



Environmental challenges related to cyanidation in Central American gold mining; the Remance mine (Panama)

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ABSTRACT

Mine tailings are a potential source of environmental pollution because they typically contain potentially toxic elements (PTEs) and the residue of chemical compounds used during extraction processes. The Remance gold mine (NW Panama) is a decommissioned mine with mining activity records dating from the 1800s and several periods of abandonment. Very little remediation work has been performed, and waste is exposed to climatic conditions. This study aimed to evaluate the PTEs and cyanide contents in mine waste after mining operations ceased some 20 years ago, and to evaluate the degree of pollution and the environmental risks they pose with the use of the Pollution Load Index (PLI) and the Ecological Risk Index (RI). Although the total cyanide (T-CN) concentration (1.4–1.9 mg kg⁻¹) found in most of the study area falls within the limits of gold mining tailing values for American sites (1.5–23 mg kg⁻¹), it is worth noting that the values of the tailings of the last used mining operation exceed it (25.2–518 mg kg⁻¹) and persist at the site. The PLI and RI suggest that the tailings from the mine and mine gallery sediments represent a source of pollution for soils and surrounding areas given their high content of PTEs (As, Cu, Sb, Hg) and T-CN, which pose serious ecological risks for biota. Therefore, it is necessary to draw up a remediation plan for this area.

1. Introduction

Gold mining is perhaps the most widespread and most practiced mining type in the world, performed by large mining companies and small groups of miners. These mining operations work with very low mineral/waste ratios and produce large volumes of waste after concentration processes. These concentration processes often employ compounds with a high toxic environment potential, such as cyanide compounds or Hg, which are also used inefficiently in Indonesia, Colombia, Brazil, Ecuador, Ghana, among other countries

(Velásquez-López et al., 2011; Seccatore et al., 2014; García et al., 2015; Clifford, 2017). Gold mining production depends on mining technology and efficiency, and the highest levels are mined in South America, with intermediate levels in Asia and Central America, and low levels in Africa (Seccatore et al., 2014). The degree of pollution produced by a gold mining operation depends on the nature and composition of the extracted minerals (Kyle et al., 2011, 2012; Luque-Almagro et al., 2016), the extraction process (Higuera et al., 2004; Olobatoke and Mathuthu, 2016) and the taken remediation measures, which are often inadequate or insufficient (O'Faircheallaigh and Corbett, 2016). Gold appears in

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ores with other elements like metals and metalloids, which can be an environmental concern when mining operations spread them in the environment (Drace et al., 2016; Pavoni et al., 2018; García-Lorenzo et al., 2019; Elmayer et al., 2020).

Mine tailings are focal pollution points for surroundings, and often contain high levels of metals and metalloids, e.g., Fe, Cu, Zn, As, Sb, Cd, Hg, Pb (Eisler and Wiemeyer, 2004; Shaw et al., 2006; Donato et al., 2007; Veiga et al., 2014; García-Lorenzo et al., 2019; BasriSakakibara and Sera, 2020). They are generally considered to be potentially toxic elements (PTEs) (Hooda, 2010), and environmental factors like rain, wind and erosion lead to the mobilisation of metals and waste (Meeussen et al., 1992; Hilson and Murck, 2001; García-Giménez and Jiménez-Ballesta, 2017; Ramappa and Muniswamy, 2018). Soil and sediment properties, such as pH, texture, clay minerals, Fe and Al oxy-hydroxides, among others, play a crucial role in the geochemical mobility of toxic metals in areas affected by gold mining (Palansooriya et al., 2020). This problem is combined with the fact that most ore-processing companies use one of the most toxic chemical products, cyanide, to leach out the valuable elements contained in mined ore (Shaw et al., 2006; Donato et al., 2008; Ramappa and Muniswamy, 2018; Anning et al., 2019). These mines require solution-processing ponds that contain alkaline waters, and high concentrations of sodium cyanide, free cyanide and metal-cyanide complexes (Donato et al., 2008). Given their cyanide content, these water bodies are an environmental risk, which is compounded in the event of accidental spillages in rivers and streams because cyanide volatilises at a high rate and releases hydrogen cyanide to air (Brüger et al., 2018). This process potentially affects agricultural areas, plants and groundwater, and also poses a risk for human and animal health (Khan et al., 2020).

The cyanide types typically found in gold mine tailings are free cyanide (HCN, CN⁻), which is easily released, weak acid dissociable (WAD) cyanide (Cu, Ni, Zn) (these complexes are unstable and break up, and free cyanide is released into the environment) and stronger cyanide complexes (Au, Co, Fe), which do not release cyanide, but decompose slowly and are stabler than WAD cyanides in sunlight and under strongly acid conditions (Meeussen et al., 1992; Anning et al., 2019). Cyanide at low concentrations in soil can biodegrade in the presence of nitrifying bacteria, but higher concentrations caused by anthropogenic activities, such as manufacturing industries, the application of herbicides and mining operations, impact both soil and the environment (ATSDR, 2006). Cyanide is harmful for human health, even at low concentrations, and over long periods. The workers who breathe in small amounts of hydrogen cyanide, i.e., between 6 and 10 mg m⁻³, for a lengthy period can have breathing difficulties, chest pains, vomiting, changes in blood, headaches and enlarged thyroid gland. Breathing high concentrations causes death, and skin that comes into contact with hydrogen cyanide and cyanide salts can be irritated with sores (ATSDR, 2006).

In light of all this, the present study aimed to assess the implications of the contents of PTEs and total cyanide that remain in Remance gold mine tailings in Panama more than 20 years after the mining company shut down its operations. The obtained information was used to evaluate the pollution risks for the environment, which were evaluated with the use of the Pollution Load Index (PLI) and the Ecological Risk Index (RI). It was hypothesized that the remaining amounts of these pollutants still pose risks for both human health and the environment. Therefore, this study lays the basis to establish remediation plans in the area.

2. Materials and methods

2.1. Study area

The Remance mine belongs to the gold-rich strip of the Veraguas province in Panama and corresponds to an epithermal gold deposit hosted on a bed of pyroclastic rocks. The hydrothermal alteration covers an area of 10 km², and gold is found in veins as either small inclusions in pyrite and marcasite (FeS₂) or free gold disseminated within quartz,

with small amounts of accessory minerals in the form of chalcopyrite (CuFeS₂), sphalerite (ZnS), galena (PbS) and arsenopyrite (FeAsS) (Nelson and Ganoza, 1999). The geochemical surveys conducted by the company Minera Remance S.A. have revealed high levels of PTEs in the area, including Au, Ag and As, as well as localised Hg and anomalously amounts of Sb (Nelson and Ganoza, 1999). The deposit comprises a system of veins, in which the principal vein contains the biggest ore quantities, along with minor, but important, veins like Santa Rosa and Consuelo, which are subterranean and have sporadic outcrops (Nelson and Ganoza, 1999). The mine was mainly operated on a subterranean basis during intermittent periods between 1800 and 1999 by three different companies (Nelson and Ganoza, 1999), and the relatively rudimentary technology employed in the process, at least by the last company, resulted in low production levels and major environmental problems (Hughes-Ortega, 1998). The features that are still visible in the mine area are two mine galleries and one mine shaft, as well as accumulated waste, which includes mine sterile rock (dumps) and dams that, in turn, include cyanidation process waste. These waste types have been exposed to local environmental effects related to the local tropical climate for at least the last two decades.

According to the Köppen climate classification map (Dirección de Hidrometeorología de ETESA, 2007), the local climate is the Am type, a humid tropical climate with a monsoon influence, annual rainfall of >2250 mm, a dry season lasting 5 months and a lengthy rainy season. The study area presents a mountainous physiography with average slopes between 35 and 50% in the central elevated area, where the presence of loose materials from mineral processing with insufficient plant cover can increase the risk of these materials' erosion, and cause their transport to lower topographically areas during torrential rain periods.

While the company Minera Remance S.A. ran its operations between 1989 and 1999 (Nelson and Ganoza, 1999), the cyanidation process was used to extract the precious metal. It stated neutralising the sodium cyanide employed during the process with sodium hypochlorite. Over the years, three tailings ponds were utilised to contain the corresponding mine waste:

- Tailings Pond 1 (TP1): used from April 1990 to March 1992. This pond is located NW of the mining concession to one side of the so-called Veneno (poison in Spanish) stream, and stored approximately 75,000 tonnes of tailings, with a front wall measuring 27 m high (Gómez, 2008). The area has a plain relief and is now employed as a football ground
- Tailings Pond 2 (TP2): it is located NE of the mining concession at the head of the *Chitreca* stream and it stored 100,000 m³ of waste material. This pond was employed from April 1992 to October 1994, when it was ordered to be closed as a result of a wall collapsing following an extreme precipitation event during the rainy season, which led to the *Chitreca* stream being polluted with mine waste material (Gómez, 2008).
- Tailings Pond 3 (TP3): it was built as a replacement following the collapse of TP2. This pond is located in the centre of the mining concession on the eastern hill slope where the 'Principal' vein is found, and the area has a less pronounced slope than that of TP2. This pond was employed from October 1994 until operation shut down (Gómez, 2008). Unlike the previous tailing points, TP3 was not positioned directly in a natural basin and, following treatment in sedimentation ponds, the tailings wash water was discharged to the *Agustina* stream, which flows into the mid course of the *Chitreca* stream which, in turn, flows into the River Santa María (Delgado, 1994).
- Secondary Ponds SP1 and SP2: the use of these ponds is unclear in terms of the local mining and mineral processing works; SP1 is located between TP2 and TP3, and SP2 is located between TP2 and the *Paisana* stream.

All three tailings ponds, along with one of the two aforementioned mine galleries, was allowed to allow uncontrollable discharges into local streams. This circumstance led to frequent complaints being made against the company, which was accused of polluting the water in streams and, hence, the River Santa María (Gómez, 2008). There were also complaints by local inhabitants about deforestation and the pollution of soil, flora and fauna (Hughes-Ortega, 1998). Given this mechanical instability scenario of tailing points and the wet tropical climate in the area, there is a real possibility of an extreme climatic rainfall event causing tailing materials to enter the local hydrological system. Such a discharge would affect the soil and groundwater in the area and would, thus, increase pressure on the River Santa María and pose a risk for the peasant population living in the region and its surroundings, who engage in subsistence agriculture and livestock farming activities. Currently, the Panamanian government has plans to reactivate the Remance mining concession, which has led to protests by residents in the area against mining because they fear that the pollution history in the region will be repeated.

2.2. Sampling of tailings

Sampling was performed between May and June 2019 during the rainy season. The location map of the taken samples is presented in Fig. 1.

The survey included 13 samples (seven from tailing ponds, six from secondary ponds and their surroundings) and one corresponding to the sediments from the mouth of a mine gallery, which actively released water to a local stream. Samples TP1 (2) corresponded to the surface samples (0–15 cm depth) taken from two locations in this accumulation of tailings; samples TP2 (3) corresponded to a single site, but at two depths (0–15 cm and 15–30 cm) near the SE border of this pond; samples TP3 (3) corresponded to two sites, with a surface sample (0–15 cm) and

two samples at different depths (0–15 cm and 15–30 cm); samples SP1 and SP2 included one taken from the button of each site and three samples from adjacent soils: one in SP1 (0–15 cm) and two in SP2 (at two depths, 0–15 and 15–30 cm). A reference sample was taken outside the mining area at the El Naranjal community, which located approximately 4 km away from the mining area.

Samples were collected with PVC tubes, which were jacked into the materials to obtain samples. The material was placed inside a plastic bag using a plastic shovel. The sediment samples (including those taken from the bottom of secondary ponds, which were flooded during sampling) were directly collected with a plastic shovel. Each sample was stored in a hermetic plastic bag, which contained approximately 3 kg of sample, to be stored at ambient temperature.

2.3. Laboratory analysis

Samples were taken to the laboratory, dried at ambient temperature, broken up with a wooden rolling pin and passed through a 2 mm sieve (Soiltest). The physicochemical analyses were conducted on the sieved sample, particularly pH, electrical conductivity (EC) and oxidation-reduction potential (ORP), which were determined in a 1:5 suspension (w/V) (ASTM D 4972) with a multiparameter benchtop Orion Versa Star Pro device. Moisture was determined by weight loss at 105 °C in an oven (P Selecta, 2000200) and on an OHAUS Adventurer Pro AV264C balance. Organic matter (OM) was established by weight loss at 455 °C (ASTM D 2974) in a Hobersal HD150 furnace. Cationic exchange capacity (CEC) was determined by the potentiometer method (Weaver et al., 1991). Colour when wet was measured by Munsell soil charts. The sample texture classification was assigned by the Unified Soil Classification System (USCS), which is a soil classification system employed in engineering and geology to describe the texture and size of the particles of a given soil. To classify soil, it is necessary to previously perform soil

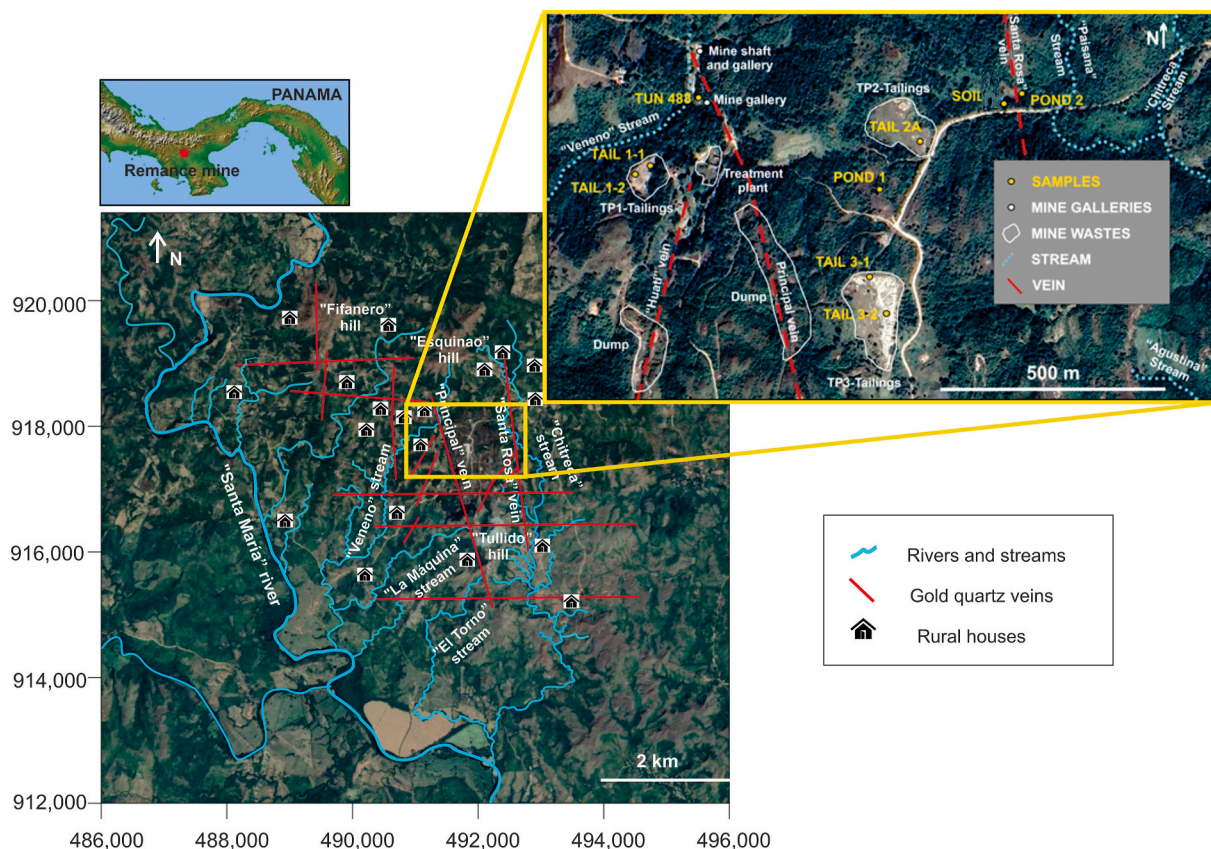


Fig. 1. Remance mine, locations of tailings, ponds and sites of samples collection.

granulometry by sieving. Then close to the Atterberg limits, the corresponding group was classified (ASTM D 2487) (ASTM, 2004). All the sieved samples were sent to Activations Laboratories Ltd. (Canada), where they were processed to determine total cyanide and PTEs. Metals were determined by partial digestion with aqua regia using a micro-processor hotbox to analyse pseudo-total concentrations (Melaku et al., 2005; Higuera et al., 2017). Extracts were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) with certified reference materials Oreas 45 d and Oreas 520, which were also digested and analysed in triplicate. Recovery percentages were 90–100%. Total cyanide was measured by NaOH leaching for 16 h in the dark. A diluted portion of the solution was analysed by a SAN Plus Segmented Flow Analyzer. For the quality control, blanks, controls and duplicates were also analysed in all the analyses.

2.4. Methods

2.4.1. Pollution Index and Pollution Load Index

A site polluted by a certain element can be assessed by the Pollution Index (PI) (Equation (1)), and the same, but with more than one element, can be determined by the Pollution Load Index (PLI) (Tomlinson et al., 1980; Pan et al., 2019) using Equation (2).

$$PI = C_{\text{soil}}/C_{\text{background}} \quad (1)$$

$$PLI = (PI_1 \times PI_2 \times \dots \times PI_n)^{1/n} \quad (2)$$

where C_{soil} and $C_{\text{background}}$ are the concentrations of the particular PTE in the soil and background samples, respectively (mg kg^{-1}). The PLI is the Pollution Load Index of several elements, and the PI is the single Pollution Index for a certain element. Table 1 presents the evaluation criteria for both the PI and PLI (Pan et al., 2019).

2.4.2. Toxicity response coefficient and the potential Ecological Risk Index

Hakanson (1980) defined RI as an index that combines environmental effects and element toxicity to consider the general ecological migration and transformation trends of these elements in soils and sediments. Pan et al. (2019) defined the toxicity response coefficient (Er) as the single potential ecological risk for a certain element (Equation (3)), which is used to obtain RI (Equation (4)):

$$Er^i = PI \times Tr^i \quad (3)$$

$$RI = Er_1 + Er_2 + \dots + Er_n \quad (4)$$

where Er^i is the single RI, the PI is the single Pollution Index for a certain element and Tr^i is the toxicity response coefficient of element i . The Er calculation is based on Hakanson's element toxicity response coefficient standards (Hakanson, 1980), which can be given as follows: Hg = 40, As = 10, Cu = 5, Zn = 1, and Sb = 7 (Wang et al., 2018). Table 1 summarises the evaluation thresholds for these indices based on the respective proposers.

2.5. Statistical analysis

Microsoft Excel spreadsheets were used to manage the results and the programme Minitab 15 was employed to analyse the statistical parameters of the analytical results. A multivariate analysis was performed by applying a factor analysis and a principal component analysis (PCA). Both were applied to search for the influence of factors, or group of

factors, using “Varimax” orthogonal rotation. The map figure was generated with the Surfer 9 software and was edited in CorelDraw 2020 licensed by the UCLM.

3. Results

3.1. Total cyanide and correlations with edaphic parameters and PTEs

Regarding cyanide and its changes over time, the Remance gold mine, where the cyanidation process was used to extract Au and other precious metals during the last mining extraction (20 years ago since mining operations shut down), presented higher total cyanide values than those reported for uncontaminated sites in America ($<0.005\text{--}0.5 \text{ mg kg}^{-1} \text{ T-CN}$) (Kjeldsen, 1999), which were similar to those reported in gold mine tailings at American sites ($1.5\text{--}23 \text{ mg kg}^{-1} \text{ T-CN}$) (Kjeldsen, 1999) in TP1, TP2, mine gallery sediments, secondary ponds and nearby soil (1.8, 1.6, 1.4, 1.9 and 1.7 mg kg^{-1} , respectively). However, TP3 had higher T-CN values ($25.2\text{--}518.0 \text{ mg kg}^{-1} \text{ T-CN}$) than these, and those reported in gold tailing 6, closed in Quebec, Canada ($4.8 \text{ mg kg}^{-1} \text{ T-CN}$) (Zagury et al., 2004). A statistical analysis was performed to seek any correlations between pollution in the area and the presence of T-CN.

The description of the taken samples, together with their colour and texture characteristics, are presented in Table 2. The colour and texture of the samples in tailings 1 (TP1), tailings 2 (TP2) and tailings 3 (TP3) were the same: yellowish brown with mainly a silty sand texture. The sediments of secondary pond 1 (SP1) and secondary pond 2 (SP2) had the same silt texture, but different colours. The soil near SP1 was silt in texture and olive brown in colour, while the soil near SP2 was sandy silt in texture. The sediment in the mine gallery differed from the rest, with a

Table 2

Description of the taken samples, Munsell colour and USCS soil classification.

| ID | Location/ Sample type | Depth (cm) | Munsell colour | Sample texture (USCS name group) |
|----------------|---------------------------|---------------|--------------------------------------|--|
| Tail 1-1 | TP1/tailing | 0–15 | Yellowish brown, 10 YR 5/6 | Silty sand |
| Tail 1-2 | TP1/tailing | 0–15 | Yellowish brown, 10 YR 5/8 | Silty sand |
| Tail 2 A | TP2/tailing | 0–15 | Yellowish brown, 10 YR 5/8 | Silty sand |
| Tail 2 B | TP2/tailing | 15–30 | Yellowish brown, 10 YR 5/8 | Silty sand |
| Tail 3-1 A | TP3/tailing | 0–15 | Yellowish brown, 10 YR 5/8 | Silty sand with gravel |
| Tail 3- 1 B | TP3/tailing | 15–30 | Yellowish brown, 10 YR 6/8 | Silty clayey sand |
| Tail 3-2 | TP3/tailing | 0–15 | Yellowish brown, 10 YR 5/8 | Silt with sand |
| Pond 1- sed | SP1/sediment | Surface | Light yellowish brown, 2.5 YR 6/4 | Silt |
| Pond 1- T | Close to SP1/ soil | 0–15 | Olive brown, 2.5 Y 4/4 | Silt |
| Pond 2- sed | SP2/sediment | Surface | Olive brown, 2.5 Y 4/4 | Silt |
| Pond 2- T A | Close to SP2/ soil | 0–15 | Dark grey, 5 Y 4/1 | Sandy silt with gravel |
| Pond 2- T B | Close to SP2/ soil | 15–30 | Dark brown, 7.5 YR 4/6 | Sandy silt |
| Tun 488 | Mine gallery/ sediment | Surface | Yellowish red, 5 YR 5/8 | High plasticity clay |

Table 1

Evaluation criteria for the Pollution Index (PI), the Pollution Load Index (PLI), the toxicity response coefficient (Er) and the Potential Ecological Risk Index (RI).

| Index | Not polluted | Slightly polluted | Moderately polluted | Considerably polluted | Seriously polluted | Extremely polluted |
|--------|--------------|-------------------|---------------------|-----------------------|--------------------|--------------------|
| PLI/PI | <1 | $1 < PLI < 2$ | $2 < PLI < 3$ | $PLI > 3$ | – | – |
| Er | <10 | <40 | $40 < Er < 80$ | $80 < Er < 160$ | $160 < Er < 320$ | $Er > 320$ |
| RI | <50 | <150 | $150 < RI < 300$ | $300 < RI < 600$ | $600 < RI < 1200$ | $RI > 1200$ |

yellowish red colour and a high plasticity clay texture.

The average and standard deviation values of the physicochemical parameters and elements of interest are found in Table 3, while all of those obtained results appear in S1. The samples taken from each tailing, the sediment samples from secondary ponds, the soil samples close to SP1 and SP2, and the mine gallery sediments were grouped. Values of pH ranged between 3.9 and 5.0. The most acidic pH corresponded to the mine gallery sediments, and the highest, to the sediments of secondary ponds. The EC ranged between 0.03 and 0.52 dS m⁻¹, and TP1 and TP2 had the lowest value, with the highest value for the mine gallery sediments. The ORP values ranged from 281.5 to 686.7 mV, with the lowest in the sediments of secondary ponds and the highest in TP3. Humidity was between 14.0% and 63.9%, with the lowest value in TP1 and the highest in the mine gallery sediments. Organic matter ranged from 0.6 to 12.9%, with the lowest value in TP2 and the highest in the mine gallery sediments. CEC went from 3.5 to 10.9 cmol kg⁻¹, with the lowest in TP3 and the highest in the mine gallery sediments.

The Fe concentration varied between 1.0 and >30.0%, and that of As was between 25.0 and 5030.0 mg kg⁻¹, with the lowest values in the sediments of secondary ponds and the highest in the mine gallery sediments. The Au concentration varied from 29.9 to >1000.0 ng g⁻¹, and that of Cu, between 10.4 and 403.0 mg kg⁻¹, with the lowest values in the soils close to the secondary ponds and the highest in the mine gallery sediments. The Zn concentration ranged from 14.5 to 153.0 mg kg⁻¹; that of Co, from 0.9 to 13.7 mg kg⁻¹; and that of V, from 14.5 to 27.0 mg kg⁻¹, with the lowest values in TP2 and the highest in the mine gallery sediments. The average Ag concentration varied between 0.7 and 5.7 mg kg⁻¹, and that of Hg, between 0.6 and 1.4 mg kg⁻¹, with the lowest concentration in the soils near secondary ponds and the highest in TP3. The average total cyanide (T-CN) concentration varied from 1.4 to 187.9 mg kg⁻¹, and that of Pb, from 1.7 to 24.9 mg kg⁻¹, with the lowest concentration in the mine gallery sediments and the highest in TP3. The average Ba concentration ranged between 55.4 and 514.5 mg kg⁻¹, and that of Cr, between 2.0 and 6.5 mg kg⁻¹, with a low concentration in the mine gallery sediments and the highest in TP2. The Sb range went from 0.9 to 20.7 mg kg⁻¹, with a low concentration in the soils near secondary ponds and the highest concentrations in TP2. The gallery mine sediments corresponded to the sediments that left a tunnel located in the 'Principal' vein, where an underground water current flows and discharges into the 'Veneno' stream. TP3 corresponded to the last tailings used until 1999, when mining operations ceased. Although the use of secondary ponds is unclear, they may have acted as a stage prior to downloading in streams (González Valoys et al. 2021).

According to the Panama Soil Standard (Gaceta Oficial Digital, 2009) (Table 4), the soils near secondary ponds exceeded the limit that the standard sets out for industrial use for As, and also for residential use for Ba and Zn.

Although tailings are not soil, their behaviour with the climatic conditions is similar, and also they can favour the dispersion of pollutants to surrounding soils and water bodies (González-Valoys et al., 2021; Rodríguez-Hernández et al., 2021). This is why the study materials relating to mining operations were compared to a soil standard, either from Panama or Costa Rica (Table 4), which gave the following PTEs of interest: As, Hg, Ba, Zn (Gaceta Oficial Digital, 2009), Cu, Sb (Ministerio de Salud, 2010), for exceeding the limit for some agricultural, residential or industrial uses. Local residents employ tailings, one as a football ground (TP1) and another to graze animals on, e.g. horses (TP3). Hence the need to study and compare them to regulations, and to evaluate their degree of pollution and the ecological risk they pose.

3.2. Statistical analysis

Fig. 2 presents the multivariate PCA performed of the samples studied for the T-CN physicochemical parameters, the precious elements that were the object of the mining process (Au, Ag), Fe, and the PTEs of interest (Cu, Zn, As, Sb, Ba, Hg). According to the strengths of the

Table 3 Average values and standard deviations of the physicochemical parameters and PTEs (mg kg⁻¹) in the different material types in the sampled areas. Abbreviations: EC, electrical conductivity; ORP, oxidation reduction potential; OM, organic matter; CEC, cationic exchange capacity; T-CN, total cyanide.

| Location | Sample type | pH | EC dS m ⁻¹ | ORP mV | Humidity % | OM % | CEC cmol kg ⁻¹ | Fe % | Au ng g ⁻¹ | Ag mg kg ⁻¹ | T-CN mg kg ⁻¹ | As mg kg ⁻¹ | Hg mg kg ⁻¹ | Sb mg kg ⁻¹ | Cu mg kg ⁻¹ | Ba mg kg ⁻¹ | Zn mg kg ⁻¹ | Pb mg kg ⁻¹ | Co mg kg ⁻¹ | Cr mg kg ⁻¹ | V mg kg ⁻¹ |
|--------------|-------------|-----------|-----------------------|---------------|-------------|-------------|---------------------------|-----------|-----------------------|------------------------|--------------------------|------------------------|------------------------|------------------------|------------------------|------------------------|------------------------|------------------------|------------------------|------------------------|-----------------------|
| TP1 | Tailing | 4.5 ± 0.4 | 0.03 ± 0.01 | 612.6 ± 23.1 | 14.0 ± 4.9 | 0.8 ± 0.2 | 4.8 ± 0.2 | 3.4 ± 1.7 | 617.5 ± 190.2 | 1.2 ± 0.2 | 1.8 ± 0.1 | 658.0 ± 410.1 | 1.1 ± 0.7 | 18.4 ± 10.1 | 34.4 ± 17.1 | 407.0 ± 227.7 | 18.0 ± 8.5 | 13.4 ± 6.7 | 1.3 ± 0.1 | 3.5 ± 0.7 | 18.0 ± 4.2 |
| | | 4.5 ± 0.3 | 0.03 ± 0.01 | 505.5 ± 76.3 | 14.5 ± 1.0 | 0.6 ± 0.1 | 4.5 ± 0.1 | 2.5 ± 0.2 | 507.0 ± 26.9 | 1.9 ± 0.3 | 1.6 ± 0.1 | 465.5 ± 37.5 | 1.0 ± 0.3 | 20.7 ± 2.5 | 21.5 ± 1.1 | 514.5 ± 135.1 | 14.5 ± 2.1 | 11.7 ± 1.6 | 0.9 ± 0.1 | 6.5 ± 0.7 | 14.5 ± 0.7 |
| | | 3.6 ± 0.7 | 0.49 ± 0.71 | 686.7 ± 66.2 | 19.5 ± 2.8 | 1.0 ± 0.4 | 3.5 ± 0.5 | 3.6 ± 1.5 | 464.7 ± 217.8 | 5.7 ± 8.7 | 5.7 ± 8.7 | 187.9 ± 285.9 | 1.4 ± 1.6 | 15.2 ± 2.3 | 109.1 ± 138.5 | 305.7 ± 102.7 | 40.3 ± 6.8 | 24.9 ± 22.8 | 1.4 ± 0.3 | 6.0 ± 2.6 | 16.3 ± 1.5 |
| SP1, SP2 | Sediment | 5.0 ± 0.4 | 0.08 ± 0.03 | 281.5 ± 6.6 | 40.7 ± 8.4 | 5.4 ± 2.9 | 10.2 ± 2.1 | 1.0 ± 0.3 | 86.4 ± 53.2 | 0.4 ± 0.4 | 1.9 ± 0.1 | 25.0 ± 10.8 | 0.2 ± 0.1 | 1.7 ± 0.8 | 12.8 ± 10.4 | 298.5 ± 40.3 | 23.5 ± 2.1 | 7.4 ± 0.0 | 2.4 ± 2.7 | 2.0 ± 0.0 | 19.0 ± 1.4 |
| | | 4.8 ± 0.2 | 0.11 ± 0.06 | 453.8 ± 153.3 | 26.5 ± 8.1 | 5.3 ± 1.2 | 9.6 ± 0.3 | 2.3 ± 0.7 | 29.9 ± 19.3 | 0.1 ± 0.1 | 1.7 ± 0.2 | 56.0 ± 4.2 | 0.1 ± 0.1 | 0.9 ± 0.6 | 10.4 ± 4.3 | 196.3 ± 45.4 | 44.0 ± 23.4 | 5.5 ± 1.9 | 1.7 ± 1.1 | 2.7 ± 0.6 | 26.7 ± 8.5 |
| Mine gallery | Sediment | 3.9 ± 0.2 | 0.52 ± 0.52 | 542.3 ± 542.3 | 63.9 ± 63.9 | 12.9 ± 12.9 | 10.9 ± 10.9 | >30.0 | >1000 | 0.7 ± 0.7 | 1.4 ± 1.4 | 5030.0 ± 5030.0 | 0.6 ± 0.6 | 2.2 ± 2.2 | 403.0 ± 403.0 | 55.4 ± 55.4 | 153.0 ± 153.0 | 1.7 ± 1.7 | 13.7 ± 13.7 | 2.0 ± 2.0 | 27.0 ± 27.0 |
| | | 3.9 ± 0.2 | 0.52 ± 0.52 | 542.3 ± 542.3 | 63.9 ± 63.9 | 12.9 ± 12.9 | 10.9 ± 10.9 | >30.0 | >1000 | 0.7 ± 0.7 | 1.4 ± 1.4 | 5030.0 ± 5030.0 | 0.6 ± 0.6 | 2.2 ± 2.2 | 403.0 ± 403.0 | 55.4 ± 55.4 | 153.0 ± 153.0 | 1.7 ± 1.7 | 13.7 ± 13.7 | 2.0 ± 2.0 | 27.0 ± 27.0 |

Table 4
Soil guidelines for the PTEs for Panama and Costa Rica (all the values are expressed as mg kg⁻¹).

| Soil Guidelines | Uses | Ag | As | Hg | Sb | Cu | Ba | Zn | Pb | Co | Cr | V | Reference |
|---|---|--------------|-----|-----|----|-----|------|------|-----|-----|------|-----|-------------------------------|
| Panama Maximum permissible limits of soil contaminants for human health | Others (agricultural) | | 4 | 1.4 | | | 10 | 3 | | | 10 | | Gaceta Oficial Digital (2009) |
| | Residential | | 20 | 14 | | | 100 | 30 | | | 100 | | |
| | Industrial | | 30 | 140 | | | 1000 | 300 | | | 1000 | | |
| Costa Rica | Prevention Value | 2 | 5 | 0.5 | 2 | 20 | 150 | 300 | 72 | 25 | 2 | 52 | Ministerio de Salud (2010) |
| | Intervention Value (Concentration above which there are potential direct or in direct risks for human health) | Agricultural | 25 | 35 | 12 | 5 | 20 | 300 | 450 | 180 | 35 | 40 | |
| | Residential | 50 | 55 | 36 | 10 | 50 | 500 | 1000 | 300 | 65 | 270 | 250 | |
| | Industrial | 100 | 150 | 70 | 25 | 100 | 750 | 2000 | 400 | 90 | 270 | 250 | |

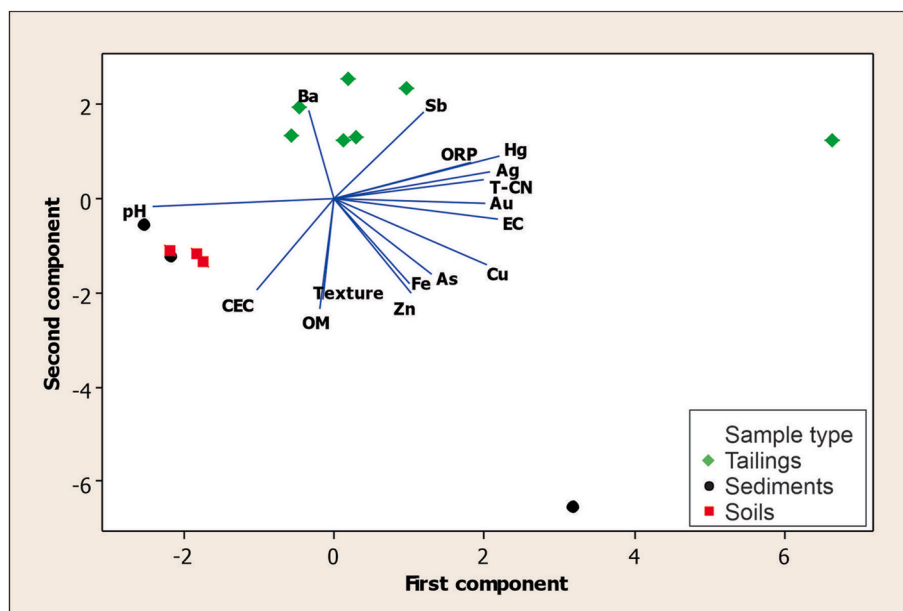


Fig. 2. PCA of the relations among T-CN, the main parameters and the PTEs concentrations. Generated with the data from tailings, sediments and soils.

relations (Table 5), the most significant relations were: for the first principal component (PC1), a positive relation was observed among T-CN (0.310), Ag (0.322), Cu (0.316), Au (0.313), Hg (0.343), EC (0.337) and ORP (0.282), and a negative relation to pH (-0.371). The second principal component (PC2) was positively related to Ba (0.316) and Sb (0.313), and negatively to OM (-0.396), texture (-0.364), Zn (-0.342), CEC (-0.330), Fe (-0.307) and As (-0.274).

Table 5
Principal component analysis matrix for the relation between T-CN (total cyanide) and the physicochemical parameters. Numbers in bold correspond to PC1 or PC2 and are more significant.

| Variable | PC1 | PC2 |
|----------|--------------|---------------|
| pH | -0.371 | -0.026 |
| EC | 0.337 | -0.075 |
| ORP | 0.282 | 0.130 |
| OM | -0.028 | -0.396 |
| CEC | -0.157 | -0.330 |
| Fe | 0.158 | -0.307 |
| Au | 0.313 | -0.016 |
| Ag | 0.322 | 0.096 |
| T-CN | 0.310 | 0.069 |
| As | 0.203 | -0.274 |
| Hg | 0.343 | 0.153 |
| Sb | 0.184 | 0.313 |
| Cu | 0.316 | -0.237 |
| Ba | -0.049 | 0.316 |
| Zn | 0.159 | -0.342 |
| Texture | -0.021 | -0.364 |

The PCA diagram also revealed how samples were grouped according to their nature, the tailings in the upper zone, the sediments of secondary ponds and the soils close to them on the left, and the mine gallery sediments far from the two previous groups (at the bottom) and to the right point Tail 3-2 of TP3, whose values were higher than the other tailings samples. This point had the highest T-CN concentration (518.0 mg kg⁻¹).

3.3. Pollution Index (PI) and Pollution Load Index (PLI)

Table 6 presents the PI and PLI values. According to the evaluation criteria for the PI and PLI in Table 1, the PI indicated considerable pollution (PI > 3) by T-CN, As, Hg, Sb and Cu, with the highest values in the tailings and the mine gallery sediments, followed in order of affection by secondary ponds and the soils near them. It is worth noting the very marked T-CN pollution that TP3 presented, and the mine gallery sediments with As.

The PLI came in the following order: TP3 > mine gallery sediments > TP1 > TP2 > secondary pond sediments > soil close to secondary ponds. These findings indicated that the gallery mine and tailings were sources of pollution for the surrounding area.

3.4. Toxicity response coefficient (Er) and the potential Ecological Risk Index (RI)

The values calculated for Er and RI of Cu, Zn, As, Sb and Hg are shown in Table 7. According to the evaluation criteria in Table 1 for Er and toxicity according to each element, the Er values suggests that TP1,

Table 6

The average values and standard deviations of the Pollution Index (PI) and the Pollution Load Index (PLI) calculated for the different sample types.

| | Sample type | PI T-CN | PI As | PI Hg | PI Sb | PI Cu | PI Ba | PI Zn | PLI |
|------------------------|-------------|---------------|-------------|------------|-------------|-------------|-----------|-----------|------------|
| TP 1 | Tailing | 3.5 ± 0.1 | 35.4 ± 22.0 | 7.1 ± 4.7 | 61.2 ± 33.7 | 4.9 ± 2.4 | 1.7 ± 1.0 | 0.6 ± 0.3 | 6.0 ± 2.9 |
| TP 2 | Tailing | 3.2 ± 0.3 | 25.0 ± 2.0 | 6.7 ± 1.7 | 68.8 ± 8.2 | 3.1 ± 0.2 | 2.2 ± 0.6 | 0.5 ± 0.1 | 5.3 ± 0.6 |
| TP 3 | Tailing | 375.9 ± 571.7 | 34.0 ± 5.2 | 9.4 ± 10.5 | 50.6 ± 7.5 | 15.6 ± 19.8 | 1.3 ± 0.4 | 1.4 ± 0.2 | 12.6 ± 8.2 |
| SP1, SP2 | Sediment | 3.7 ± 0.1 | 1.3 ± 0.6 | 1.4 ± 0.5 | 5.7 ± 2.8 | 1.8 ± 1.5 | 1.2 ± 0.2 | 0.8 ± 0.1 | 1.8 ± 0.4 |
| Soil close to SP1, SP2 | Soil | 3.3 ± 0.3 | 3.0 ± 0.2 | 0.7 ± 0.3 | 3.1 ± 1.9 | 1.5 ± 0.6 | 0.8 ± 0.2 | 1.6 ± 0.8 | 1.6 ± 0.3 |
| Mine gallery | Sediment | 2.8 | 270.4 | 4.1 | 7.3 | 57.6 | 0.2 | 5.5 | 7.7 |

TP2 and TP3 pose a serious risk for their As, Hg and Sb contents, as well as mine gallery sediments, given their As, Hg and Cu contents. The extreme risk of the mine gallery sediments is highlighted for their high As content.

The average RI value presented the following order of potential ecological risk: mine gallery sediments (extreme risk); TP3, TP1, TP2 (serious risk); secondary pond sediments and nearby soils (considerable risk). This means that the gallery mine and tailings are sources of pollution for the surrounding area and represent a high ecological risk.

4. Discussion

Remance mine tailings still contain large amounts of PTEs and, together with the mine gallery sediments, they pose a risk as source of environmental pollution, given their total As, Cu, Sb, Ba, Hg and Zn contents, as verified by the presence of these PTEs in nearby soils and the sediments of the water network in the study area (González-Valoys et al., 2021). All these pollutants can cause several health problems: As can cause skin, liver and lung cancers; Cu can provoke abnormalities to the nervous system; Sb can harm the respiratory system; Ba can favor muscle paralysis; Hg produces neurological damage; Zn can weaken the immune system (Bini and Wahsha, 2014).

Although the T-CN concentrations found in most of the studied area fell within the ranges reported for gold mine tailings at American sites (Kjeldsen, 1999), T-CN at TP3 was higher than that reported in the literature for closed gold tailings in Canada (Zagury et al., 2004). Although we would expect cyanide to have evaporated as free cyanide 20 years after mining operations ceased, its concentrations should be similar to those in other areas, but it persists in TP3. The high T-CN concentration at TP3 did not seem to be linked with sample texture as the texture and colour of the three tailings were similar, and they also showed similar Fe, As, Hg and Sb concentrations. Nevertheless, the pH at TP3 was more acidic, both EC and ORP were higher, as were the Cu, Zn and Ag concentrations. The PCA showed a close relation between T-CN and Au, Ag, Hg and Cu, with which it would seem complexed and favoured by today's EC and ORP conditions. These cyanide complexes formed under certain conditions like sunlight and a strongly acidic medium slowly decompose (Meeussen et al., 1992; Anning et al., 2019) and are released to the environment, a phenomenon that has been reported by the study of Johnson et al. (2002), as photochemical changes in cyanide in tailing piles. The presence of T-CN and complexed cyanide was found in surrounding soils, stream sediments and the terrace sediments near the Remance mine tailings, while easily released cyanide was below the detection limit (González-Valoys et al., 2021), which demonstrates that cyanide complexes affect soils and water bodies, favoured

by the slope at the site and by runoff. All this suggests that the cyanides at TP1 and TP2 were treated after mining closed. However, high T-CN concentrations persist at TP3, which is the largest of the three and the last to be used, due to inappropriate abandonment and no residual cyanide decomposition.

In turn, T-CN is slowly released to the environment, and favours the release of other pollutants, such as Cu and Hg, with which it travels complexed. They have also been found in nearby soils, streams and terrace sediments (González-Valoys et al., 2021). Ba in the PCA analysis did not appear to be bound to T-CN, but to a second factor, Ba, which was inert, and could be bound due to its relations with Au–Sb in mineralisation as inert Ba.

The acidic pH at certain points like TP3 (pH 3.6), and the mine gallery sediments (pH 3.9), which are potential acid mine drainage (AMD) generators, could be due to their content of pyrite and marcasite (FeS₂), chalcopyrite (CuFeS₂) and arsenopyrite (FeAsS) minerals, as reported in the mining mineralogy by the company Remance S.A. (Nelson and Ganoza, 1999). AMD is produced as a result of abiotic and biotic reactions that involve water and air, and with sulphide minerals present in mine wastes (e.g., pyrite). These reactions produce acidic effluents that tend to be loaded with several heavy metals and metalloids (e.g., Fe, Cu, As, Hg) (Nadeif et al., 2019).

Arsenic came at high concentrations (5030.0 mg kg⁻¹) in the mine gallery sediments, and had to take an inorganic form after being released from minerals, such as arsenopyrite (FeAsS) from the 'Principal vein', from which the underground water stream came. The mine gallery sediments were characterised for their clay texture composition, acidic pH (3.9) and with high CEC (10.9). The high concentration of these PTEs (As, Cu, Zn) at this point could be associated with clay fractions, such as kaolinite and illite (Palansooriya et al., 2020), which were detected in the soil and sediment samples taken from streams near the area (González-Valoys et al., 2021), for which the best explanation would be the absorption/adsorption of PTEs by clays (González-Valoys et al., 2021).

The PI and PLI allowed a comparison to be made of the pollution level in an area to background areas. Thus the PI of T-CN showed marked pollution (PI > 3) in the tailings area and its surroundings, and TP3 obtained the highest value (PI 375.9) and, therefore, poses a source of pollution for the surroundings given its T-CN, As, Hg, Sb and Cu contents, which can have adverse health effects. For the PLI, TP3 was also the area with the highest pollution, followed by the mine gallery sediments, TP1 and TP2. This would be the order to be followed to recover areas in a recovery management plan for this area. To evaluate the possible effect of PTEs on the biota, the RI and Er were used, which complement information about the PLI. They revealed that the mine

Table 7

Average values and standard deviations of the toxicity response coefficient (Er) and the Potential Ecological Risk Index (RI) for the different sample types.

| | Sample type | Er As | Er Hg | Er Sb | Er Cu | Er Zn | RI |
|------------------------|-------------|---------------|---------------|---------------|-------------|-----------|------------|
| TP 1 | Tailing | 353.8 ± 220.5 | 284.0 ± 186.7 | 428.2 ± 235.9 | 24.6 ± 12.2 | 0.6 ± 0.3 | 1091 ± 656 |
| TP 2 | Tailing | 250.3 ± 20.1 | 266.7 ± 67.9 | 481.8 ± 57.7 | 15.3 ± 0.8 | 0.5 ± 0.1 | 1015 ± 145 |
| TP 3 | Tailing | 340.3 ± 51.9 | 376.0 ± 421.0 | 353.9 ± 52.5 | 77.9 ± 98.9 | 1.4 ± 0.2 | 1150 ± 592 |
| SP1, SP2 | Sediment | 13.4 ± 5.8 | 56.0 ± 18.9 | 39.7 ± 19.8 | 9.1 ± 7.4 | 0.8 ± 0.1 | 119 ± 40 |
| Soil close to SP1, SP2 | Soil | 30.1 ± 2.2 | 29.3 ± 13.9 | 21.8 ± 13.5 | 7.4 ± 3.1 | 1.6 ± 0.8 | 90 ± 27 |
| Mine gallery | Sediment | 2704.3 | 165.3 | 51.3 | 287.9 | 5.5 | 3214 |

gallery sediments and tailings posed a serious ecological risk for the biota in this place, as well as a considerable ecological risk for the surrounding area.

Tailings with high Au and Ag contents must be reprocessed to extract the amount of remaining precious metals, which should then be properly remediated to avoid them becoming a source of pollution for the community via polluting soils, stream sediments and terrace sediments, which pose a risk for the ecology and human health of its inhabitants (González-Valoys et al., 2021). Longer-term remediation should be implemented using the measures proposed in the literature, such as chemical degradation of residual cyanide (Ebbs, 2004), covering tailing ponds (Henny et al., 1994) and the bioremediation (Viera and Stefenon, 2017) of not only surrounding soils, but also of stream sediments and sediments.

The problem of improperly abandoned mines is a latent problem in Panama and other countries. For this reason, there is a dire need to implement some type of economic surety for mining companies to raise funds with which to remedy the area if the company goes bankrupt. It is also necessary to reinforce measures to monitor proper compliance with environmental regulations and to promote scientific research in this field.

5. Conclusions

The tailings from the Remance gold mine and mine gallery sediments present a high degree of pollution given their PTEs (As, Hg, Sb, Cu) and T-CN contents. The highest PLI went to tailings 3, due to their high T-CN content. Hence, they represent a source of pollution for the surrounding areas and pose a serious ecological risk (RI) for the biota in this place.

Total cyanide content is similar to that of gold mine tailings at American sites in most of this area, but the last used tailings from mining operations contained a highest T-CN concentration. This clearly demonstrates that this pollutant persists more than 20 years after closing mining operations. This finding suggests that in later tailings, cyanide contents were not chemically treated before shutting down mining operations.

T-CN under certain specific pH, EC and ORP conditions is slowly released to the environment, which favours the release of other pollutants like Cu and Hg, with which it travels in a complexed manner. The pollutants that were also found in nearby soils, streams and terrace sediments also pose a risk to both the environment and human health of Remance community inhabitants (González-Valoys et al., 2021).

The Remance mine is an abandoned area with no environmental control. An environmental monitoring plan should be set up to avoid undesirable water, soil and plant uses by the local population. We recommend remediating the tailings ponds and mine works areas because they pose a serious environmental risk. We also recommend conducting bioavailability and bioaccessibility studies on these materials.

Declarations

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Authors' contributions

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Animal research

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Ethics approval

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Consent to participate

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Declaration of competing interest

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Appendix A. Supplementary data

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