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Copper mining in the eastern Amazon: an environmental perspective on potentially toxic elements

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Abstract Mining activity is of great economic and social importance; however, volumes of metallic ore tailings rich in potentially toxic elements (PTEs) may be produced. In this context, managing this environmental liability and assessing soil quality in areas close to mining activities are fundamental. This study aimed to compare the concentrations of PTEs—arsenic (As), barium (Ba), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), molybdenum (Mo), nickel (Ni), lead (Pb) and zinc

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E. S. de Souza e-mail: edna.souza@unifesspa.edu.br of Co, Cu and Ni in the tailings area exceeded the prevention values, soil quality reference values and average upper continental crust concentrations; however, the tailings area was considered unpolluted according to PLI and RI and presented a low potential ecological risk. The high concentrations of PTEs are associated with the geological properties of the area, and the presence of PTEs-rich minerals supports these results. For the urban and pasture areas, none of the 11 PTEs analyzed exceeded the prevention values established by the Brazilian National Environment Council.

Keywords Soil pollution · Environmental safety · Contamination factor · Ecological risk · Geoaccumulation

Introduction

Brazil is one of the largest ore exporters in the world. This activity is of fundamental economic importance because it constitutes approximately 16.8% of Brazil's industrial GDP, generating approximately 195,000 direct jobs and more than 2 million indirect jobs (IBRAM, 2019). Cu contributes 9% to the total export of ores, with the eastern Amazon being the largest contributor to Cu production (IBRAM, 2019). Despite its economic and social importance, mining can have adverse impacts on the areas influenced by these activities, including impacts arising from the movement and removal of large volumes of soil/rocks and the production of tailings/mining waste (Zhao et al., 2013). These materials may contain potentially toxic elements (PTEs) that may be toxic to living organisms, depending on the nature of the exploited ores. Thus, the management of these environmental liabilities is a critical and necessary activity (Li et al., 2018).

High levels of PTEs, including As, Ba, Cd, Cu, Hg, Pb and Zn, can cause concern. Therefore, environmental studies are important; such studies aim to highlight anthropic areas with high levels of these elements and propose measures to mitigate the corresponding problems (Birani et al., 2015; Souza et al., 2019). Soil fertility provides information beyond the total PTEs concentration by indicating the actual plant-available portion of micro- and macronutrients and can be used in combination with soil texture analysis to estimate environmental risk or, in situations related to the need to grow plants, whether to revegetate or produce food.

The total levels of the PTEs present in a studied area are usually determined in the initial stages of characterization; these levels are then compared with reference values established by regulatory agencies and scientific studies (Shah et al., 2012). There are several classes of reference values that consider the intended use, exposure levels and acceptable risks. Among them, the following guidance values for soil quality were established by the Brazilian National Environment Council (CONAMA, 2009): quality reference value, prevention value and intervention value. Additionally, several environmental indices can be used to more fully characterize studied areas, such as the contamination factor (CF), degree of contamination (Cdeg), potential ecological risk index (RI), geoaccumulation index (Igeo) and pollution load index (PLI). These indices represent a reliable strategy for a broader evaluation of soil PTEs levels (Gao et al., 2013; Zhuang & Gao, 2014). In addition, performing studies aimed at characterizing accumulated mining substrates, such as tailings, is a good strategy for identifying the limitations and qualities of these substrates to form more effective guidelines for rehabilitation in mined areas (Gastauer et al., 2018).

Keeping the above points in mind, the present study aimed to identify the PTEs levels in four different environments (tailings, native land, urban land and pasture) located in the proximity of a Cu mining complex in the eastern Amazon. These areas are located in the Carajás Mineral Province, which is characterized by several mineral deposits (Teixeira et al., 2007). Additionally, fertility analysis of the tailings was performed to establish future revegetation strategies for Cu mining tailings.

Materials and methods

Location and characterization of the study area

The studied areas, influenced by a Cu mine, are located in the municipality of Canaã dos Carajás in the state of Pará, Brazil. This mining complex is used for the extraction and processing of Cu ore. The tailings produced as a result of these operations are stored in a tailings dam, which was one of the sample collection areas for this study; the other sample collection areas were pastures, urban areas and native areas with vegetation typical of the region and without human management (Fig. 1).

Collection, preparation and chemical analysis of the samples

Nine simple sediment samples were collected from the material deposited in the Cu mine tailings dam. In addition, soil samples were obtained: three from urban areas, four from native areas and three from pastures close to the Cu mining complex. The samples were collected at a depth of 0–20 cm, air-dried, sieved to 2 mm and sent to the Brazilian Laboratory of Agricultural Analysis (LABRAS).

For the determination of the total concentrations, the samples were finely ground so that they could pass through a nylon sieve of 0.106 mm. Next, the samples were digested in nitric acid and hydrochloric acid in a microwave oven according to EPA method 3051A (USEPA, 1998). The concentrations of As, Ba, Cd, Co, Cu, Cr, Mo, Hg, Ni, Pb and Zn in the extracts were quantified using inductively coupled plasma mass spectrometry (ICP-MS). All samples were analyzed in triplicate; for each set, a blank sample and a certified reference material sample (OREAS 905) were included. The obtained recovery rate ranged from 94 to 98%.

The fertility parameters (pH, organic matter (OM) content and phosphorus (P), potassium (K), boron (B), Zn, iron (Fe), manganese (Mn) and Cu concentrations) and sand, silt and clay contents of the study areas were determined according to the Brazilian Agricultural Research Corporation (Embrapa, 1997).

Determination of environmental indices

The levels of PTEs were compared with different reference values: the prevention value (CONAMA, 2009), the soil quality reference value (Fernandes



Fig. 1 Map of soil sampling areas and mining tailings

et al., 2018; Sahoo et al., 2019), the global average soil concentration (Kabata-Pendias & Mukherjee, 2007) and the average upper continental crust concentration (Rudinick & Gao, 2014). Furthermore, the contamination factor (CF, Eq. 1), degree of contamination (Cdeg, Eq. 2) and potential ecological risk index (RI, Eq. 3) were calculated according to Hakanson (1980), the pollution load index (PLI, Eq. 4) was calculated according to Tomlinson (1980) and the geoaccumulation index (Igeo, Eq. 5) was calculated according to Muller (1969):

$$CF = \frac{C_{\text{metal}}}{C_{\text{control}}} \tag{1}$$

$$C_{\text{deg}} = \sum_{i=1}^{n} CF \tag{2}$$

$$RI = \sum_{i=1}^{n} (Tr \times CF)$$
(3)

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \cdots \times CF_n}$$
(4)

$$I_{\text{geo}} = \log_2 \left(\frac{C_{\text{metal}}}{1.5 \times C_{\text{control}}} \right)$$
(5)

where C_{metal} is the concentration of the metal in the sampled area, $C_{control}$ is the concentration of the metal in a reference soil, *n* refers to the number of PTEs analyzed and Tr is the toxic response of the PTEs Cd, Cr, Cu, Hg, Ni, Pb and Zn (Hakanson, 1980), Ba (Yang et al., 2015) and As and Co (Zheng-Qi et al., 2008).

In this study, we used $C_{control}$ values established for the state of Pará for As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb and Zn (Fernandes et al., 2018) and Co (Fadigas et al., 2006) and $C_{control}$ values established for the Mineral Province of Carajás for Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb and Zn (De Lima et al., 2020) and As and Hg (Salomão et al., 2019).

Statistical analysis

The data were subjected to analysis of variance (ANOVA), and the means were compared by the Scott–Knott test at 5% probability. Principal component analysis (PCA) was performed using Statistic 14.1 software to evaluate the relationship between PTEs and soil attributes in different forms of land use.

All the variables used in the PCA, except pH, were log-transformed to obtain a normal distribution.

Results

Total concentrations of potentially toxic elements

The total levels of PTEs in the areas with different forms of land use, as well as the reference values used for comparison, are shown in Fig. 2. The As and Cd levels showed no significant differences between the different forms of land use (p > 0.05). The levels of these PTEs were below the reference quality values for the state of Pará and the prevention values established by the Brazilian National Environment Council (CONAMA, 2009). The total concentrations of Co, Cr, Mo and Ni were higher (p < 0.05) in the tailings than in the native, pasture and urban areas. The Cu concentration in the tailings was statistically (p < 0.05) similar to that in the native areas and higher than that in the other studied areas.

The concentrations of Co, Cr, Mo and Ni in the tailings were higher than the quality reference values for the state of Pará and of Co and Ni higher than the Brazilian prevention values (Fernandes et al., 2018; CONAMA, 2009). The levels of Ba, Pb and Zn in the native area were statistically higher (p < 0.05) than those in the other areas studied. The Ba, Pb and Zn levels exceeded the quality reference values of the state of Pará (Fernandes et al., 2018). It is important to note that the levels of analyzed PTEs in the urban and pasture areas were lower than the prevention values proposed by the Brazilian National Environment Council (CONAMA, 2009), making the use of these areas for urbanization and agriculture permissible.

Levels of contamination of potentially toxic elements

The values of CF, Igeo, Cdeg, PLI and RI calculated according to the $C_{control}$ values established for the state of Pará are shown in Table 1, and those established according to the $C_{control}$ values established for the Carajás Mineral Province are shown in Table 2.

The CFs presented in Table 1 show high or very high contamination of Co, Cu, Mo and Ni in the tailings, as well as of Cu, Mo, Ni and Pb in the native soils. The CFs in urban and pasture areas were



Fig. 2 Total PTEs concentrations in the study areas, soil prevention values, soil quality reference values, global average soil concentrations and average upper continental crust concentrations

classified as low, except that Mo in both areas was classified as highly contaminated, and Cu in both areas, Ni in urban areas and Pb in pasture areas were classified as moderate (Table 1). The CFs presented in Table 2 show different results, where only Cu and Ni for tailings areas and Cu and Pb for native areas presented values classified as moderate.

For the Igeo results presented in Table 1, Cu in native and tailings areas had values in the range of extreme contamination, and only Mo and Ni in the tailings area were in the range of extreme contamination. Different results are observed in Table 2, where values above 1 in the moderately contaminated range were obtained for Ni in the tailings area and values in the practically uncontaminated range were obtained for Cu in the tailings and native area and Pb in the native area.

Based on the Cdeg results presented in Table 1, the tailings were the most contaminated, with a value twice that of the native soil. The secondary Cdeg

Area	Contamination factor—CF										
_	As	Ba	Cd	Со	Cr	Cu	Hg	Мо	Ni	Pb	Zn
Tailings	0.47	1.36	0.02	3.60	1.53	31.28	0.02	53.56	30.93	0.99	0.60
Native	0.56	2.10	0.04	0.74	0.18	30.80	0.15	12.28	3.11	7.41	1.69
Urban	0.23	0.45	0.03	0.15	0.72	1.98	0.08	5.11	1.68	0.88	0.76
Pasture	0.22	0.90	0.04	0.13	0.74	1.45	0.13	3.89	0.51	1.16	0.70
	Geoaccumu	ilation inde	x—Igeo								
Tailings	- 1.67	- 0.14	- 6.16	1.26	0.03	4.38	- 6.08	5.16	4.37	- 0.60	- 1.33
Native	- 1.43	0.48	- 5.38	- 1.01	- 3.09	4.36	- 3.32	3.03	1.05	2.30	0.17
Urban	- 2,70	- 1.74	- 5.49	- 3.32	- 1.06	0.40	- 4.20	1.77	0.16	- 0.77	- 0.98
Pasture	- 2.78	- 0.74	- 5.20	- 3.49	- 1.02	- 0.04	- 3.57	1.37	- 1.56	- 0.37 -	- 1,10
					Degree o	f contamin	ation—Cde	g			
Tailings					124.36						
Native					59.05						
Urban					12.07						
Pasture					9.87						
					Pollution	load index-	—PLI				
Tailings					1.43						
Native					1.26						
Urban					0.48						
Pasture					0.47						
					Potential	ecological	risk index–	–RI			
Tailings					346.59						
Native					229.20						
Urban					33.11						
Pasture					28.70						

Table 1 Environmental indices for the studied areas according to C_{control} values established for the state of Pará

The classification levels for each variable are defined as follows: CF: < 1 (low), 1–3 (moderate), 3–6 (high) and > 6 (very high); Cdeg: < 8 (low), 8–16 (moderate), 16–32 (high) and > 32 (very high); RI: < 150 (low), 150–300 (moderate), 300–600 (high) and > 600 (very high), established by Hankanson (1980); PLI: > 1 (polluted) and < 1 (unpolluted), established by Tomlinsom (1980); and Igeo: < 0 (uncontaminated), 0–1 (practically uncontaminated), 1–2 (moderately contaminated), 2–3 (moderately to heavily contaminated), 3–4 (heavily contaminated), 4–5 (heavily to extremely contaminated) and > 5 (extremely contaminated), established by Müller (1969)

pasture areas are smaller than the other study areas, i.e., 14 times smaller than the tailings area and seven times smaller than the native area. In contrast, the Cdeg values in Table 2 also show that the tailings have the highest value, corresponding to moderate contamination, in addition to presenting a value very close to that of the native area.

According to the PLI results in Table 1, the tailings area was the most polluted, followed by the native

area, but it should be noted that the native area is naturally enriched in certain elements. The urban and pasture areas were classified as unpolluted. According to the PLI values in Table 2, all areas are unpolluted.

Regarding the RI values in Table 1, the highest values were those for the tailings area (classified as high) and the native area (classified as moderate). All of the values in Table 2 are classified as low.

Area	Contamination factor—CF											
	As	Ba	Cd	Co	Cr	Cu	Hg	Мо	Ni	Pb	Zn	
Tailings	0.67	0.17	0.001	0.76	0.25	2.06	0.08	0.74	3.04	0.32	0.14	
Native	0.79	0.27	0.001	0.16	0.03	2.03	0.56	0.17	0.31	2.39	0.40	
Urban	0.33	0.06	0.001	0.03	0.12	0.13	0.31	0.07	0.16	0.28	0.18	
Pasture	0.31	0.11	0.002	0.03	0.12	0.10	0.47	0.05	0.05	0.37	0.17	
	Geoaccu	mulation in	dex—Igeo									
Tailings	- 1.17	- 3.11	- 10.82	- 0.98	- 2.59	0.46	- 4.17	- 1.02	1.02	- 2.24	- 3.41	
Native	- 0.92	- 2.49	- 10.03	- 3.25	- 5.71	0.44	- 1.42	- 3.14	- 2.30	0.67	- 1.91	
Urban	- 2.19	- 4.71	- 10.15	- 5.56	- 3.68	- 3.52	- 2.30	- 4.41	- 3.19	- 2.40	- 3.06	
Pasture	- 2.28	- 3.71	- 9.86	- 5.73	- 3.63	- 3.97	- 1.67	- 4.80	- 4.91	- 2.01	- 3.18	
					Degree of	of contamin	ation—Cde	g				
Tailings					8.24							
Native					7.10							
Urban					1.67							
Pasture					1.79							
					Pollution	load index	—PLI					
Tailings					0.26							
Native					0.23							
Urban					0.09							
Pasture					0.08							
				Potential ecological risk index-RI								
Tailings					41.92							
Native					55.85							
Urban					19.12							
Pasture					25.41							

Table 2 Environmental indices for the studied areas according to C_{control} values established for the Carajás Mineral Province

The classification levels for each variable are defined as follows: CF: < 1 (low), 1–3 (moderate), 3–6 (high) and > 6 (very high); Cdeg: < 8 (low), 8–16 (moderate), 16–32 (high) and > 32 (very high); RI: < 150 (low), 150–300 (moderate), 300–600 (high) and > 600 (very high), established by Hankanson (1980); PLI: > 1 (polluted) and < 1 (unpolluted), established by Tomlinsom (1980); and Igeo: < 0 (uncontaminated), 0–1 (practically uncontaminated), 1–2 (moderately contaminated), 2–3 (moderately to heavily contaminated), 3–4 (heavily contaminated), 4–5 (heavily to extremely contaminated) and > 5 (extremely contaminated), established by Müller (1969)

Physical and chemical attributes

PCA was used to evaluate the PTEs and the physical and chemical attributes of the soil and tailings, and two principal components were identified, which explained 68.5% of the total variation (Fig. 3 and Table 3). The first component (PC1), which explained 49.7% of the total variation, was moderately and positively correlated with Cr (0.66) and pH (0.75); strongly and positively correlated with Co (0.90), Mo (0.86), Ni (0.95), P (0.79) K (0.91) and sand (0.84); negatively and strongly correlated with Mg (- 0.88), OM (- 0.83), B (- 0.80), Fe_a (- 0.89) and Mn_a (- 0.95); and negatively and moderately correlated with Zn_a (- 0.64), Ca (- 0.65) and Cd (- 0.72).

PC2 showed strong and positive factor loadings with Ba (0.83) and Cu_a (0.89) and moderate loadings with As (0.65), Cu (0.72), Pb (0.61), S (0.58) and Zn (0.71). Factor loadings > 0.75 were considered strong, those between 0.75 and 0.5 were considered moderate, and those between 0.5 and 0.3 were considered weak.

It is also noteworthy that the tailings samples were aligned with Cu, Co, Mo, P, Ni, Cr, K, As, Hg, sand and pH and therefore demonstrated alignment and affinity with PC1; in contrast, samples from native areas were aligned with S, Zn and Pb and therefore demonstrated alignment and affinity with PC2 (Fig. 3).

The physical and chemical attributes of the studied areas exhibited significant differences (p < 0.05) (Table 4). In general, all areas had a predominance of sand in relation to clay and silt; the tailings area had the highest sand content, 84%, while the pasture area had the lowest sand content, 59.3%. Clay represented less than 40% in all areas, and the pasture area had the highest clay level, 37%. In all areas, the silt content represented less than 10% of the total, and the urban and native areas showed the highest proportions, 8.3 and 6.3%, respectively.

The pH values of the areas did not differ statistically; the values ranged from 5.8 in the pasture areas to 6.9 in the tailings area and were classified as slightly acidic (Table 3). The OM content of the tailings area



Fig. 3 Principal component analysis of potentially toxic elements in different forms of land use near a copper mine in southeastern Pará state, Brazil

 Table 3
 Matrix of components for PTE concentrations in soils and copper mine tailings

Variables	Matrix of components					
	PC1	PC2				
As	0.40	0.65				
Ba	0.26	0.83				
Cd	- 0.72	0.24				
Co	0.90	0.40				
Cr	0.66	- 0.15				
Cu	0.65	0.72				
Hg	0.23	0.09				
Мо	0.86	0.38				
Ni	0.95	0.20				
Pb	- 0.38	0.61				
Zn	- 0.39	0.71				
pH	0.75	-0.08				
Р	0.79	0.36				
K	0.91	0.22				
S	- 0.16	0.58				
Ca	- 0.65	0.29				
Mg	-0.88	0.15				
OM	- 0.83	0.46				
B _a	-0.80	0.34				
Cu _a	0.06	0.89				
Fe _a	- 0.89	0.14				
Mn _a	- 0.95	0.20				
Zn _a	- 0.64	0.33				
Clay	- 0.93	- 0.06				
Silt	- 0.48	0.18				
Sand	0.84	0.09				
Eigenvalue	12.9	4.9				
% Total variation	49.7	18.8				
% Cumulative	49.7	68.5				

Note: The pH, P, K, S, Ca, Mg, OM (organic matter), Ba, Cu_a, Fe_a, Mn_a, and Zn_a are fertility parameters of soils

was very low (1.05%), and that of the native area was moderate (3.4%). The native area presented a high P concentration (89.5 mg dm⁻³), which was five times higher than that found in the state of Pará in Gleysols, the class of soil with the highest level of P in the state (Souza et al., 2018). The tailings area presented eight times more P than the native area. The pasture area had the lowest P concentration among the studied areas, but the value was still higher than the average levels found in sandy-textured soils of the state of Pará (Souza et al., 2018). The K levels were similar to the P levels, with a higher level in the tailings area and a lower level in the pasture area. The K concentration in the tailings area was 3.8 times higher than that in the native area and 6.2 times higher than that in the pasture area. Additionally, the average K concentration in the pasture area was higher than the average found in soils from the state of Pará (Souza et al., 2018). Lower levels of Ca and Mg were present in the tailings area than in the other areas; the pasture areas presented the highest Ca levels, while the highest Mg level was found in the urban area. The native area had the lowest levels of exchangeable cations among the areas studied.

The available levels of the micronutrients Cu, Fe, Mn and Zn were significantly higher (p < 0.05) in the native area than in the tailings area. The levels of Fe and Mn in the native and pasture areas were similar. The urban area generally had low micronutrient concentrations. The native area had 13 times more available Cu than the tailings area and 74 times more Cu than the pasture area.

Discussion

Potentially toxic elements of concern in mining tailings

A comparison of the PTEs concentrations in mining tailings with the values in soils in areas close to the mine is essential for assessing environmental risks and establishing policies for the protection of the environment and human health (Da Silva et al., 2015) because high levels of PTEs in the soil can be absorbed by food crops and can contaminate groundwater, which may affect food safety and water quality (Dung et al., 2013; Esmaeili et al., 2014). In the present study, the sampled areas are very close to the copper extraction complex, and the local land use is constantly changing due to mining activity, which leads to environmental concerns, mainly regarding soil quality.

Although high levels of PTEs are frequently associated with mining and several studies have noted contamination of PTEs in areas adjacent to mining activities (Afonso et al., 2020; Giri et al., 2017; Pereira et al., 2020), in this study, the pasture and urban areas, which are adjacent to the mining area, did not present such contamination. This finding can be explained by

 Table 4
 Chemical and physical attributes of Cu tailings from the Sossego Dam, native areas, urban areas and pasture areas and available element concentrations

Parameters	Tailings	Native	Urban	Pasture	CV (%)
pH in water	$6.9 \pm 0.3a$	$6.3 \pm 0.5a$	$6.5\pm0.2a$	5.8 ± 0.04 b	2.5
OM (%)	$1.1 \pm 0.4 \mathrm{b}$	$3.4 \pm 0.8a$	$1.6 \pm 1.1 \mathrm{b}$	$2.9\pm0.9a$	15.9
$P (mg dm^{-3})$	$715.7 \pm 392a$	$89.5 \pm 13.8b$	$71.6 \pm 90.4b$	$26.9 \pm 2.3b$	40.3
K (mg dm^{-3})	$312.9 \pm 99.7a$	$82.7\pm26.7b$	$62.7 \pm 19.1 b$	$50.2 \pm 4b$	17.1
S (mg dm^{-3})	$5.0\pm24.7a$	9.3 ± 1.6a	$5.9\pm2.5a$	6.2 ± 0.8 a	20.6
Ca $(\text{cmol}_{c} \text{ dm}^{-3})$	$2.3 \pm 0.3a$	$3.6\pm0.5a$	$3.2 \pm 1.9a$	$4.5 \pm 1.8a$	15.9
Mg (cmol _c dm ^{-3})	$0.1\pm0.04\mathrm{b}$	$0.7\pm0.1a$	$0.9\pm0.3a$	$0.6 \pm 0.4a$	13
CEC ($\text{cmol}_{c} \text{ dm}^{-3}$)	$4.8\pm0.01a$	$7.5\pm0.6a$	$5.6\pm0.5a$	$6.9 \pm 0.1a$	13
B (mg dm^{-3})	$0.1 \pm 0.02c$	$0.2\pm0.01\mathrm{b}$	$0.1 \pm 0.1c$	$0.3 \pm 0.2a$	5.8
$Zn (mg dm^{-3})$	$0.5 \pm 1.7c$	$2.8\pm3.8a$	$1.2 \pm 2.9 \mathrm{b}$	1.3 ± 0.04 b	9.5
Fe (mg dm^{-3})	$15.4 \pm 11.1c$	$103 \pm 42a$	$57 \pm 22.5b$	$122.5\pm4.5a$	13.8
$Mn (mg dm^{-3})$	$2.2 \pm 0.8c$	$24.3 \pm 7.4 \mathrm{a}$	$10.4 \pm 2.3b$	$23.4\pm8.9a$	20
Cu (mg dm^{-3})	$14.1 \pm 9.2b$	$184.2 \pm 54.8a$	$6.1 \pm 3.4c$	$2.5\pm0.5c$	11.6
Silt (%)	$2.5\pm31.4b$	$6.3 \pm 28a$	$8.3 \pm 31.1a$	$3.6\pm10.2b$	18.2
Clay (%)	$12.3 \pm 18.4c$	$28.3\pm41.5b$	$29.5\pm54b$	$37 \pm 20.4a$	7.9
Sand (%)	$84.1\pm48.7a$	$65.5\pm68.5b$	$62.2\pm82.5\mathrm{b}$	$59.3\pm30.6b$	4.6

Means followed by the same letter in the row do not differ by the Scott-Knott test at 5% probability

the adequate and efficient methods used to deposit, handle and contain the tailings generated in mining, which allow environmental control of the generated waste. The conditions here thus diverge from the results found in several works in the literature related to mining areas with inadequate waste handling.

The concentrations of elements As, Ba, Cd, Hg, Pb and Zn in the tailings area, urban areas and pasture areas were similar to or lower than the concentrations found in the native area, which suggests that mining does not contribute to the enrichment of these PTEs in this region and that there are no environmental concerns regarding the use of these areas as pasture despite proximity to a Cu mine. The results indicate that the concentrations of these PTEs are related to the source materials in the region (Licina et al., 2017), which provides further evidence of their geogenic origin (Sahoo et al., 2019; Salomão et al., 2019).

Notably, even though the levels of Ba, Pb and Zn found in the native area were higher than those found in the other study areas, except for Pb, they were still below the reference values established for the state of Pará (Fernandes et al., 2018) and below the world average. This result indicates a low risk of environmental contamination and to human health. Even the highest values of these elements found in native areas are characteristic of soils in the Carajás Mineral Province, which is due to the high mineralogical diversity (quartz, kaolinite, hematite, biotite, rutile, magnetite, goethite, gypsum and vivianite, bernilite, chlorocalcite, nitrocalcite, muscovite and halloysite) (De Lima et al., 2020), with mafic and ultramafic rocky substrates presenting soils naturally rich in PTEs (Berni et al., 2014; Schaefer et al., 2015).

Additionally, the As and Cd levels did not differ statistically among the studied areas, and the levels of these elements are also apparently related to the lithology of the region, as indicated in regional geochemical studies developed in the Carajás region (Sahoo et al., 2019; Salomão et al., 2019). These PTEs are commonly found in Cu mining areas, and they are usually present as sulfides containing As and Cd, including arsenopyrite (FeAsS) and cobaltite (CoAsS) (Wang et al., 2016). The levels of these PTEs are below the reference values for the state of Pará and the prevention values of CONAMA (2009), which suggests a low risk of human and ecological contamination and geogenic origin (Souza et al., 2018). Co and Ni levels exceeded the Brazilian prevention values in the tailings, and Co concentrations were lower and Ni were greater than the intervention values. The concentrations of elements below the intervention values suggest minimum risks for human and ecological health and indicate that the soil remains functional. Conversely, metal levels higher than intervention values suggest direct or indirect potential risks to human health (CONAMA, 2009). Prolonged exposure to Ni in humans can trigger adverse health effects such as contact dermatitis, cardiovascular disease, asthma, lung fibrosis and respiratory system cancer (Genchi, et al., 2020).

The high levels of these PTEs in the tailings may be related to the processing of Cu by flotation, which involves electrochemical reactions where insoluble metal compounds are formed, sulfide minerals undergo intense oxidation, and Cu and several other PTEs, such as Co, Cr, Ni and Mo, are enriched (Silva, 2011). Another factor explaining the high levels of these elements in a Cu mining area is the fact that siderophores are found in native ores and host rocks in Cu mining areas (Wang et al., 2016). Corroborating the high levels of elements found in the present study, Chileshe et al. (2019) reported high levels of elements in copper mining tailings in Zambia: 12,000 mg kg⁻¹ Cu, 300 mg kg⁻¹ Co, 20 mg kg⁻¹ Ni and 15 mg kg⁻¹ Cr. In turn, metal concentrations lower than those found in the present study were reported by Afonso et al. (2020) for Cu mining tailings in southern Brazil, where the Cu concentration was 259.7 mg kg⁻¹, the Cr concentration was 15.3 mg kg^{-1} and the Ni concentration was 9.0 mg kg⁻¹. Differences in PTEs concentrations in different mining areas indicate a strong relationship between PTEs concentrations and the geological formation of each region (Chileshe et al., 2019).

The similar Cu concentrations of the native and tailings areas suggest that the beneficiation process was efficient and allowed the extraction of more than 90% of the Cu present in the ore, reducing the risks of environmental contamination. In a Cu mine in Zambia, the tailings had a mean Cu concentration of 12,237.3 mg kg⁻¹, while a forest area had a mean Cu concentration of 50.5 mg kg⁻¹ (Chileshe et al., 2019). The authors attributed the Cu concentration in the tailings to not only the mineralogical composition but also the ore extraction and processing method, which presented an efficiency of only 40%. Thus, the similar

Cu concentrations of the tailings and native areas indicate that mining processes are not causing enrichment of this metal and that the soils of the region are naturally rich in this element and other elements. Such natural abundance is common in metalliferous regions (Chileshe et al., 2019) and is clearly demonstrated by metallogenic (Moreto et al., 2015) and geochemical studies (Sahoo et al., 2019; Salomão et al., 2019) that provide evidence for the existence of two large copper mineralized belts in the Carajás region; the study area is located in the southern mineralized copper belt.

It is important to note that the available Cu levels in the tailings are 13-fold smaller than those in the native area, which suggests that the tailings present a low risk of contamination. High levels of elements in the available form are worrisome because this fraction has high mobility and bioavailability and consequently presents a higher risk of environmental contamination than do other forms (Pereira et al., 2020). The lower available Cu concentration in the tailings may also be associated with higher losses due to the solubilization of Cu mineral sources, given the low cation exchange capacity (CEC) in this area (Pereira et al., 2020).

The high available Cu concentration in the native area may also be attributed to a higher OM content and nutrient cycling (Sundaray et al., 2011). Cu has a high affinity for organic ligands due to the ease of formation of highly stable compounds between organic fractions and elements, reducing mobility (Pereira et al., 2020). The ability of OM to adsorb elements and modify mobility and bioavailability is well documented in the literature (Pereira et al., 2020; Souza et al., 2019; Wu et al., 2020).

A comparison of the Co, Cu, Mo, Ni and Zn concentrations in the native area with those in the pasture area showed that the latter area had lower levels of these PTEs. As the sampled areas are close and belong to the same geological formation, the low levels of these elements, which are also nutrients for plants, suggest that the form of land use, combined with inadequate management practices, leads to nutrient losses and soil degradation (Wu et al., 2020).

Environmental indices

The results presented in Table 2 will be used as the basis for the discussion of the environmental indices of the studied areas; since these results are based on regional reference values that best represent the areas

of this study, they may better reflect the contamination conditions. In addition, the environmental indices in Table 1 overestimate the obtained results since they are based on reference values established for the state of Pará and are lower than the natural values of the areas of this study. Therefore, it is fundamentally important to use the appropriate reference values for the areas to be investigated. This difference between the values in Tables 1 and 2 demonstrates a high natural enrichment of PTEs in the region where the study area is located (De Lima et al., 2020).

None of the forms of land use in this study showed contamination by As, Ba, Cd, Co, Cr, Hg, Mo or Zn. The Igeo values for Cu and Ni for the tailings areas and Cu and Pb for the native areas indicated practically no contamination or moderate contamination. In addition, these elements were the ones that most contributed to PLI. The Pb CF in the native area was 7.5 times greater than that in the tailings area; this finding can be explained by the geological formation of the study area, which is composed of metavolcanic and sedimentary rocks rich in PTEs (Berni et al., 2014). These geochemical characteristics indicate that the high Pb values are influenced by geology (De Lima et al., 2020). The Cu and Ni CFs in the tailings area were higher than those in the native area, which may be an alert for possible environmental risks.

Despite the high concentrations of Cu, Co and Ni in Cu tailings in native areas (Fig. 2), the PLI suggests that these areas are not polluted, and the RI suggests that there is a low potential ecological risk. These results may be associated with high background values of these PTEs in the region. Nevertheless, urban and pasture areas do not present significant environmental risks in relation to the studied PTEs, as they present low values for all the established environmental indices. The low risk of pollution in these areas suggests low risks of contamination of the ecosystem and human health. The high natural concentrations of PTEs in the native areas suggest the need to adopt management practices to reduce the risk of pollution and the risks of contamination of other ecosystems (Christou et al., 2017). The form of land use is one of the main factors that influence the concentration of PTEs in the soil and the risk of human exposure; thus, the tailings areas and their surroundings must be monitored and managed to avoid additional pollution (Zhang et al., 2018).

Fertility parameters suggest concern regarding the revegetation of tailings areas

Regarding the granulometric distribution of the sampling points, all areas had a sandy texture, especially the tailings area, which presented a granulometric fraction greater than 80%, resulting from the comminution process. The predominance of sand and the low contents of clay and silt imply a low aggregation capacity, low nutrient adsorption and low water retention. Soils or sediments with a predominance of clay and silt present a relatively high CEC and the ability to retain nutrients and immobilize contaminants (Souza et al., 2018).

The concentrations of available P and K present in the tailings area are classified as very high according to Brasil, Cravo and Viegas (2020). For some environmental conditions, high concentrations of these elements may be a concern due to the risk of leaching, erosion and atmospheric dust deposition (Wang et al., 2018), which can enrich the soils, sediments and waters of the surrounding areas. For example, although P is a plant nutrient, when discharged in large quantities into water, it can lead to eutrophication (Azam et al., 2019). Therefore, even though these elements are not considered potentially toxic, measures to prevent their uncontrolled dispersion in the environment are recommended. The revegetation of areas is an alternative method of reducing the contamination level in surrounding areas (Wang et al., 2018). However, it is noteworthy that tailings areas are established in an industrial mining context and are subject to measures that minimize dispersion to the environment.

The low OM, Ca and Mg concentrations and sandy texture of the tailings can limit revegetation because these factors can result in nutrient scarcity, poor structure, a low moisture retention capacity and high erodibility (Afonso et al., 2020). These conditions can be accentuated by the intense rainfall conditions of the Amazon region, which facilitates the loss of bases (Pereira et al., 2020). Additionally, a sandy texture favors the loss of polluting elements, which can cause contamination of the water table and adjacent areas. Management of these areas requires techniques that maintain soil cover, increase OM content and reduce the availability of contaminants (Afonso et al., 2020; Zhang et al., 2018). The native area had an OM content of 34 g kg^{-1} , which is considered average according to the classification by Venegas et al. (1999). A high OM content in sandy-textured soils, as found in this region, contributes to increased CEC and nutrient availability. The high levels of micronutrients, even given the predominance of sand, indicate that OM contributes substantially to sustaining soil fertility in this region. The high levels of Cu, P and K in this area are related to both OM and mineralogy. In tropical regions, where highly weathered soils predominate, OM is the main attribute responsible for soil fertility (Souza et al., 2018).

The pasture area had the lowest values of pH and macro- and micronutrients, which suggests that the form of land use contributes to the loss of native fertility and consequent land degradation. Management practices such as the integration of livestock and fertilization may be an alternative to maintain a viable pasture system in the region.

The low CEC observed in the studied areas is consistent with the results commonly found in Amazonian soils, where predominantly dystrophic soils are highly weathered and thus have low levels of exchangeable bases (Souza et al., 2018).

The concentration of PTEs in soil is influenced by both soil genesis and soil physicochemical properties, such as pH and the clay, OM and oxide contents (Licina et al., 2017). For example, the PCA revealed a strong association between the As, Cu, Cr, Co, Mo and Ni concentrations and the sand fraction in PC1, which suggests that these PTEs are associated with lithogenic origin and have affinity and similar geochemical behavior. However, the association of these factors with sand also suggests that these elements are associated with the primary minerals found in this fraction (Fernandes et al., 2018). These results agree with those of Licina et al., (2017), who verified a correlation between Ni and Cr and between Fe and Co, a positive correlation between sand and PTEs and a negative correlation between PTEs and clay.

The association of the available concentrations of Mn, B, Zn and Fe with clay and OM indicates that pedogenesis contributed to the origin of these PTEs. OM and clay are the main factors responsible for soil CEC because they have a high surface area and a high capacity to retain nutrients by adsorption on the surface of colloids (Fernandes et al., 2018; Pereira et al., 2020).

Soil attributes such as pH, OM and grain size distribution influence the mobility and bioavailability of PTEs in soils (Gao et al., 2013). The tailings, for example, had the lowest available concentrations of Mn, B, Zn and Fe combined with the highest sand percentage; in contrast, the native and pasture areas were characterized by high clay contents, high levels of OM and higher available concentrations of PTEs.

The inverse behavior of pH and the available elements Mn, B, Zn, Fe and Mg indicates that soil acidity can reduce the concentrations of these PTEs and favor losses by leaching. The positive association between the total concentrations of PTEs and pH is related to the near-neutral pH found in the tailings area, which reduces the solubility of elements. Thus, immobilization of elements occurs on the surface of colloids at neutral or alkaline pH, while solubilization occurs at acidic pH.

Conclusions

The levels of Co, Cu and Ni in the tailings area exceeded the prevention values, soil quality reference values and average upper continental crust concentrations; however, the tailings area was considered unpolluted according to the PLI and RI and presented a low potential ecological risk since the regional background values for the studied areas indicate soils that have high natural concentrations of PTEs and are therefore enriched by geological influence.

The highest concentrations of PTEs found in the native areas reflect the natural enrichment of the soils in the region. Thus, moderate environmental values were found for more enriched elements, and low values were found for elements present in lower concentrations.

For the urban and pasture areas, none of the 11 PTEs analyzed exceeded the prevention values established by the Brazilian National Environment Council, and for these areas, all the environmental indices corresponded with the lowest classification values, demonstrating that there are no contamination and little natural enrichment of these areas by PTEs.

The chemical and granulometric characteristics of the tailings area differ the most from those of the other study areas, mainly due to the high concentrations of PTEs, phosphorus and potassium, which require future management if a vegetation cover is to be established in this area.

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Declarations

Conflicts of interest Not applicable.

Human or animal rights Not applicable

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