

# Contamination Potential of an Urban Mine Tailings Deposit in Central Mexico —A Preliminary Estimation

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## Abstract

This study consisted of a geochemical analysis of the Dos Carlos tailings' deposit located in the Mining District of Pachuca-Real del Monte in the state of Hidalgo, Central Mexico. The goal of the study was to determine the potential effects of this deposit on the environment and health of the population of the metropolitan area of Pachuca. Sampling was conducted from the top to the base of two raised sections at opposite ends of the deposit, and macroscopic features (profiles A and B) of these deposits were evaluated. Subsequently, mineralogical analyses of the collected samples were performed using X-ray diffraction and physico-chemical analysis of the leachates. The results were compared with the maximum permissible limits established by different national and international standards for drinking water and hazardous waste. In addition, geochemical modeling was conducted using PHREEQC to calculate the distribution of aqueous species, ionic activities and saturation indices. Of the 35 samples of leachate analyzed for profile A, two samples had two to nearly three times the national permissible maximum limits of Cr<sup>6+</sup> in drinking water, four samples had NO<sub>3</sub><sup>-</sup> levels that exceeded these limits and 19 samples had NO<sub>2</sub><sup>-</sup> levels that exceeded these limits. Of the 10 samples analyzed from profile B, six samples had NO<sub>3</sub><sup>-</sup> levels that were up to fourteen times higher than the limit for human consumption, and six samples had levels of NO<sub>2</sub><sup>-</sup> that were up to 28 times higher than the permissible limit. The predominant minerals found in the Dos Carlos tailings' deposit were quartz, feldspar, clay from the kaolinite group, and calcite and or-

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thopyroxene (enstatite-ferrosilite series). The samples from profile A indicated that minerals containing  $\text{Ag}_{\text{metallic}}$ ,  $\text{Cd}^{2+}$ ,  $\text{Cu}^+$ ,  $\text{Ni}^{2+}$ ,  $\text{Pb}^{2+}$  and  $\text{Zn}^{2+}$  were dissolved, whereas minerals containing  $\text{Fe}_{\text{total}}$ ,  $\text{Mn}^{2+}$ ,  $\text{Pb}(\text{OH})_3$  and  $\text{Zn}^{2+}$  were in a state of precipitation. In profile B, the dissolution of minerals containing  $\text{Ag}_{\text{metallic}}$ ,  $\text{As}^{5+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Cu}^+$ ,  $\text{Mn}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Pb}^{2+}$  and  $\text{Zn}^{2+}$  was observed only in a superficial sample, whereas  $\text{Ag}_{\text{metallic}}$ ,  $\text{Fe}_{\text{total}}$ ,  $\text{Mn}^{2+}$ ,  $\text{Pb}_3(\text{PO}_4)_2$  and  $\text{PbHPO}_4$  were found in a precipitated state. The above-mentioned species that occurred in a state of dissolution pose an environmental threat to the groundwater in the study area; indeed, the tailings' deposit provides the conditions necessary for these minerals to leach and infiltrate into the subsoil or be transported by wind, deposited in the soil of the surrounding area and inhaled by the nearby population, potentially compromising respiratory health.

## Keywords

Contamination; Dos Carlos Tailings; Mining District Pachuca-Real del Monte

## 1. Introduction

In Mexico, one of the major byproducts of mining activity is the generation of millions of tons of tailings or slurries. Some mining sites in the country have had produced tailings with environmental impacts, such as those located in the Mexican localities of Taxco, Guerrero [1]-[3]; Guanajuato, Guanajuato [4]-[7]; Santa Bárbara, Chihuahua [8] and Tizapa, Estado de México [9]-[11]. However, there are many sites where the source of contamination from tailing deposits is not obvious. One example is the tailings from the mining unit of Charcas, San Luis Potosí, which does not represent potential generators of acidity and does not contain soluble toxic elements; therefore, it is not considered hazardous [12].

The state of Hidalgo, an area with significant environmental problems, contains mining waste from the Zimapán district. The waste from this district is classified as dangerous because of its toxicity; in particular, even in stable forms, the arsenic content of the waste can contaminate the environment [13]. In the city of Pachuca, the accumulation of tailings in the urban area began in 1912 by order of the Department of Mining of the Secretariat of Industry and Trade [14]; this order was aimed at storing mining waste until techniques to recover the silver contained within the waste could be improved [15]. The Dos Carlos tailings' deposit began in response to this order. In 1959, this deposit contained approximately 55 million tons of waste [16]. Forty-seven million tons of these tailings were produced by the cyanidation of minerals from all parts of the mining district; eight million tons came from the only flotation plant of the Mining District of Pachuca-Real del Monte [17]. To date, the tailing deposits of the district have been investigated for a variety of purposes [18] [19]; however, few investigations have evaluated the individual deposits for environmental purposes. In this context, this paper presents a preliminary investigation of the contamination potential that the Dos Carlos tailings' deposit of the Mining District of Pachuca-Real del Monte in the state of Hidalgo presents to the environment and the health of the surrounding population.

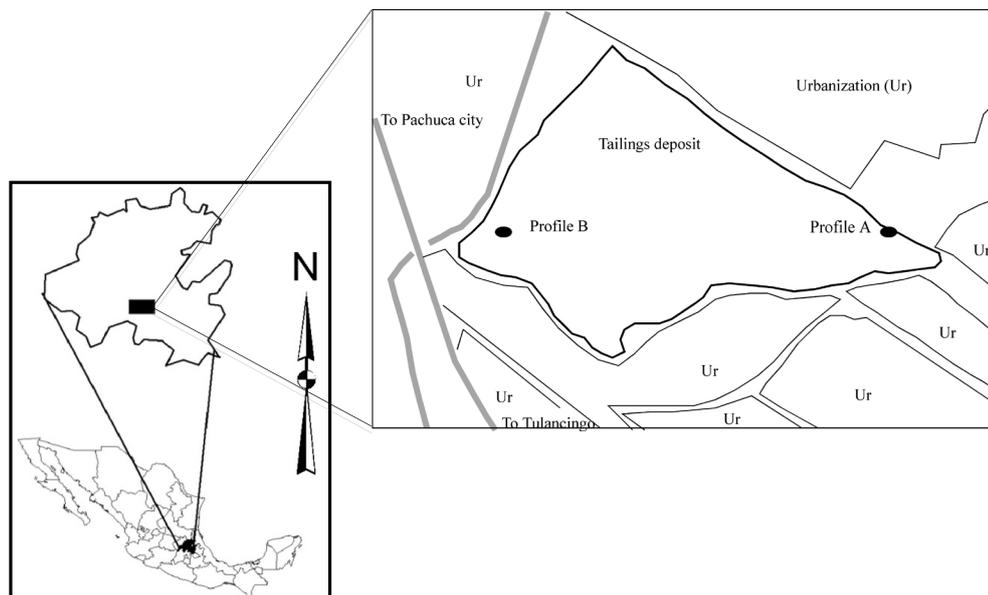
## 2. Description of the Study Area

### 2.1. Location

The tailings deposit known as Dos Carlos is located at the coordinates  $20^{\circ}06'20''$  latitude north and  $98^{\circ}42'46.8''$  longitude west to the north of the municipality of Pachuca in the state of Hidalgo (Figure 1). The deposit has an elevation of 2437 meters above sea level (masl) [20].

### 2.2. Climate

According to the climatic classification of [21] and the modified classification of [22], the city of Pachuca has a dry climate of subtype BS1k'w(w)(I'). The rainy season usually occurs from April to September, and maximum average precipitation levels (203.9 mm) occur in the month of May; however, in some years, the rainy season can begin in March and extend through late October or early November. In general, Pachuca has a temperate



**Figure 1.** Location of the study area (No scale).

climate. Its annual average temperature oscillates between 11.3°C and 16.6°C. The highest temperatures occur in April (maximum temperatures of 28.8°C) and the lowest temperatures occur in January (minimum temperatures of -3°C). Most of the year is dominated by winds that blow in a north-south and northeast-southwesterly direction and reach an average speed of 22 to 24 m/s [23].

### 2.3. Description of the District and Tailings Deposit

Pachuca-Real del Monte is one of the oldest silver-producing mining districts of Mexico. It is located in Sierra de Pachuca in the metallogenic province of the Mexican Neovolcanic Axis. The site is characterized by its epithermal polymetallic deposits of silver, lead, zinc, copper and gold [24]. Hundreds of productive veins are scattered along a surface with an area of 70 km<sup>2</sup>; this area has been exploited for more than 500 years and has yielded 40 million kg of silver from a total of 100 million metric tons of ore with 1% of total sulfides. These veins have a series of calc-alkaline volcanic rocks from the Oligocene-Miocene age (Figure 2) with a thickness of 2700 m [25].

The tailings deposit, which is surrounded by housing complexes, is located in the northern part of the city, approximately 10 meters from the Sosa riverbed. Because it is directly exposed to the weather conditions, the mining waste deposit site was selected according to the area and volume of the site and its state of neglect and inactivity. We found no indication that the site is monitored, and no studies have been performed to analyze the behavior of the chemical speciation of the minerals of the site; consequently, the environmental contamination potential of these tailings and their effects on the health of the population are unknown.

## 3. Methods

### 3.1. Field Work

Two raised sections at opposite ends of the Dos Carlos tailings deposit were described based on the specifications established by the NOM-141-SEMARNAT-2003 [26], the NMX-AA-132-SCFI-2005 [27] and the NOM-052-SEMARNAT-2006 [28]. The selection criterion was that the deposit has had several periods of deposition, therefore, the profiles selected represent the oldest and most recent stage of mineral processing. The northeast end (profile A), which is approximately 5.32 m high with respect to the natural terrain, contains the most recently deposited tailings (deposited during the 1980s); the southwestern end (profile B), which is approximately 7 m high, contains the older tailings deposited in the 1950s (Figure 1). Macroscopic characteristics such as the thickness of the layer or stratum, particle size, color and presence of oxidation were described in both sections (Tables 1(a) and (b)).

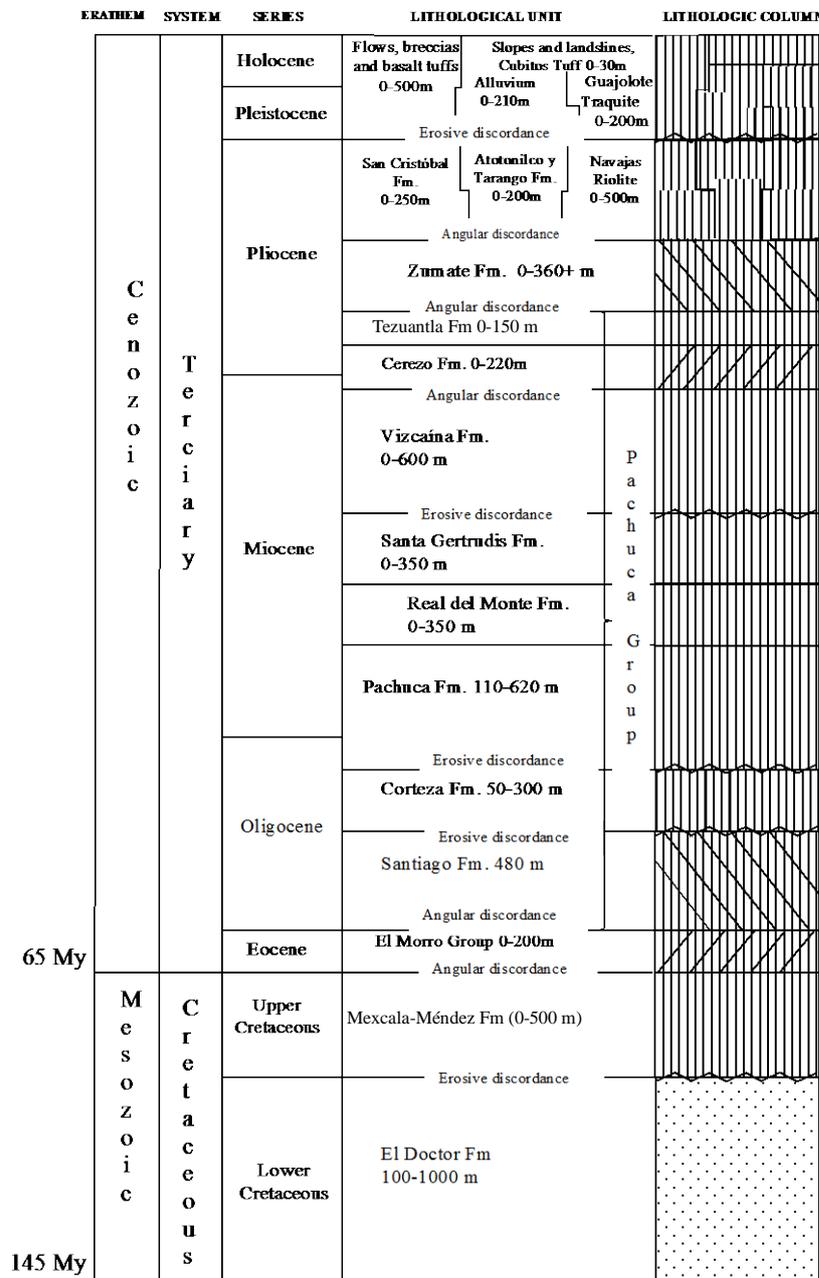


Figure 2. Stratigraphic column of the Mining District Pachuca-Real del Monte. No scale (Edited from [17]).

A total of 45 samples were collected from the top to the base of the studied sections based on changes in the different aforementioned macroscopic characteristics (35 from profile A and 10 from profile B). The collected samples were sieved (mesh #60) to remove plant debris and non-tailing rock fragments. Subsequently, the samples were weighed to determine their total weight in grams, placed in pre-labeled polyethylene bags and transported to the Environmental Geology laboratory of the Academic Area of Earth and Material Sciences of the Autonomous University of the state of Hidalgo for analysis.

### 3.2. Laboratory Work

The following parameters were evaluated in the leachates: a) percent humidity, b) physical parameters such as

**Table 1.** (a) Macroscopic characteristics of leachates in profile A. (b) Macroscopic characteristics of profile B.

(a)

Sample	Stratum thickness (cm)	Depth (cm)	Macroscopic characteristics
1A	110	0 - 110	Loose material with colors ranging from dark brown to orange, presence of weathering and plant roots. Represents the top of the profile.
2A	5	115	Black coloration
3A	24	139	Silty, dark brown to gray
4A	1	140	Sandy, light gray
5A	16	156	Silty toward the base and at the top, dark brown; sandy in the center, gray
6A	1	157	Sandy, dark gray
7A	3	160	Silty, dark to light brown
8A	4	164	Sandy, light gray
9A	25	189	Intercalated layers of sand and dark brown silt
10A	5	194	Sandy, light gray
11A	20	214	Silty, light to dark brown
12A	4	218	Sandy, dark gray with oxidation stains
13A	3.5	221.5	Sandy, light to dark gray
13A'	4	222	This sample comes from the same layer as sample 13A. It has the same texture and coloration; in addition, reddish to orange oxidation is present.
14A	12	233.5	Silty, light brown to dark brown
15A	2	235.5	Sandy, light gray to dark gray
16A	15	250.5	Silty, light brown to dark brown
17A	7	257.5	Sandy, very fine texture, light gray color, very compact
18A	17	274.5	Silty, light brown, very compact
19A	13	287.5	Intercalated layers of sand and silt, light gray to light brown
20A	18	305.5	Silty, light brown with sandy intercalated layers
21A	9	314.5	Silty, light brown, very compact
22A	16	330.5	Sandy, light gray; in the center, there is a light brown silt layer (1.5 cm)
23A	37	367.5	Silty, light brown with small alternating layers of light gray sand (3 - 4 cm)
24A	19	386.5	Alternating layers of sand and silt, dark gray to light gray; each sublayer is 3 to 4 cm thick
25A	33.5	420	Sandy with alternating layers of silt and very fine light brown to light gray sand with oxide lenses 1 to 2 cm thick
Q	4	424	Sandy, light gray, slightly compact
27A	7	431	Silty, dark brown with black nodules, very compact, moist
28A	5.5	436.5	Sandy, light gray, very loose material
29A	19.5	456	Silty, dark brown, with a layer of medium-grain dark gray sand with small black spots
30A	22.5	478.5	At the top, there is a layer of medium-grained light gray sand with a sheet of nodules. The center is light brown and silty with oxide lenses. At the base, there is a layer of fine to medium sand with oxide lenses
31A	32	510.5	Clay with alternating cream to very light brown layers; the last 9.1-cm-thick layer from the base consists of alternating layers of cream-colored clay and dark material with small oxide lenses
32A	10	520.5	Silty, dark brown, very compact
33A	11	531.5	Fine sand, light gray at the top and base and dark gray in the center

(b)

# of escarpment	Sample	Macroscopic characteristics
1	10B	Clay with thin layers of fine light gray sand. Has a horizontal fracture filled with fine light gray sand. Presence of vegetation on the top of this escarpment.
2	9B	Clay, dark to light brown, moist, very compact except on the top where the material is soft
3	8B	Clayey, with intercalating dark and light brown layers, presence of small reddish surfaces
	7B	Very narrow intercalating light and dark brown layers; the sublayers show surface runoff
	6B	Intercalating layers of clay with very fine dark to cream brown sand, slightly compact, presence of small red surfaces, dry and rough
4	5B	Clay, light brown, with one end cradled horizontally
	4B	Has the appearance of a clayey dark brown conglomerate; toward the top, small reddish surfaces with white edges
	3B	Toward the base, light brown clay; toward the top, fine, light gray sand with black spots, very compact
5	2B	Sandy, fine grain, with yellow to light reddish brown oxidation
	1B	Intercalating layers of clay and fine sand at the base; the center is made up of reddish clay; at the top, intercalating layers of clay and fine sand are found.

temperature, pH, electrical conductivity (EC), redox potential (Eh) and dissolved solids (measured with electrodes, models HI98120 and HI98129-Hanna<sup>®</sup> instruments, USA-), c) levels of ions such as Cr<sup>6+</sup>, Cu<sup>+</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, P<sup>+</sup>, SiO<sub>2</sub> and Zn<sup>2+</sup>, d) mineralogical contents using X-ray diffraction, e) geochemical modeling, and f) average daily dose.

### 3.2.1. Percent Humidity

To determine the percent humidity, the samples were sieved and weighed in the field (Ph) and desiccated in an ORL-V (O.R.L., USA) oven for 24 hours at 150°C to eliminate the existing moisture. The samples were then weighed again (Ps). The percentage of total humidity (%th) was calculated:

$$\%th = \left[ \frac{(Ph - Ps)}{Ph} \right] \times 100 \quad (1)$$

### 3.2.2. Physical Parameters and Levels of Ions

Parameters such as temperature, dissolved solids, pH and electrical conductivity were evaluated with a Combo pH and EC electrode, model HI98129 (-Hanna<sup>®</sup> instruments, USA), that was previously calibrated with reference buffers (pH 4.01 ± 0.01 at 25°C and pH 7.00 ± 0.01 at 25°C). The oxidation-reduction potential (ORP or Eh) was measured under the same conditions used for the pH and EC with a Hanna Instruments ORP potentiometer, model HI 98120 calibrated by the manufacturer (-Hanna<sup>®</sup> instruments, USA).

Subsequently, the samples were prepared to evaluate the above-mentioned physical and chemical parameters: 100 g of tailings were added to 500 ml of distilled water and left for 48 hours. Three vials of distilled water without tailings were used as controls in all analyses. The analytical quality was calibrated using samples of certified High Purity Standards. Detection limits were as follows: Cr<sup>6+</sup> (0.005 mg/L), Cu<sup>+</sup> (0.001 mg/L), NO<sub>3</sub><sup>-</sup> (0.5 mg/L), NO<sub>2</sub><sup>-</sup> (0.02 mg/L), PO<sub>4</sub><sup>3-</sup> (0.04 mg/L), P<sup>+</sup> (0.3 mg/L), SiO<sub>2</sub> (0.03 mg/L) and Zn<sup>2+</sup> (0.003 mg/L); these limits were determined using a multiparametric photometer, model HI83206 (-Hanna<sup>®</sup> instruments, USA). The error in the ionic balance was verified to be less than 10%.

The values obtained in the leachate analysis were compared with the maximum permissible limits (MPL) a) for drinking water established by the Mexican standards NOM-127-SSA1-1994 [29], by the United States [30] and by the World Health Organization [31]; b) for water discharged into rivers and artificial reservoirs established by the NOM-001-SEMARNAT-1996 [32] and c) for the classification of waste considered hazardous to the environment because of its toxicity according to the NOM-052-SEMARNAT-2005 [28].

### 3.2.3. Mineralogical Contents

The mineralogical analysis was conducted using X-ray diffraction (XRD). Fourteen samples with oxidation or high concentrations of  $\text{Cr}^{6+}$ ,  $\text{Cu}^+$ ,  $\text{SiO}_2$  and/or  $\text{Zn}^{2+}$  were selected from each of the samples; ten of these samples corresponded to profile A and four to profile B. The analyses were conducted at the Department of Geochemistry of the Geological Institute of the National Autonomous University of Mexico. The diffractograms were obtained with a Shimadzu XRD-6000 diffractometer (Shimadzu Corp., USA) equipped with a monochromator and copper tube. Two grams were required for each sample. The samples were milled with an agate mortar and analyzed in an aluminum sample holder (non-oriented fraction) at an angle of  $2\theta$  with intervals ranging from  $4^\circ$  to  $70^\circ$  and velocities of  $2^\circ/\text{minute}$ .

### 3.2.4. Geochemical Modeling

The PHREEQC software for Windows (version 2.18.5570) was used for environmental geochemical modeling [33]. The speciation code WATEQ4F Version 2.63 [34] was selected to calculate the distribution of aqueous species, ionic activities and saturation indices based on the values of temperature, pH, Eh, and ionic concentrations determined in the laboratory. Using this simulation software, it was possible to analyze trends in dissolution, precipitation or equilibrium of a mineral in contact with water. The activity coefficients were obtained with the Debye-Huckel or Davis equations depending on the ionic strength of the solution [35]. The product of ionic activity (PIA) is used to calculate the saturation index (SI):

$$\text{SI} = \log(\text{PIA}/\text{Ksp}), \quad (2)$$

where Ksp is the solubility constant. With these data, it was possible to determine the tendency of water to dissolve a mineral ( $\text{SI} < 0$ ), precipitate ( $\text{SI} > 0$ ) or maintain equilibrium ( $\text{SI} = 0$ ).

### 3.2.5. Average Daily Dose

The risk to the surrounding population associated with the oral ingestion of  $\text{Cr}^{6+}$  was measured and was calculated as follows:

$$\text{ADD} = (\text{C} \times \text{IRw} \times \text{ED} \times \text{EF}) / (\text{BW} \times \text{AT} \times 365) \quad (3)$$

The average daily dose (ADD) specifies the amount of contaminant that is ingested by a Mexican adult with an average weight [36]. C is the concentration of the contaminant ( $\text{Cr}^{6+}$ , in this study mg/L), IRw is the ingestion rate per unit time (L/day), ED is the exposure duration (years), EF is the exposure frequency (days/year), BW is the body weight of the receptor (kg), and AT is the averaging time (years), equal to the life expectancy, and 365 is the conversion factor from year to days.

## 4. Results

### 4.1. Physical Characteristics of Tailings

Profiles A and B both have materials with a heterogeneous granulometry. Profile A consists of a 5.32-m-thick material composed of particles with sizes varying from fine sand to silt fractions, with the fine sand fraction being predominant (Tables 1(a) and (b)). The deposits are arranged in horizons with Munsell scale colors of light brown (6/3 7.5YR) and light gray (6/2 7.5YR) and thicknesses that vary from 1 to 37 cm; one of the horizons at the top has a peak thickness of 110 cm. In addition, the tailings have intercalated slightly darker (5/3 7.5YR) horizons with thicknesses ranging from 1 to 5 cm. The coloration is not associated with granulometry. Occasionally, profile A has 4-cm (12A) and 33.5-cm-thick (25A) gray (7/1 7.5YR) horizons with yellowish brown (7/8 10YR) and reddish brown (5/8 7.5YR) bands that were considered to be iron oxide. Profile B has the same granulometric characteristics and colorations as profile A; however, the reddish iron oxide layers (5/8 7.5YR) are more frequently encountered in profile B than in profile A. Toward the top of the deposit and distributed throughout, a 3-m-thick layer of intermediate gray color (4/1 5Y) that corresponds to a sandy loam soil type is found. On this soil horizon, vegetation from the families Cactaceae, Asteraceae, Poaceae, Leguminosae, Chenopodiaceae, Solanaceae and Cruciferae and the species *Brickelia veronicifolia*, *Zaluzania triloba*, *Solanum elaeagnifolium*, *Stipa tenuissima*, *Bouteloua curtipendula*, *Haplopappus venetus* and *Cinodon dactylon* can be found [37].

In both profiles A and B, the humidity content varies from 1% to 15%. The strata with the highest percent

humidity in both profiles consist mainly of sand-sized particles that easily lose moisture and are generally not observed in an oxidation state; oxidation was detected in both of the strata consisting of fine-textured particles of silt and/or clay (**Tables 1(a)** and **(b)**).

Variations in potential of hydrogen (pH) were observed within each profile. In profile A, the pH varies from 7 to 7.64; in profile B, the pH varies from 7.27 to 7.85. In both cases, the slightly acidic medium becomes slightly alkaline. Additionally, horizons with pH values of 6 occasionally exhibit reddish to brown sheets with thicknesses of only a few millimeters, which indicate the oxidation of Fe sulfides; in contrast, these sheets are absent in the horizons with pH values greater than 6 (**Tables 2(a)** and **(b)**).

## 4.2. Chemistry of the Tailings

The results of the chemical analysis of the leachates ( $\text{Cr}^{6+}$ ,  $\text{Cu}^+$ ,  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ ,  $\text{PO}_4^{3-}$ ,  $\text{P}^+$ ,  $\text{SiO}_2$  and  $\text{Zn}^{2+}$ ) for profiles A and B are summarized in **Tables 2(a)** and **(b)**, respectively. The levels in the leachates were compared to the national [29] and international [30] [31] environmental standards for drinking water and for water discharged into rivers and artificial reservoirs [32]. These levels were also examined in the context of the classification of hazardous waste considered toxic to the environment [28] to evaluate whether the infiltration of these leachates into the ground water poses a risk to inhabitants consuming the drinking water. Of the 35 leachates samples analyzed for profile A, only samples 2A and 33A, had levels of  $\text{Cr}^{6+}$  (maximum = 0.014 mg/L; average = 0 mg/L) that were, respectively, twice- and almost three-fold higher than the maximum permissible limit (MPL) established for drinking water by [29]-[31]. Samples 26A, 29A, 31A and 32A had levels of  $\text{NO}_3^-$  (maximum = 12.85 mg/L; average = 4.25 mg/L) that slightly exceeded the MPLs established by the standards [29] [30]. Samples 1A, 3A, 5A, 10A, 11A, 12A, 16A, 17A, 18A, 19A, 20A, 21A, 23A, 25A, 26A, 27A, 29A, 31A and 32A had  $\text{NO}_2^-$  levels (maximum = 1.57 mg/L; average = 0.30 mg/L) that exceeded the MPL [29] [30].

Of the 10 samples analyzed in profile B, samples 1B, 2B, 3B, 4B, 7B and 8B had  $\text{NO}_3^-$  levels (maximum = 146.19 mg/L; average = 38.96 mg/L) that were 14 times higher than the MPL for drinking water established by the [29] [30] and three times higher than the MPL established by the [31]. Samples 1B, 2B, 3B, 6B, 9B and 10B had levels of  $\text{NO}_2^-$  (maximum = 1.57 mg/L; average = 0.48 mg/L) that were up to 28 times higher than the MPLs established by the [29] and slightly higher than the MPL established by the [30].

**Table 2.** (a) Concentrations from the chemical analysis of Profile A. (b) Concentrations from the chemical analyses of Profile B.

(a)											
	pH	Eh	E.C.	$\text{Cr}^{6+}$	$\text{Cu}^+$	$\text{NO}_3^-$	$\text{NO}_2^-$	$\text{PO}_4^{3-}$	$\text{P}^+$	$\text{SiO}_2$	$\text{Zn}^{2+}$
		mV	$\mu\text{S/cm}$					mg/L			
Maximum	7.64	217	1102.9	0.075	5.1067	146.19	1.406	3.575	1.3	2.2	1.96
Minimum	7	175	723.2	0	0	0	0	0.74	0	0	0
Average	7.375	193.5	938.73	0.0162	1.0791	56.81	0.48	1.91	0.64	1.74	0.32
Std. Dev.	0.17	13.48	0.17	0.0231	1.479	63.13	0.45	0.84	0.47	0.65	0.59

(b)											
	pH	Eh	E.C.	$\text{Cr}^{6+}$	$\text{Cu}^+$	$\text{NO}_3^-$	$\text{NO}_2^-$	$\text{PO}_4^{3-}$	$\text{P}^+$	$\text{SiO}_2$	$\text{Zn}^{2+}$
		mV	$\mu\text{S/cm}$					mg/L			
Maximum	7.85	2248.92	270	0.014	3.675	12.84	1.57	5.08	3	2.75	1.93
Minimum	7.27	237.5	182	0	0	0	0	0	0	0	0
Average	7.568	1165.3	228.02	0.00261	0.491	4.14	0.30	1.81	0.46	2.05	0.32
Std. Dev.	0.162	542.91	22.86	0.00381	0.669	3.90	0.36	1.37	0.66	0.51	0.53

### 4.3. Mineralogical Analyses Using XRD

The mineralogical variability of the samples of tailings was minimal (**Table 3**). The predominant minerals and mineralogical groups were quartz, feldspar, clays of the kaolinite group (to 7.3 Å), as well as calcite and orthopyroxene (enstatite-ferrosilite series). Phase identification was performed using the PDF-2 database of the International Center for Diffraction Data (ICDD) with the program Jade v.5. The Powder Diffraction File (PDF) patterns associated with peak identification were 00-019-0002, 00-046-1045, 00-014-0164, 00-047-1743 and 00-019-0607, which correspond to orthoclase, quartz, kaolinite-1 Å, calcite and ferrosilite, respectively. The orthoclase pattern was associated with potassium feldspars, and ferrosilite was associated with a mineral of the enstatite-ferrosilite series. The adjustment process of the diffractometer allowed for refinement of the unit cell parameters. Phase quantification was carried out using the Reference Intensity Ratio (RIR) method [38].

In profile A, high concentrations of quartz were found in all samples (77.3% - 91.79%), followed by orthoclase (4.28% - 20.5%) and kaolinite (2.2% - 10.08%); calcite (0.84% - 5.10%) was found at a depth of 250.5 cm, and ferrosilite (5.5% - 8.9%) was found only in the first 115 cm of depth. In profile B, quartz, orthoclase and kaolinite were present in the four selected samples (5.34% - 19.53%, 9.5% - 27.2%, and 58% - 76.5%, respectively); calcite and ferrosilite were only found in one surface level sample (1.54% and 3.4%, respectively).

### 4.4. Mineral Speciation and Geochemical Modeling

The results of the selected samples from profiles A and B are shown in **Figure 3**. In profile A, geochemical modeling showed that minerals containing  $\text{Ag}_{\text{metallic}}$ ,  $\text{Cd}^{2+}$ ,  $\text{Cu}^{+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Pb}^{2+}$  and  $\text{Zn}^{2+}$  were dissolved, whereas minerals containing  $\text{Fe}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Pb}(\text{OH})_3$  and  $\text{Zn}^{2+}$  were in a state of precipitation. In the case of profile B, minerals that contain  $\text{Ag}_{\text{metallic}}$ ,  $\text{As}^{5+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Cu}^{+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Ni}^{2+}$  and  $\text{Pb}^{2+}$  were dissolved, and dissolved  $\text{Zn}^{2+}$  was only found in a surface sample. Minerals containing  $\text{Ag}_{\text{metallic}}$ ,  $\text{Fe}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Pb}_3(\text{PO}_4)_2$  and  $\text{PbHPO}_4$  were found in a precipitated state. The ionic strength in solution for samples of profiles A and B varied from  $2.198^{-3}$  to  $3.755^{-3}$  and from  $2.662^{-3}$  to  $7.777^{-3}$ , respectively.

**Table 3.** Mineralogical composition of samples of tailings (XDR).

Sample	Mineral species (%)				
	Quartz SiO <sub>2</sub>	Orthoclase (K,Na)AlSi <sub>3</sub> O <sub>8</sub>	Kaolinite Al <sub>2</sub> O <sub>3</sub> ·2SiO <sub>2</sub> ·2H <sub>2</sub> O	Calcite CaCO <sub>3</sub>	Ferrosilite Fe <sub>2</sub> Si <sub>2</sub> O <sub>3</sub>
PROFILE A					
1A	83.3	6.8	4.4		5.5
2A	78.9	9.7	2.4		8.9
7A	85.8	9.2	4.9		
16A	81.49	6.10	10.08	2.33	
17A	84.75	10.8	3.61	0.84	
23A	77.3	20.5	2.2		
27A	81.26	7.14	6.51	5.10	
33A	91.79	4.28	2.66	1.27	
34A	82.03	9.85	7.02	1.10	
PROFILE B					
1B	67.53	12.94	19.53		
2B	76.5	9.5	14.0		
7B	58	27.2	11.4		3.4
10B	71.75	21.36	5.34	1.54	

Blank cells indicate that the mineral species was not identified or was below the detection limit of the analytical equipment.

## 4.5. Risk for Human Exposure

This risk determination defines the risk of contaminants entering the body via a variety of routes (*i.e.*, via inhalation or oral ingestion of contaminated water). The magnitude, frequency and duration of the contact between human beings and the contaminant were estimated [39]. For this study, exposure to  $\text{Cr}^{6+}$  was assumed to principally occur via oral ingestion of contaminated water, as levels of this toxic contaminant in the leachates exceeded the MPL for drinking water. **Table 4** summarizes the data used in calculations (Equation (3)) and the average daily dose for  $\text{Cr}^{6+}$  that an inhabitant of the city of Pachuca might consume:  $1.45 \times 10^{-4}$  mg/L/day.

## 5. Discussion

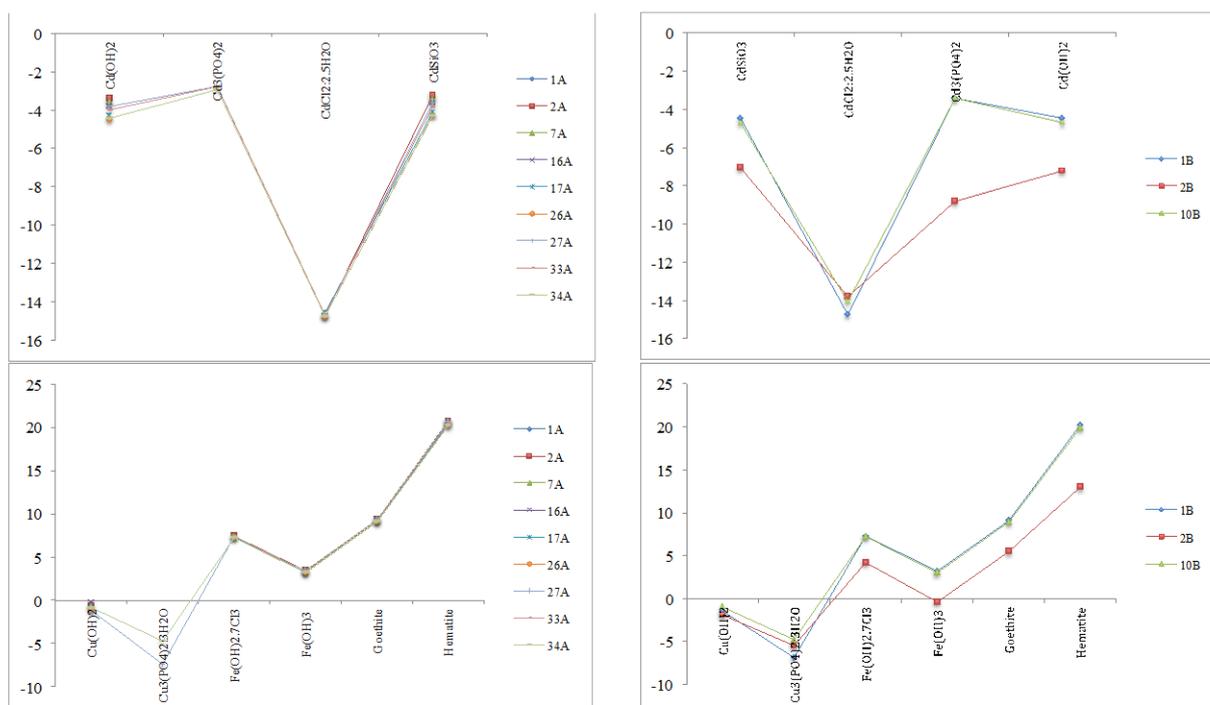
The integration of the results obtained by different analyses and data from previous studies conducted in the area provides a general overview of the effect that the tailings from the Dos Carlos deposit can have on the environment and, consequently, on the population of Pachuca and its urban area.

### 5.1. Physical Characteristics of the Tailings Deposit

The tailings deposit is predominantly composed of fractions of fine sand, with some fractions of silt and clay. [40] suggests that there is a direct relationship between fine particle size and the ease with which particles release their minerals when they come in contact with atmospheric oxygen or water. Previous work [41] reports that minerals are released more easily in fractions with sizes of  $70 \mu\text{m}$  or less than in fractions with sizes greater than  $70 \mu\text{m}$ . In contrast, studies carried out in a mining site in the State of Mexico [11] reported that mineral release occurred to the greatest extent in particulate fractions with sizes of  $70 \mu\text{m}$ , which correspond to fine sands and represent the finest fraction observed in the tailings.

It is known that the greater the dilution imposed on the particle, the greater the separation efficiency of its chemical components [42]. Similar results were obtained in laboratory [43], observing that tailing particulates less than  $1000 \mu\text{m}$  in size had high levels of heavy metals. [44] used wet centrifugation and observed that the weight of the smallest particles in the tailings allowed the particles to more easily separate from sulfuric minerals; however, this separation occurred only in the fraction of fine sand.

The preceding discussion suggests that the fine sands of the Dos Carlos tailings are less likely to become saturated in water than the silt particles; however, mineral release is favored in both fractions (although to a lesser





**Table 4.** Exposure factors for an adult Mexican living near the tailings deposit.

Factor/Parameter	Symbol	Unit	Residential	Data source
Concentration of the contaminant: Cr <sup>6+</sup>	C	mg/L	0.014	This study
Ingestion rate (Drinking water)	IR <sub>w</sub>	L/day	2.0	[39]
Exposure Duration	ED	Years	30	[56]
Exposure Frequency	EF	Days/year	365	[56]
Body Weight	BW	Kg	74.8	[36]
Averaging Time	AT	Years	77	[36]

tained in the host rock. This type of leaching into the Dos Carlos deposit was reflected in the slightly acidic horizons (pH values of 5.93 to 6.83) where evidence of the oxidation of iron sulfides was observed.

## 5.2. Chemistry of the Tailings Deposit

Chemical analyses of the tailings deposit identified the presence of Cr<sup>6+</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup> [28]-[32]; levels of these contaminants were two, three and up to 14 times higher, respectively, than the maximum permissible limits (Tables 2(a) and (b)).

An analysis of leachates from eight samples collected on the surface (first 50 cm) of the Dos Carlos tailings [18] reported levels of Cu<sup>+</sup> (maximum = 1.045 mg/L; average = 0.43 mg/L), Cd<sup>2+</sup> (maximum = 0.625 mg/L; average = 0.244 mg/L), Fe<sup>3+</sup> (maximum = 4.16 mg/L; average = 1.07 mg/L), Mn<sup>2+</sup> (maximum = 53.55 mg/L; average = 29.83 mg/L), Pb<sup>2+</sup> (maximum = 1.685 mg/L; average = 0.8 mg/L), Ag (maximum = 0.41 mg/L; average = 0.12 mg/L) and Ni<sup>2+</sup> (maximum = 0.6 mg/L; average = 0.12 mg/L). This analysis concluded that the tailings of the Dos Carlos deposit are potential sources of acid mine drainage [18]. The values obtained in this study were compared with the values obtained by [18] and with the standards established [28]-[32]. Two, eight, three, eight, five, one and three samples had levels of Cu<sup>+</sup>, Cd<sup>2+</sup>, Fe<sup>3+</sup>, Mn<sup>2+</sup>, Pb<sup>2+</sup>, Ag and Ni<sup>2+</sup>, respectively, that exceeded the MPLs [30] [31].

When the leachates mix with ground water, the concentrations of pollutants are diluted in the new water body, resulting in pollutant levels that are only slightly higher than the MPLs; consequently, the risk of compromising the water quality is negligible or nonexistent. However, for ions with levels that were several-fold higher than the MPL (as in the case of Cd<sup>2+</sup>), the risk is greater. Cd<sup>2+</sup> in particular should be given special attention, as this ion can be a gastric irritant and can cause vomiting and diarrhea in humans. In some short-term studies, the oral administration of soluble Ni to animals resulted in the accumulation of this compound in the kidneys, lungs, liver and heart (in that order), and absorption via the lungs resulted in toxicity and carcinogenicity [47].

The particulates in the tailings were observed to influence the chemical composition of the soil in the metropolitan area of Pachuca. [48] reported Cu<sup>+</sup> concentrations of 34.68 - 119.88 mg/kg and Cd<sup>2+</sup> concentrations of 0.13 - 20.07 mg/kg in the soil around Pachuca; the authors concluded that the presence of these elements was associated with activities of the Mining District Pachuca-Real del Monte and that these compounds may have come from the Dos Carlos tailings. In contrast, these authors concluded that the concentrations of Mn<sup>2+</sup> (188 - 780 mg/kg) and Pb<sup>2+</sup> (9.64 - 114.6 mg/kg) were associated with atmospheric emissions produced by vehicles. However, the possibility that these elements could also come from the Dos Carlos tailings should not be excluded. In this context, performing atmospheric measurements to estimate the wind-borne spread of some of the ions from the tailings will be critical, as, for example, manganese can affect the nervous system in humans and cause deleterious neurological effects [49].

As shown in the correlation matrix of the physico-chemical parameters measured in this study (Table 5), strong positive correlations ( $r^2 > 0.50$ ) were observed for the following pairs in the indicated order: pH and E.C. > Eh, Cu<sup>+</sup> and Zn<sup>2+</sup> > E.C. > pH, NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup>, and *vice versa*. These correlations indicate that variations in the pH of the leachates could contribute to the dissolution or precipitation of ions, thereby producing a more acidic or neutral-alkaline environment, respectively. The correlation between Cu<sup>+</sup> and Zn<sup>2+</sup> is related to the type of deposit exploited; therefore, as the pH decreases, these ions will continue to be released. The presence of NO<sub>3</sub><sup>-</sup> can be related with agriculture soil eroded that was deposited by aeolian transportation; other source for nitrates can be associated with atmospheric emissions produced by vehicles [48].

**Table 5.** Cross correlation matrix of physico-chemical parameters of the tailings deposit.

	pH	Eh	E.C.	Cr <sup>6+</sup>	Cu <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>	NO <sub>2</sub> <sup>-</sup>	PO <sub>4</sub> <sup>3-</sup>	P <sup>+</sup>	SiO <sub>2</sub>	Zn <sup>2+</sup>
<b>pH</b>	1.00										
<b>Eh</b>	<b>0.53</b>	1.00									
<b>E.C.</b>	<b>0.67</b>	<b>0.68</b>	1.00								
<b>Cr<sup>6+</sup></b>	0.47	0.31	-0.03	1.00							
<b>Cu<sup>+</sup></b>	0.21	<b>0.55</b>	<b>0.70</b>	-0.23	1.00						
<b>NO<sub>3</sub><sup>-</sup></b>	0.14	-0.11	-0.04	0.09	-0.19	1.00					
<b>NO<sub>2</sub><sup>-</sup></b>	-0.39	-0.24	-0.10	-0.42	0.13	<b>0.62</b>	1.00				
<b>PO<sub>4</sub><sup>3-</sup></b>	-0.64	-0.37	-0.75	0.13	-0.38	-0.03	0.16	1.00			
<b>P<sup>+</sup></b>	-0.60	-0.40	-0.69	-0.03	-0.46	0.31	0.22	<b>0.74</b>	1.00		
<b>SiO<sub>2</sub></b>	0.14	0.43	0.26	0.29	0.27	-0.46	-0.34	-0.08	-0.46	1.00	
<b>Zn<sup>2+</sup></b>	0.27	<b>0.65</b>	<b>0.78</b>	-0.12	<b>0.96</b>	-0.22	0.00	-0.34	-0.48	0.36	1.00

### 5.3. Mineralogy and Speciation

The mineralogical analysis of the Dos Carlos tailings and the comparison of mineral speciation results in profiles A and B suggest that minerals in a state of dissolution are releasing Ag<sub>metallic</sub>, Cd<sup>2+</sup>, Cu<sup>+</sup>, Ni<sup>2+</sup>, Pb<sup>2+</sup> and Zn<sup>2+</sup> into the environment. Minerals that contain Fe<sup>3+</sup>, Ag<sub>metallic</sub>, Pb(OH)<sub>3</sub>, ZnSiO<sub>3</sub> and ZnO are found in a precipitated state. This indicates that certain minerals such as quartz, calcite and kaolinite could promote an atmosphere in which low levels of acidity in the deposit can be neutralized [50]. [51] stated that the interaction of calcite with fluids that contain phosphate in slightly acidic to slightly alkaline environments results in the dissolution of calcite and promotes the precipitation of Ca-phosphate phases. In contrast, [52] determined the acidity constants of several clays and found that the kaolinite group favors the retention of heavy metals. The mobility of certain ions in the soil could also influence the mobilization of the dissolved ions; for example, Pb<sup>2+</sup> has negligible mobility in soils (or tailings) with a slightly alkaline pH, whereas Cu<sup>+</sup> and Zn<sup>2+</sup> have greater mobility in this type of soil [53].

Minerals in the tailings such as kaolinite and calcite that are found at high concentrations could neutralize the dissolution of minerals containing Cu<sup>+</sup>, Cr<sup>6+</sup>, and Zn<sup>2+</sup>. [54] reported that when the pH is almost neutral (6.5 - 7), the neutralization of calcite occurs as follows:  $\text{CaCO}_3 + 2\text{H}^+ = \text{Ca}^{2+} + \text{CO}_2 + \text{H}_2\text{O}$ . However, it is necessary to conduct other studies to determine the neutralization capacity of these minerals in this deposit.

### 5.4. Oral Exposure of Cr<sup>6+</sup>

The mean daily dose of Cr<sup>6+</sup> in groundwater (the main source of water for the population) that an inhabitant of the study area could safely consume is  $1.45 \times 10^{-4}$  mg/L/d. The carcinogenic potential of chrome ingested orally has not yet been determined. It has been determined that inhabitants of a village in China who consumed drinking water with 20 ppm of this contaminant suffered adverse health effects [55]. However, in a toxicological review of hexavalent chromium, the [56] reported that 5% of orally ingested Cr<sup>6+</sup> is absorbed by the gastrointestinal track. Once transported into the bloodstream, a portion of this concentration can remain in the body for a long time without causing apparent damage.

## 6. Conclusions

This preliminary study showed that the granulometric fraction of tailings with fine sand and a lower percentage of silts and clays favors the diffusion of oxygen (atmospheric or from water infiltration). This finding was confirmed by the fact that both profiles had moisture contents that could promote the oxidation of minerals, especially in the horizons with a slightly acidic pH. Cr<sup>6+</sup>, NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup> were found at different depths in contaminant levels. A correlation matrix showed that the behaviors of the physical and chemical parameters of the

tailings were strongly related. This indicates that the environmental conditions (diffusion of atmospheric oxygen or rainfall) affect the physico-chemical composition of the tailings. In addition, the origin of the deposit, the mining activities and the mineral separation that occur during the mining process also influence the chemical composition of the tailings. Therefore, the concentrations of the previously mentioned elements vary with pH and with the diffusion of oxygen through the pores of the tailings.

The analysis of the geochemical modeling showed that minerals containing ions characteristic of acid mine drainage were dissolved in both profiles, as it was also reported in a previous study. In contrast, minerals such as calcite and kaolinite were observed to be capable of being neutralized.

The above-mentioned findings and the concentrations of  $\text{Cr}^{6+}$  measured in the leachates from the tailings suggest that the oral ingestion of this  $\text{Cr}^{6+}$  mixed with groundwater should not cause adverse health effects. However, it is necessary to investigate the mean daily dose that the population currently ingests through inhalation. Therefore, the infiltration of leachates of the tailings into the groundwater and the wind-borne transport of contaminants from the tailings' deposit present a potential risk of contamination to the environment and human population. However, a detailed chemical and atmospheric analysis is necessary to definitively confirm whether this deposit is the source of short-term contamination of the environment and the health of the population.

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