonates (calcite and dolomite) have been also found in the mining tailings, which are more abundant in the sulfur-rich ones; the presence of carbonates is important due to the control over the

pH, since in case of acid generation, they are important consumers of protons, favoring pH stabilization between 6 and 8 (Sherlock et al. 1995; Iii and Trahan 1999). Silicates, sulfides and carbonates found in tailings can be considered as “primary” (Lindsay et al. 2015), considering that they were already in the hosting rocks or were formed during alteration and mineralization processes.

Regarding secondary minerals, that is, minerals formed because of oxidation processes of mine tailings (Lindsay et al. 2015), the most representative are sulfates and oxyhydroxides and hydroxy sulfates. Gypsum was commonly found in the majority of studied samples from sulfide- and oxidized tailings, indicating that there is a constant oxidation and dissolution of sulfides. The precipitation of gypsum limits the mobility of PTE (Lin et al. 2013). Anglesite is another insoluble sulfate detected, resulting from oxidation of galena, characterized by limiting mobility of lead. Hydroxylsulfates of the alunite group such as jarosite were detected, indicating the possible presence of argentojarosite, hydroniojarosite and natrojarosite. Jarosite is formed under oxidizing and low pH conditions, favoring, on the one hand, the oxidation of sulfides that contribute to Fe(III) and  $SO_4$ , while dissolution of silicates contributes to  $K^+$  and  $Na^+$  (Lindsay et al. 2015); this is an important mineral phase that controls the mobility of PTE (e.g., Zn, Cu, Ni, As). Although jarosite is metastable and is transformed to Fe(III) oxyhydroxides (e.g., goethite) or hematite, the resulting mineral phases continue fixing PTE (Romero et al. 2007). If acid conditions intensify ( $pH < 3.5$ ), goethite becomes unstable releasing the PTE, which may explain the low abundance of goethite in San Felipe oxidized tailings (average pH of 3.7). Another insoluble hydroxylsulfate of the alunite group corresponds probably to beudantite, which is very important since it controls the mobility of As and Pb in mining residues (Romero et al. 2007; Loredó-Portales et al. 2017). This mineral species was only found in un-oxidized mine tailings samples (Table 1). Jarosite and beudantite are in a continuous solid solution, and more detailed analyses could identify more accurately the presence of mineral phases of this mineral group. Finally, another group of sulfates detected is simple divalent hydrated metal sulfates from the rozenite (rozenite and starkeyite), kieserite (kieserite, szolmonokita) and epsomite groups (Table 1, Fig. 3). These are highly soluble efflorescent minerals, which are formed by evaporation of fluids resulting from oxidation and

**Table 1.** Mineral species determined by X-ray diffraction in samples of abandoned tailings and road dust from the San Felipe Jesús area, in central Sonora

Mineral	Formula	Mineral group	UT (%)	OT (%)	EUT (%)	EOT (%)	RD
Calcite	CaCO <sub>3</sub>	Carbonate	15–3	+	4–0	–	+
Dolomite	CaMg (CO <sub>3</sub> ) <sub>2</sub>	Carbonate	2–0	–	–	–	–
Ankerite	CaFe <sup>2+</sup> (CO <sub>3</sub> ) <sub>2</sub>	Carbonate	–	–	–	–	+
Magnesioferrite	(MgFe) <sub>2</sub> O <sub>4</sub>	Oxide	2–0	–	–	–	–
Cristobalite	SiO <sub>2</sub>	Oxide	+	–	–	–	–
Goethite	FeO(OH)	Oxide-hydroxide	4–0	–	–	–	–
Quartz	SiO <sub>2</sub>	Silicate	42–8	47–33	35–8	26–16	+
Kaolinite	Al <sub>2</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub>	Silicate	38–3	–	+	–	–
Illite	(K,H <sub>3</sub> O)(Al,Mg,Fe) <sub>2</sub> (SiAl) <sub>4</sub> O <sub>10</sub>	Silicate	–	11–0	–	–	–
K-feldspar	K(SiAl) <sub>4</sub> O <sub>8</sub>	Silicate	48–12	–	–	–	+
Albite	NaAlSi <sub>3</sub> O <sub>8</sub>	Silicate	–	–	–	–	+
Richterite	Na (CaNa)Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub>	Silicate	9–0	–	–	–	–
Diopside	MgCaSi <sub>2</sub> O <sub>6</sub>	Silicate	20–0	–	–	–	–
Enstatite	MgSiO <sub>3</sub>	Silicate	1–0	–	–	–	+
Muscovite	KAl <sub>2</sub> (AlSi <sub>3</sub> O <sub>10</sub> )(F,OH) <sub>2</sub>	Silicate	24–0	–	11–0	+	–
Microcline	KAlSi <sub>3</sub> O <sub>8</sub>	Silicate	+	–	–	–	–
Spessartine	(Mn <sup>2+</sup> ) <sub>3</sub> Al <sub>2</sub> (SiO <sub>4</sub> ) <sub>3</sub>	Silicate	–	+	–	–	–
Mordenite	(Ca,N <sub>a</sub> ,K <sub>2</sub> )Al <sub>2</sub> Si <sub>10</sub> O <sub>24</sub> ·7H <sub>2</sub> O	Silicate	–	–	–	73–6	–
Gypsum	CaSO <sub>4</sub> ·2H <sub>2</sub> O	Sulfate	50–6	60–26	39–5	15–5	–
Jarosite	KFe <sup>3+</sup> (OH) <sub>6</sub> (SO <sub>4</sub> ) <sub>2</sub>	Sulfate	21–5	17–4	27–0	14–0	–
Hydroniumjarosite	(H <sub>3</sub> O)Fe <sub>3</sub> (SO <sub>4</sub> ) <sub>2</sub> (OH) <sub>6</sub>	Sulfate	–	–	+	–	–
Kieserite	MgSO <sub>4</sub> ·H <sub>2</sub> O	Sulfate	50–0	10–0	–	50–27	–
Epsomite	MgSO <sub>4</sub> ·7H <sub>2</sub> O	Sulfate	23–0	–	6–0	–	–
Szomolnokite	FeSO <sub>4</sub> ·H <sub>2</sub> O	Sulfate	–	–	–	64–21	–
Rozenite	FeSO <sub>4</sub> ·4H <sub>2</sub> O	Sulfate	–	–	–	14–0	+
Coquimbite	Fe <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ·9H <sub>2</sub> O	Sulfate	15–7	–	–	–	–
Aluminocoquimbite	FeAl(SO <sub>4</sub> ) <sub>3</sub> ·9H <sub>2</sub> O	Sulfate	–	–	+	–	–
Copiapite	Fe <sup>2+</sup> Fe <sub>4</sub> <sup>3+</sup> (SO <sub>4</sub> ) <sub>6</sub> (OH) <sub>2</sub> ·20H <sub>2</sub> O	Sulfate	–	–	26–0	–	–
Starkeyite	MgSO <sub>4</sub> ·4(H <sub>2</sub> O)	Sulfate	63–0	–	51–0	46–0	–
Beudantite	PbFe <sub>3</sub> (OH) <sub>6</sub> SO <sub>4</sub> AsO <sub>4</sub>	Sulfate	12–0	–	–	–	–
Kieserite	MgSO <sub>4</sub> ·H <sub>2</sub> O	Sulfate	–	–	22–0	–	–
Anglesite	PbSO <sub>4</sub>	Sulfate	5–0	7–0	–	–	–
Argentojarosite	AgFe <sub>3</sub> <sup>3+</sup> (SO <sub>4</sub> ) <sub>2</sub> (OH) <sub>6</sub>	Sulfate	+	–	+	–	–
Hydroniumjarosite	(H <sub>3</sub> O)Fe <sub>3</sub> (SO <sub>4</sub> ) <sub>2</sub> (OH) <sub>6</sub>	Sulfate	+	+	–	–	–
Hydroniumjarosite	(H <sub>3</sub> O)Fe <sub>3</sub> (SO <sub>4</sub> ) <sub>2</sub> (OH) <sub>6</sub>	Sulfate	–	+	–	–	–
Pyrite	FeS <sub>2</sub>	Sulfide	26–6	–	19–3	2–0	–
Sphalerite	ZnS	Sulfide	8–2	3–0	–	8–6	–
Arsenopyrite	FeAsS	Sulfide	17–0	–	–	–	–

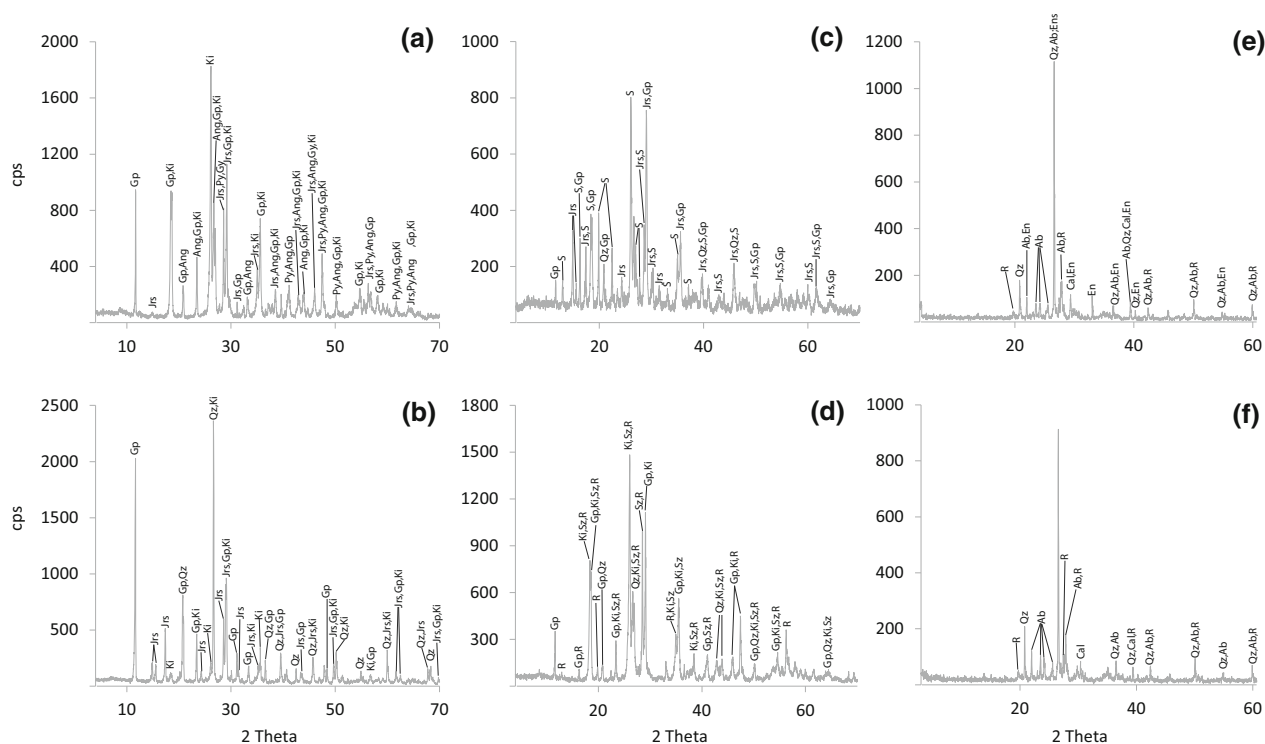
Abundances (%) were calculated using Reference Intensity Ratio (RIR) method, therefore abundance percentage is semi-quantitative. Mineral species detected in samples (+), but unable to calculate abundance: mineral species not detected (–). *UT* un-oxidized tailings, *OT* oxidized tailings, *EUT* efflorescence from un-oxidized tailings, *EOT* efflorescence from oxidized tailings, *RD*: road dust

acid mine drainage (Lindsay et al. 2015). Although these mineral species are mostly found in efflorescent crusts, they were also detected in un-oxidized tailings (Fig. 6a), and to a lesser extent, in oxidized mine tailings samples (Fig. 3b). Other highly soluble species detected by XRD are simple hydrous sulfates with trivalent metal cations (coquimbite) and mixed divalent-trivalent hydrous metal sulfates (copiapite). Efflorescent minerals are potentially responsible for the release and dispersion of PTE to environmental media, through aeolian and hydric erosion processes. Mobility of PTE can be associ-

ated with rainy season, enhancing dispersion in solution due to the high solubility of efflorescence minerals.

Regarding the mineral associations detected by XRD in road dust samples from San Felipe de Jesús town, the most representative ones are silicates followed by carbonates (Table 1). This is reasonable considering the surrounding lithology. In addition, rozenite was found, which is an efflorescent mineral also detected in the nearby mine tailings (Fig. 3e and f); this dispersion of mineral particles is supported by predominant wind directions as evidenced by





**Fig. 3.** Representative XRD spectra of (a) un-oxidized tailings, (b) oxidized tailings, (c) efflorescence crusts from un-oxidized tailings, (d) efflorescence crusts from oxidized tailings, (e–f) road dust samples from San Felipe de Jesús town. Gp: gypsum; Jrs: jarosite; Ki: kieserite; Ki: anglesite; Ang: quartz; Qz; starkeyite; Sz; rozenite; R; starkeyite; S; albite; Ab; calcite; Cal; enstatite: En.

wind rose in Figure 1. The presence of rozenite in road dust is an important finding since it can be considered as mineral indicator that evidences dispersion processes of particulate material originated in the tailings. In fact, dispersion phenomenon in arid and semi-arid regions is favored by wind erosion through dust, which is an effective transporter agent of pollutants from local to global scales (Csavina et al. 2012). Efflorescent minerals are more susceptible to wind erosion due to their low cohesion between particles (Sánchez-Bisquert et al. 2017), especially during dry seasons. Therefore, dispersion of efflorescent minerals could potentially impact surrounding areas. The fact that rural dust is impacted indicates that population might be exposed to PTE contained in soluble minerals, implying a potential health risk by inhalation or ingestion.

### Elemental Concentrations and Enrichment

Descriptive statistical data of Pb, As, Zn, Cu, Sb, Cd, Fe, Mn and Ti concentrations determined in samples from abandoned mine tailings, road dust,

agricultural and non-impacted soils are shown in Table 2.

No significant differences were found when in elemental concentrations (As, Cu, Pb, Sb, Fe and Ti) between un-oxidized and oxidized mine tailings (Table 2); the only elements found with significant differences were Cd, Zn and Mn. Higher concentrations of Cd and Zn characterize un-oxidized tailings, which are expected because these types of tailings represent the original composition of the waste. Conversely, higher Mn concentrations were recorded in oxidized tailings, which can be explained by their higher mobility during acidic conditions and further fixation. Regarding efflorescent salts developed on un-oxidized and oxidized mine tailings, no significant differences were found in concentrations of As, Cd, Cu, Zn and Fe. However, significant differences were found in concentrations of Pb, Sb and Mn. High concentrations of Pb and Sb characterize efflorescent salts from un-oxidized tailings, which can be explained by the high concentrations of these elements in the un-oxidized tailings, whereas high Mn concentrations are found in efflorescent salts from oxidized tailings (Table 2).

**Table 2.** Elemental concentrations ( $\text{mg kg}^{-1}$ ) of samples from abandoned mine tailings, soils and road dust from the San Felipe de Jesús area, in central Sonora

	UT ( $n = 14$ )	OT ( $n = 6$ )	EUT ( $n = 3$ )	EOT ( $n = 4$ )	UR ( $n = 3$ )	AS ( $n = 4$ )	RD ( $n = 8$ )	NS ( $n = 5$ )
pH	6.5	3.7	3.7	4.9	7.1	7.3	8.9	7.7
E.C.	298.8	415.7	68.0	83.3	2281.4	51.0	120.7	40.5
As	25,177.4–342.2	17,265.2–221.4	26,787.3–7392.9	2554.9–56.7	146.5–33.1	91.1–26.8	258.5–83.7	21.7–10.7
Ave $\pm$ SD	10,098.5 $\pm$ 8990.2	6213.2 $\pm$ 6873.0	16,756.4 $\pm$ 9714.4	1305.8 $\pm$ 1766.5	89.8 $\pm$ 80.2	47.2 $\pm$ 29.9	181.3 $\pm$ 89.2	15.0 $\pm$ 4.8
( $P > t$ )	0.395	0.395	0.125	0.125	0.413	0.296	0.084	–
Cd	904.7–138.2	163.4–15.8	1027.1–369.4	829.7–327.4	nd	nd	nd	nd
Ave	327.3 $\pm$ 210.6	67.7 $\pm$ 66.7	664.7 $\pm$ 334.0	487.9 $\pm$ 230.3	nd	nd	nd	nd
( $P > t$ )	0.001*	0.001*	0.441	0.441	–	–	–	–
Cu	939.2–176.3	501.2–83.8	1719.3–484.7	10,967.5–382.7	41.5–22.2	94.6–65.1	83.9–59.3	40.8–19.6
Ave	491.9 $\pm$ 220.8	338.3 $\pm$ 162.4	1052.9 $\pm$ 623.1	5691.4 $\pm$ 5292.5	31.8 $\pm$ 13.6	73.2 $\pm$ 14.3	70.1 $\pm$ 15.3	28.3 $\pm$ 8.3
( $P > t$ )	0.179	0.179	0.267	0.267	0.679	0.001*	0.001*	–
Pb	26,944.4–6857.5	12,687.2–7524.6	9698.9–7645.1	1449–206.7	227.9–225.8	536.5–320.1	376.9–72.5	70.7–45.0
Ave	14,161.9 $\pm$ 6839.2	10,464.5 $\pm$ 2061.4	8672.0 $\pm$ 1452.2	831.9 $\pm$ 621.5	226.9 $\pm$ 1.5	382.9 $\pm$ 102.8	274.1 $\pm$ 113.4	62.0 $\pm$ 12.0
( $P > t$ )	0.111	0.111	0.003*	0.003*	0.0001*	0.0008*	0.0002*	–
Sb	955.5–83.2	770.6–56.0	616.1–360.3	164.3–28.0	213.9–21.6	nd	24.8–15.8	nd
Ave	481.0 $\pm$ 329.4	415.8 $\pm$ 350.7	491.9 $\pm$ 128.1	81.9 $\pm$ 72.5	117.8 $\pm$ 135.9	nd	19.0 $\pm$ 3.6	nd
( $P > t$ )	0.699	0.699	0.008*	0.008*	–	–	–	–
Zn	176,589.6–22,802.7	16,167.6–1854.2	195,607.7–132,210.6	185,665.3–170,684.8	469.8–435.1	626.2–531.1	956.7–444.4	292.5–120.1
Ave	60,709.3 $\pm$ 41,828.3	8285.4 $\pm$ 6585.0	163,909.1 $\pm$ 44,828.5	176,218.7 $\pm$ 8221.0	452.5 $\pm$ 24.5	576.5 $\pm$ 47.7	705.7 $\pm$ 171.9	193.0 $\pm$ 66.1
( $P > t$ )	0.001*	0.001*	0.649	0.649	0.004*	0.0001*	0.0001*	–
Fe	363,510.7–129,349.6	307,864.9–126,774.2	437,486.7–176,677.9	161,951.2–7068.1	20,541.7–18,115.1	25,246.4–23,112.5	35,306.0–26,093.6	27,731.4–21,160.3
Ave	212,377.1	307,864.8	288,915.3	61,942.1	19,328.4	24,047.9	30,388.9	24,537.4
( $P > t$ )	0.794	0.794	0.070	0.070	0.057	0.740	0.008*	–
Mn	57,851.0–6502.2	21,780.6–11,036.4	58,236.2–25,200.7	237,150.6–68,464.1	1113.4–852.4	1755.9–1355.8	1382–969.1	1153.4–604.2
Ave	29,519.6 $\pm$ 17,622.8	16,255.1 $\pm$ 4652.6	41,562.1 $\pm$ 16,519.9	117,418.4 $\pm$ 44,140.2	982.9 $\pm$ 184.6	1489.9 $\pm$ 184.8	1165.8 $\pm$ 137.7	885.2 $\pm$ 248.8
( $P > t$ )	0.025*	0.025*	0.049*	0.049*	0.644	0.005*	0.0301*	–
Ti	942.1–356.4	1309.0–716.8	731.3–251.4	–	2795.3–2091.4	3414.7–3196.1	4626.9–3499.8	4232.9–3986.4
Ave	728.8 $\pm$ 171.4	989.6 $\pm$ 233.9	487.0 $\pm$ 240.1	nd	2340.0 $\pm$ 394.8	3312.0 $\pm$ 92.5	3864.9 $\pm$ 368.5	4143.1 $\pm$ 116.8
( $P > t$ )	0.537	0.537	–	–	0.0001*	0.0001*	0.184	–

\*Significant differences at 95% confidence

UT un-oxidized tailings, OT oxidized tailings, EUT effluence from un-oxidized tailings, EOT effluence from oxidized tailings, UR unpaved road, AS agricultural soils, RD road dust, NS natural soils. Electric conductivity ( $\mu\text{S/cm}$ ). Detection limits (ppm) are As (3), Cd (7), Cu (12), Pb (5), Sb (12), Zn (6), Fe (35), Mn (55), Ti (10). nd: below detection limit.  $T$  test ( $P > t$ )

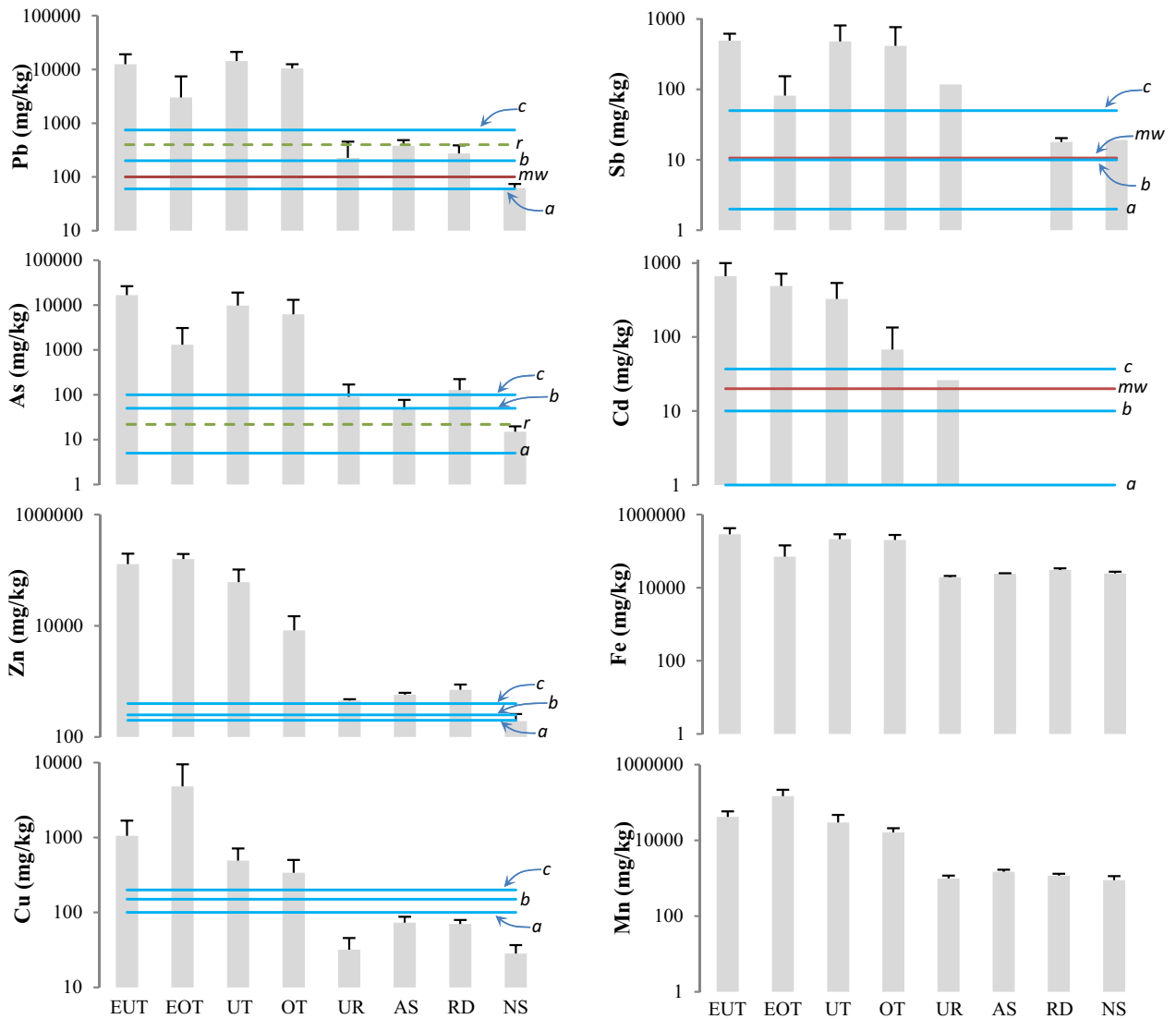
Regarding the comparison of elemental concentrations between natural and unpaved road soils, no significant differences were detected in As, Cu, Fe and Mn; however, significant differences were detected for Pb, Zn and Ti (Table 2). Concentrations of Cd and Sb in natural soil were below detection limit, whereas Cd was also below detection level in unpaved road soils samples, and Sb showed a concentration of 117.8 ppm in road soil. Regarding the comparison between natural soil and road dust, no significant differences were found in concentrations of As and Ti; however, significant differences were detected between Cu, Pb, Zn, Fe and Mn (Table 2), which might be explained by anthropogenic input. Finally, statistical analyses suggest significant differences in concentrations of As and Fe between natural and agricultural soils, while differences were found for Cu, Pb, Zn Mn and Ti.

The results indicate that PTE contents in the San Felipe tailings are elevated in As, Cu, Pb, Sb and Zn. The average concentrations ( $\text{mg kg}^{-1}$ ) followed the decreasing order  $\text{Fe} > \text{Zn} > \text{Mn} > \text{Pb} > \text{As} > \text{Cu} > \text{Sb} > \text{Cd}$  for un-oxidized tailings,  $\text{Fe} > \text{Mn} > \text{Pb} > \text{Zn} > \text{As} > \text{Sb} > \text{Cu} > \text{Cd}$  oxidized tailings,  $\text{Fe} > \text{Zn} > \text{Mn} > \text{As} > \text{Pb} > \text{Cu} > \text{Cd} > \text{Sb}$  for efflorescence formed on un-oxidized tailings and  $\text{Zn} > \text{Mn} > \text{Fe} > \text{Cu} > \text{Pb} > \text{As} > \text{Cd} > \text{Sb}$  for efflorescence formed on oxidized tailings. The average concentrations found in agricultural and natural soils followed the decreasing order  $\text{Fe} > \text{Mn} > \text{Zn} > \text{Pb} > \text{Cu} > \text{As}$ , whereas unpaved roads surrounding mine tailings showed  $\text{Fe} > \text{Mn} > \text{Zn} > \text{Pb} > \text{Sb} > \text{As} > \text{Cu}$ . Finally, the average elemental concentrations obtained in road dust samples from San Felipe de Jesús town generated the decreasing order  $\text{Fe} > \text{Mn} > \text{Zn} > \text{Pb} > \text{As} > \text{Cu} > \text{Sb}$ .

On the one hand, elemental concentrations obtained from mine tailings samples were compared with the maximum allowed concentrations of toxic elements in mining waste according to the Secretariat of Environment and Natural Resources of Mexico (SEMARNAT, Spanish acronym), which establishes the procedures of mining residue management in Mexico (SEMARNAT 2009). Although these concentrations are considered during project, construction, operation and closing stages of mine residues, these values were used as a comparative approach given the lack of information regarding concentration limits for abandoned mine tailings. Average concentrations of Pb-As-Sb-Cd in re-suspendible fractions of abandoned mine tailings con-

siderably exceed the allowed limits for active mining waste (Fig. 4). On the other hand, obtained values were compared with those for agricultural, residential and commercial soils according to SEMARNAT (2004), which establishes concentrations of contaminated soils in Mexico (Fig. 4), where average As concentrations exceed the values for agricultural soils and road dust.

With the purpose of comparing obtained concentrations data in soils to a global level, the Finnish standard values have been chosen since they represent good approximation average concentrations in Europe, India and agricultural soils by United Nations Environment Programme (Tóth et al. 2016). The Finnish standard system (MEF 2007) set: (a) the threshold value for exploratory assessment; (b) the guideline value (higher and lower) where contamination might present ecological or health risks (Tóth et al. 2016). In soils studied in the San Felipe de Jesús area, the threshold values are exceeded by Zn, As, Pb, Sb and Cd (Fig. 4). Lower guideline value is exceeded by average concentrations of Zn, As, Pb and Sb, whereas average concentrations of Zn and As exceeded the higher guideline values (Fig. 4). The data obtained in this investigation suggest that the San Felipe de Jesús region should be environmentally assessed because elemental concentrations (Pb, As, Zn, Sb) may indicate an ecological risk in the area, and particularly health risk in the case of Pb and Sb. In addition, Table 3 shows PTE concentrations at regional and global scale, in order to compare obtained results with other localities contaminated by abandoned mining sites. In general, agricultural soils and road dust from San Felipe de Jesús showed higher concentrations of Zn and Pb, and As to a lesser extent. Table 3 highlights the problem of high PTE concentrations in agricultural soils from regions with similar socioeconomic conditions in South America, where sources of pollutants are related to mining activity affecting at local or regional scale (e.g., De Gregori et al. 2003; Veado et al. 2006; Garrido et al. 2012). A feature observed in these agricultural soils is the high variability of PTE associations and concentrations, which may be the resulting combination of factors such as the type of ore deposit, mining, metallurgical processes, climate (Romero et al. 2008; Kossoff et al. 2014), as well as the lack of preventive and/or remediation policies regarding abandoned mining waste. This highlights the need for constraining sources of PTE that can be potentially dispersed and cause environmental and health problems in developing



**Fig. 4.** Average concentrations in mine tailings, agricultural, natural soils and road dust samples from San Felipe de Jesús town. EUT and EOT: efflorescence crusts developed on un-oxidized and oxidized tailings, respectively; UT: un-oxidized tailings; OT: oxidized tailings; UR: unpaved roads surrounding mining residue; AS: agricultural soils; RD: road dust from San Felipe de Jesús town; NS: natural soils. Blue lines represent (a) threshold values, (b) lower guideline value, (c) higher guideline value, according to the elemental values established for soils (MEF 2007); (r) elemental values established for agricultural, residential and commercial soils according to the Secretariat of Environment and Natural Resources of Mexico (SEMARNAT 2004); (mw) maximum allowed limit according to the procedures of mining residue management of Mexico (SEMARNAT 2009).

countries, particularly if PTE are incorporated in the food chain (Bech et al. 2012).

Establishing globally accepted elemental values to evaluate whether soils have concentrations above the established in environmental assessments is challenging, mainly because of the variety of geological features in each region. This problem is even more complex when it comes to abandoned mining waste due to the lack of threshold values. Therefore,

with the purpose of determining the enrichment of PTE in mine tailings samples, in addition to agricultural and road dust samples, enrichment factors (EF) were applied, using Ti as a normalizer because of the low coefficient of variation ( $CV < 3\%$ ) in natural soils samples, from which it is assumed as the local geochemical background. As expected, EF values (As, Zn, Pb, Cu, Mn) of mine tailings exceed the very severe enrichment limit, followed by un-

**Table 3.** Comparative data of PTE concentrations (mg kg<sup>-1</sup>) from different countries including data from this investigation

	As	Cd	Cu	Pb	Zn	References
<i>Agricultural soils</i>						
Guangdong Province, China	5.3	0.1	12.2	51.9	56.3	Cai et al. (2019)
Wuxi, China	14.3	0.1	40.4	46.7	112.9	Yan-Feng et al. (2007)
Nkpuma Ekwoku, Nigeria	1.8	8.6	8.2	109.6	210.0	Chukwu and Oji (2018)
Argolida basin, Greece	7.0	0.5	74.7	19.7	74.9	Kelepertzis, (2014)
Murcia, Spain	–	0.2	11.0	48.9	18.4	Acosta et al. (2011)
Alicante, Spain	–	0.3	22.5	22.8	52.8	Micó et al. (2007)
Isfahan, Iran	–	0.4	35.4	34.6	111.5	Esmaeili et al. (2014)
Tarapacá, Chile <sup>b</sup>	34.0	–	31.0	–	–	De Gregori et al. (2003)
Antofagasta, Chile <sup>b</sup>	202.0	–	35.0	–	–	De Gregori et al. (2003)
Valparaíso, Chile <sup>b</sup>	52.0	–	530.0	–	–	De Gregori et al. (2003)
O'Higgins, Chile <sup>b</sup>	–	0.3	450.0	48.0	145.0	Badilla-Ohlbaum et al. (2001)
Fuente de Oro, Colombia	–	nd	17.6	16.7	58.6	Mahecha-Pulido et al. (2015)
Sinú basin, Colombia <sup>b</sup>	–	0.04	1149.0	0.07	1365.0	Marrugo-Negrete et al. (2017)
Río Moche middle basin, Peru <sup>b</sup>	0.02	0.02	1.1	0.4	0.3	Huaranga Moreno et al. (2012)
Gerais, Brazil <sup>b</sup>	650.0	–	60.0	–	–	Veado et al. (2006)
San Felipe de Jesús área, Mexico	47.2	–	73.2	382.9	576.5	This study
<i>Road dust</i>						
San Francisco de Assis, Portugal	810.2	5.4	338.9	43.9	464.4	Candeias et al. (2014)
Nacozari de García, Mexico	15.0	–	278.0	51.7	184.7	Meza-Figueroa et al. (2009)
Bayan Obo, China	12.0	2.2	36.4	183.9	299.4	Li et al. (2015)
Huainan, China	6.6	0.3	36.3	42.2	–	Tang et al. (2017)
Murcia, Spain	–	0.4	116.0	123.0	176.0	Acosta et al. (2011)
Suzhou, China	–	–	27.5	45.2	225.3	Lin et al. (2017)
San Felipe de Jesús área, Mexico	181.3	–	70.1	274.1	705.7	This study
<i>Abandoned mine tailings</i>						
Wadi El-Allaqi, Egypt	8.0	0.3	56.5	14.0	54.5	Rashed (2010)
Sardinia, Italy	–	62.0	128.5	4700.0	12,400.0	Concas et al. (2006)
Nacozari de García, Mexico	29.3	<4.0	400.5	39.0	78.8	Meza-Figueroa et al. (2009)
Gabellino, Italy	138.0	18.0	299.0	2450.0	6080.0	Benvenuti et al. (1997)
Kettara, Morocco	–	65.0	1950.0	77.5	272.5	Hakkou et al. (2008)
San Felipe de Jesús área, Mexico	6213.2	67.7	338.3	10,464.5	8285.4	This study <sup>a</sup>

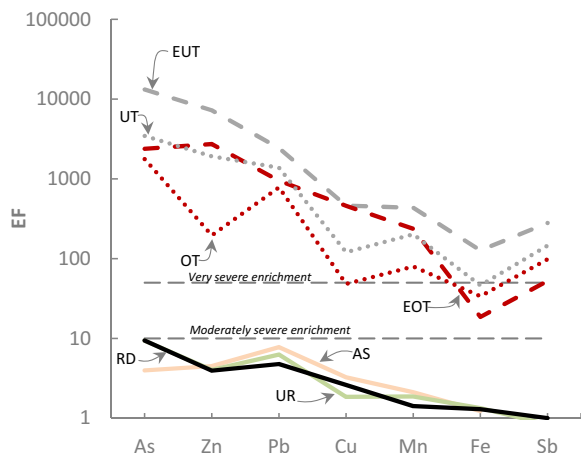
nd below detection limit

<sup>a</sup>Un-oxidized mine tailings

<sup>b</sup>Agricultural soils affected by mining activity

paved road soil surrounding the tailings with enrichment between very-severe-to-moderately-severe enrichment. The lowest enrichment factors recorded belong to agricultural soils and street dust samples (Fig. 5). Although EF has been widely used to determine the degree of anthropogenic contribution in soils and sediments (Zhang and Shan 2008; Çevik et al. 2009; Molamohyeddin et al. 2017; Pan et al. 2018), investigations do not recommend this approach (Reimann and de Caritat 2000, 2005). In this investigation, instead of determining anthropogenic contributions with EF, normalization factor is assumed here to construct multi-elemental patterns to identify geochemical similarities in environmental assessments. Multi-elemental normalized patterns have been widely used in geosciences to

trace sources and processes (e.g., Rollinson 2014), as well as in environmental sciences (e.g., Moreno-Rodríguez et al. 2015). The EF spectrum of oxidized tailings and their respective efflorescent crusts show the mobility from source (mine tailings) to efflorescent salts, indicating in general, a higher elemental concentration in efflorescence phase, mainly in Zn and Cu, and Mn to a lesser extent (Fig. 5). The EF spectra of un-oxidized tailings and their respective efflorescent minerals are similar, except that efflorescent crusts are slightly enriched in PTE. The spectra that characterize unpaved road, agricultural soils and road dust are similar, indicating less enrichment than nearby abandoned tailings (Fig. 5). However, despite the differences in enrichment, the spectrum of unpaved road, agricultural soils, road



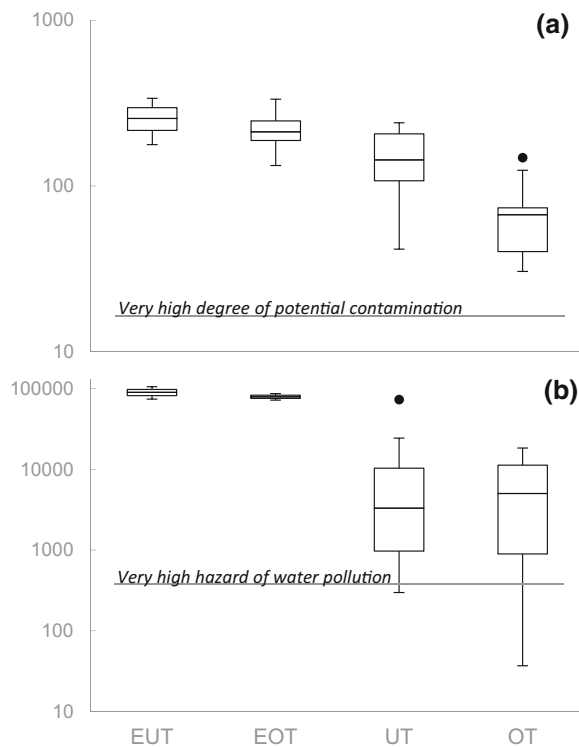
**Fig. 5.** Enrichment factors patterns of efflorescence crusts (EUT and EOT) formed on un-oxidized and oxidized tailings, respectively, un-oxidized tailings (UT), oxidized tailings (OT), unpaved roads surrounding mining residue (UR), agricultural soils (AS), road dust (RD) from San Felipe de Jesús town.

dust is similar to oxidized mine tailings, highlighting the impact of small-scale historical mining in the rural area of San Felipe de Jesús, which has been ignored for decades.

### Potential Pollution and Toxicity

The approach used to evaluate possible contamination of PTE, considering the abandoned mine tailings as the main source, consists of a methodology that classifies abandoned mining residues proposed by Arranz-González et al. (2016). According to the values obtained, average contamination index (CI) obtained, from highest to lowest, is 256.8 (efflorescence from un-oxidized tailings), 222.7 (efflorescence from oxidized tailings), 149.9 (un-oxidized tailings), 70.0 (oxidized tailings). According to the toxicity factor proposed by Alberruche del Campo et al. (2014), the CI obtained in studied tailings represent a high potential of pollution (Fig. 6a) if dispersing toward surrounding soils and sediments.

Considering that mine tailings can interact with environmental humidity or rainwater if infiltrates through it, oxidation processes enhance liberation of soluble PTE, potentially impacting superficial and underground water quality. With the purpose to estimate that impact, a comparative exercise was carried out to determine, in case mining tailings interact with water, which elements can be solubilized and potentially reach bodies of water. The high



**Fig. 6.** Box plot diagram showing variation of (a) index of contamination (IC) and (b) hazard average quotient (HAQ) of efflorescence crusts from un-oxidized tailings (EUT), efflorescence crusts from oxidized tailings (EOT), un-oxidized tailings (ST), oxidized tailings (OT) of abandoned mine tailings from San Felipe de Jesús town. Degree of potential contamination for soils and sediments and hazard of water pollution inferred from Alberruche del Campo et al. (2014).

elemental concentrations recorded in studied samples (Table 4) exceed internationally established values for some PTE to determine water quality (WHO 1993; Secretaría de Salud 1994; European Council 1998; USEPA 2002, 2012). Soluble concentrations of As, Cd, Cu, Pb, Sb and Zn were used to determine the hazard average quotient (HAQ) and toxicity factor proposed by Alberruche del Campo et al. (2014). The results indicate an average HAQ of 89,680 (efflorescence from oxidized tailings), 79,180 (efflorescence from un-oxidized tailings), 17,655 (un-oxidized tailings) and 7107 (oxidized tailings). According to the high HAQ values determined in tailings and efflorescence crusts generated on surfaces of both types of tailings, the potential of contaminating water bodies (e.g., surficial, underground water by infiltration) is very high (Fig. 6b). It is worth noting that obtained CI and HAQ values of abandoned mine tailings from San Felipe de Jesús

**Table 4.** Concentrations of soluble elements (mg L<sup>-1</sup>) in samples of abandoned tailings from San Felipe de Jesús area

	As	Cd	Cu	Fe	Mn	Pb	Zn	Sb
<i>Efflorescence crusts from oxidized tailings</i>								
EO2	0.3	> 100	92.6	24	> 5000	141.5	> 2500	0.04
EO4	1.4	> 100	> 2000	61	> 5000	109.0	> 2500	0.22
<i>Efflorescence crusts from un-oxidized tailings</i>								
ENO2	10.4	> 100	85.0	157	> 5000	5.0	> 2500	1.29
ENO3	53.0	> 100	930.0	26,200	> 5000	0.5	> 2500	0.87
<i>Un-oxidized tailings</i>								
JNO8	8.1	> 100	0.7	7	> 5000	184.5	> 2500	2.44
JNO9	1.5	4.5	< 0.05	4	96.3	8.7	104.0	0.88
JNO10	0.9	1.2	< 0.05	5	78.7	7.6	11.1	1.41
JNO11	3.4	0.2	< 0.05	5	28.8	4.2	5.3	4.82
JNO12	0.6	8.1	0.06	163	1740	31.3	1945	0.02
<i>Oxidized tailings</i>								
JO1	0.3	0.5	4.1	211	44.1	31.9	59.2	0.02
JO3	7.6	10.1	35.3	731	862	0.5	1020	0.19
JO4	0.1	10.6	186.5	114	1740	119.5	2240	0.002
JO5	0.01	0.02	0.1	4500	75.3	0.006	6.3	0.48
<i>Road dust</i>								
PC1	0.8	0.008	2.0	6	1.7	0.3	0.9	0.14
PC5	0.1	0.021	0.9	17	1.0	1.6	3.6	0.02
Reference values	0.01 <sup>a,b</sup>	0.00025 <sup>c</sup>	0.01 <sup>c</sup>	0.3 <sup>d</sup>	0.15 <sup>d</sup>	0.01 <sup>b</sup>	0.12 <sup>c</sup>	0.02 <sup>e</sup>

<sup>a</sup>USEPA (2012)<sup>b</sup>Council Directive 98/83/EC (1998)<sup>c</sup>USEPA (2002)<sup>d</sup>Secretaría de Salud (1994)<sup>e</sup>WHO (2003); concentration values below lower detection limit (<) and above upper detection limit (>). Detection limits (mg/L) are As (0.01), Cd (0.001), Cu (0.05), Fe (1.0), Mn (0.05), Pb (0.005), Zn (0.05), Sb (0.0005)

region, so far, exceed those obtained for abandoned mine wastes inventoried in Spain (Arranz-González et al. 2016), which highlights the environmental and health concerns associated with abandoned mining sites in semi-arid zones from northwestern Mexico.

### Multivariate Analysis

A PCA was performed considering element concentrations (total and soluble) and mineralogy, determined in samples of mining waste and road dust from the San Felipe de Jesús town, with the purpose of identifying the relationships among the mentioned variables. The results of principal component analysis identify four components explaining 94.4% of the total variance (Table 5). Factor 1 explains 32.8% of variance, which defines the association of primary minerals (sulfides, carbonates), total elemental concentrations of As and Sb, and soluble concentrations of Sb, and showed lower weighted variables with Pb and Fe; additionally, Factor 1 is associated with un-oxidized mine tailings. The relationship between high concentrations of As,

Pb, Sb and Fe is expected to occur due to the presence of sulfides. In spite of the higher concentrations of As and Sb, in Factor 1, only soluble concentrations of Sb are related, which is explained by the differences in solubility between As and Sb (e.g., Mitsunobu et al. 2006). However, some aspects regarding the geochemical behavior of Sb in comparison with As still need to be understood (Wilson et al. 2010). The high soluble contents of Sb, obtained in un-oxidized tailings from San Felipe de Jesús, should be studied in detailed because of the ecological and health risk that might represent. On the other hand, it is interesting to note the absence of relationship of PTE such as Zn and Cu with this factor, since these elements are characteristic of mineralization ores exploited in the area. Factor 2 explains 30.63% of variance (Table 5), and relates concentrations of Zn, Mn and Cd, with soluble concentrations of same elements, and to a lesser extent with efflorescent salts (MgSO<sub>4</sub> + H<sub>2</sub>O); this relationship is expected due to the presence of highly soluble minerals. The samples related to Factor 2 are efflorescent minerals developed on oxidized mine tailings, oxidized tailings and road

**Table 5.** Rotated component matrix of a principal component analysis of PTE concentration, solubility and mineralogy obtained in un-oxidized tailings, oxidized tailings, efflorescence crusts developed on both types of tailings and road dust samples from San Felipe de Jesús area

	F1	F2	F3	F4
Jarosite (%)	0.004	0.003	<b>0.849</b>	0.010
MgSO <sub>4</sub> + H <sub>2</sub> O (%)	0.136	<b>0.592</b>	0.047	0.109
Sulfides (%)	<b>0.956</b>	0.003	0.027	0.001
Calcite (%)	<b>0.712</b>	0.042	0.113	0.007
As(s)	0.041	0.034	<b>0.872</b>	0.008
Cd(s)	0.103	<b>0.813</b>	0.023	0.054
Cu(s)	0.018	0.086	0.028	<b>0.862</b>
Fe(s)	0.007	0.016	<b>0.862</b>	0.024
Pb(s)	0.270	0.270	0.257	0.044
Sb(s)	<b>0.809</b>	0.086	0.011	0.013
Zn(s)	0.103	<b>0.786</b>	0.026	0.050
Mn(s)	0.103	<b>0.812</b>	0.024	0.053
Pb	<b>0.603</b>	0.025	0.335	0.007
As	<b>0.967</b>	0.012	0.012	0.001
Zn	0.001	<b>0.930</b>	0.005	0.030
Cu	0.023	0.068	0.013	<b>0.851</b>
Fe	<b>0.613</b>	0.032	0.287	0.021
Sb	<b>0.905</b>	0.039	0.046	0.009
Cd	0.039	<b>0.865</b>	0.017	0.030
Mn	0.147	<b>0.616</b>	0.110	0.040

Bold values indicate highly weighted variables for the respective principal components. Soluble concentrations (s). Rotation method: Varimax with Kaiser normalization

dust of San Felipe de Jesús town; this relationship is supported by the mineralogy and enrichment factor spectra. Factor 3 explains 19.80% of variance and correlates soluble concentrations of As and Fe, with Jarosite; Factor 3 is related to efflorescent salts developed on un-oxidized tailings. This relationship is unusual because the presence of insoluble minerals, such as jarosite, is assumed to act as metal and metalloid fixer (e.g., Corrales-Pérez and Romero 2018). Finally, Factor 4 explains 11.10% of variance, describing the relationship between total concentration and soluble Cu, and related to efflorescent minerals developed on oxidized tailings, indicating that the formation of efflorescent crusts is complex because of the variability of total and soluble concentrations of PTE.

## CONCLUSIONS

Abandoned mine tailings, located near San Felipe de Jesús town, act as an important source of pollutants. The semi-arid conditions of the region enhance oxidation processes, allowing the formation

of efflorescent minerals (hydrated sulfates) over tailings surface, such as rozenite, starkeyite, kieserite, szolmonokita, epsomite, which are by highly soluble and may carry potentially toxic elements. Wind dispersal of the mine residue material is demonstrated by the presence of rozenite in rural dust, which in this study, serves as a mineral index that support mobility. Concentrations of potentially toxic elements (Pb, As, Zn, Sb) in abandoned mine tailings, agricultural soils and road dust from San Felipe de Jesús area not only exceed the limits set for environmental Mexican regulations, but also exceed international threshold and guideline values, representing some of them as an ecological (Pb, As, Zn, Sb) and health risk (Sb and Pb).

Normalized multi-elemental patterns of enrichment factors of the studied sample types, multivariate analyses and mineralogy support that abandoned mine tailings material impacts rural dust from San Felipe de Jesús. Contamination index values on un-oxidized and oxidized mine tailings, as well as efflorescent crusts, indicate a very high potential of pollution if dispersing toward surrounding soils and sediments. Hazard average quotient values of un-oxidized and oxidized tailings, as well as efflorescence crusts generated on both types of tailings, exceed the maximum toxicity value, suggesting a very high risk of contamination of surface water bodies by runoff, or underground water by infiltration, implying that mobility of soluble PTE may represent a potential health risk via ingestion route. Finally, based on the findings obtained, it is highly recommended the application of phytostabilization techniques to attenuate the dispersal of PTE from the studied abandoned tailings deposit.

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