Hydrogeological analysis of sulfide tailings at a uranium mine using geophysical and hydrochemical methods

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Abstract

A hydrogeological analysis was conducted of aquifer systems within a sulfide-rich tailings pile and underlying rocks at a uranium mine to understand the generation and transport of AMD. A geophysical investigation was carried out along seven transects spaced 30 m apart using electrical resistivity tomography and a Wenner-Schlumberger array. The results were used as input to 2D and pseudo-3D models of resistivity and chargeability parameters. The geophysical results and models contrasted unaffected saturated zones and zones impacted by AMD, in addition to three areas within the tailings with a high ulfide content. The tailings and underlying fractured bedrock aquifer are hydrologicall connected, which promotes the exchange of mine-influenced water into the regional aquifer. Releases from the site's tailings and other wastes can severely affect regional groundwater.

Introduction

The mining industry has experienced vigorous growth in recent decades due to profound socioeconomic changes around the world. When economic performance is high, investments in technology and effective solutions in environmental remediation of degraded areas is assumed from the earliest stages of mining. However, decommissioning and mine closure may be considered secondary priorities in mine planning because these activities add cost without financial return compared to mining and ore processing (Burritt e Christ, 2018).

The Osamu Utsumi Mine, located at Caldas (State of Minas Gerais) in Brazil, has decommissioned a uranium mine to reduce environmental impacts and comply with urrent environmental regulations. One of the biggest challenges in this area is the volume of acid-generating waste material and tailings (Dutta et al. 2020; Freitas et al. 2011; Lei et al. 2005).

Acid mine drainage (AMD) is produced as a consequence of chemical weathering of either a natural ore deposit or mining wastes. Consequently, the AMD can mix with groundwater flow and leach high concentrations of dissolved metals and other constituents from rocks and waste materials (Pabst et al., 2018; Simate and Ndlovu, 2014). These effluents are especially enriched in metals that are mobile in oxidizing environments (Nordstrom et al., 2015). In the case of a uranium mine, the mobilization of radioactive elements might contribute to the deterioration of the local water quality. Despite being a wellunderstood process, AMD is extremely complex to remediate and is often seen as an irreversible environmental liability with costly mitigation measures (Akcil and Koldas, 2006).

Geophysical methods have been increasingly used to investigate derelict and contaminated land and groundwater to locate contaminant plumes before digging pits and boreholes, including at sites affected by AMD from tailings (Epov et al., 2017; Martín-Crespo et al., 2018; Power et al., 2018; Targa et al., 2019).

Direct current (DC) resistivity and induced polarization (IP) methods are non-invasive, have great spatial coverage, and can be used to recognize inorganic contaminants in soils and rocks and to study the physical and geotechnical integrity of waste and tailings piles (Anterrieu et al., 2010; Casagrande et al., 2020; Spitzer and Chouteau, 2003). 2D and 3D geophysical modeling of the results from these geoelectrical methods combined with electrical resistivity tomography (ERT) can reliably and consistently identify the presence of acidic plumes and help determine their lateral and vertical extent (Bortnikova et al., 2018; Olenchenko et al., 2016).

In this context, the present work assessed underground flow of AMD within a tailings pile (BF-08) and its underlying rock at a uranium mine complex using DC resistivity and IP with 2D and pseudo-3D tomographic modeling. In addition, water quality data from groundwater samples were used to elucidate hydrogeological processes that could be occurring inside the tailings.

The Osamu Utsumi Mine

The Osamu Utsumi Mine is part of an industrial mining complex of Poços de Caldas (CIPC) owned by Nuclear Industries of Brazil (INB-Indústrias Nucleares do Brasil), a government entity, and the location of the nation's first uranium production center.

The main facilities of the Osamu Utsumi Mine include the mine open pit, tailings impoundments, the processing plant for uranium extraction, waste rock piles, and administrative and operational support buildings (Figure 1). The mine operated from 1981 to 1995, and uranium mining and milling operations ceased after depletion of the uranium ore. Its reserves have been estimated at ≈ 17,200 t of U_3O_8 (Fraenkel et al., 1985; Souza et al., 2013). About 1,242 t of ammonium diuranate (yellowcake), 4,48×107 m³ of waste material, and 2,39×106 m³ of mine tailings were produced during the mine's operational years (Franklin, 2007).

All effluents and solid waste generated by yellowcake production were sent to the wastewater treatment plant. Effluent treatment consisted of the neutralization of sulfuric acid with ground limestone to a pH of 9 to 10. Neutralization precipitated almost all iron and other metal ions (Al, Mg, Mn) as hydroxides. Likewise, the solid waste was composed of 98% low-grade uranium ore obtained by filtering, with a chemical treatment of neutralization with limestone. All waste produced from these methods was disposed in the tailings impoundment (Cipriani, 2002).

Starting in 1998, the sludge produced by effluent and solid waste treatment was placed in the open pit. Its composition consisted mainly of metal hydroxides, such as Fe, Al, Mn, and radioactive elements, in a calcium sulfate matrix. This material is locally known as "DUCA" (for calcium diuranate) and includes the sludge produced by lime treatment of open-pit contact water and acid drainage from waste piles (Cipriani, 2002).

The study area was part of the BF-08 tailings pile, which covers an area of 64,4 million m², has a volume of 15 million m³, 5 m wide benches, and 8 m long rocky slopes at an angle of 70º. The rocks in the pile are mainly composed of syenites and phonolites from the open pit area with an ore grade less than 170 ppm and grain sizes ranging from fine sand (0.05 to 0.2 mm) to boulders (> 200 mm).

The tailings came from the picking and sorting of alkaline pyritic rocks (Cipriani 2002). The material was end-dumped, so generally, fine grains remain at the top of the pile, whereas coarser cobbles and boulders are found in its basement.

The Cercado Stream had part of its channel diverted using tailings and waste rock for the new channel. The BF08 site was covered with a 20 cm of clay with a slope between 0.5% and 1%. Rainwater run on was captured by concrete ditches and a piping system discharged the water to the local hydrological system. However, due to a lack of regular maintenance, the clay layer has been progressively eroded by rainwater, some ditches were damaged, and erosion has intensified on its slopes. As a result, neutral pH precipitation currently infiltrates into the pile and reappears downstream as acid drainage in a lake known as the BIA Reservoir. It is important to note that even though AMD is observed downstream, no contaminated seeps were observed to be issuing from the impoundment slopes in the study area.

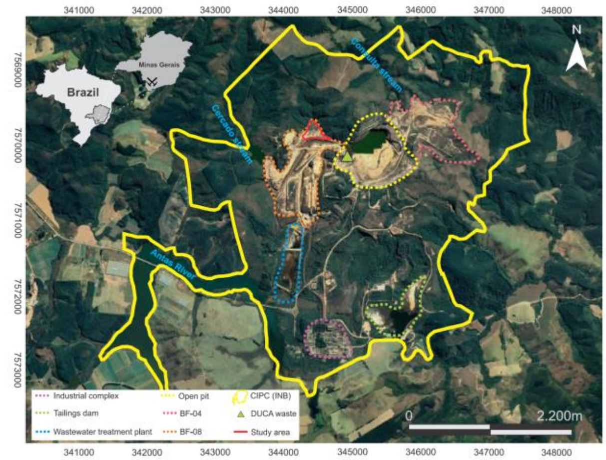

***Figure 1** – Study area location map.*

Method

The geophysical investigations were carried out along seven lines to measure resistivity and chargeability (Figure 2), and were conducted during the dry season. The geophysical lines were parallel to each other and arranged 30 m apart, with a minimum length of 120 m (line 1), a maximum length of 340 m (line 7), and an electrode spacing of 5 m. DC resistivity and IP methods were applied using ERT and a Wenner-Schlumberger array.

The ABEM Terrameter LS resistivity meter (84 channels, 250W, with a resolution of 1 μV, and a maximum current of 2.5A; ABEM, 2012) that was used consists of a single module for transmission and receiving of automated signals to obtain resistivity and chargeability parameters simultaneously. The survey acquisition parameters settings were a signal sending time of 1.5 s, electrical current of 500 mA, two fixed reading windows of 100 ms, and 0.5 s of acquisition time delay. Non-polarizable porous-pot electrodes in a Cu-CuSO4 solution were used to avoid local polarizations that would interfere with chargeability measurements during the IP survey.

The results from each geophysical line were analyzed using a data processing and inversion routine with Res2Dinv (2D) software, version 3.53 (Geotomo Software) to produce bidimensional sections with resistivity and chargeability values according to their distance from line origin and depth of acquisition. This software was designed to interpolate and invert field data from electrical geophysical prospecting according to the mathematical model of ordinary least squares (OLS). This technique smooths the extreme values using block modelling and reduces differences between measured and modeled parameters (Reynolds, 2011). Resistivity inversion models required 5 to 7 iterations to obtain the most reliable 2D sections, whereas chargeability inversion models demanded 4 to 7 iterations, as indicated in each 2D section.

Block modelling generates a standard deviation parameter called the root mean squared (RMS) factor, which mainly represents the match between the calculated pseudo-section and that obtained in the field, influenced by the presence of extreme values in the input data, potentialized by post-processing (Chulès and Delfiner 2012; Geotomo 2003; Loke 2000). Resulting resistivity and chargeability models are presented as 2D sections with a chromatic logarithmic scale and intervals of interpolation in color values. This ERT data inversion routine is used in many kinds of studies, including mineral prospecting, hydrogeology, contaminated area diagnosis, and geotechnical engineering (Berthold et al., 2004; Camarero and Moreira, 2017; Falgàs et al., 2011; Helene et al., 2016, 2020; Moreira et al., 2019).

The geophysical surveys were planned to produce 3D visualization models by interpolation of the 2D sections, in addition to isovalue surfaces for resistivity and chargeability. This procedure was carried out using the Oasis Montaj Software Geosoft platform, which consisted of a tridimensional model created from coordinating data (X, Y, and Z) from the 2D sections (Geomotosoft, 2014). Moreover, local topography was considered in all models to avoid distortions using the differential global positioning system. 3D modeling and isovalue surfaces improved the recognition and characterization of groundwater flow and sulfide zones, as well as AMD transport.

INB provided hydrochemical data from monitoring wells and surface water from 2015 to 2017 so that we could quantify chemical data and changes in physical-chemical parameters in groundwater. The nearest monitoring wells to the study area and the acid drainage lake located at the toe of BF-08 (Figure 2) (the BIA Reservoir) were selected for chemical analysis of pH, electrical conductivity (EC), redox potential, dissolved oxygen (DO), turbidity, and major element chemistry (cations and anions). The hydrochemical data complemented the geophysical interpretations from BF-08 and helped identify the most critical AMD generation zones.

Groundwater samples were collected using the micro-purge low flow sampling method. First, monitoring wells went through a purging process to remove stagnant water, since this water cannot be considered as representative of the local aquifer quality. Then, sampling was performed using the micro-purge low flow method at flows of ≤ 0.5 L/min to minimize water table lowering (Popek, 2018). This procedure ensures that the sample collected is consistent and representative of the contaminants in the local aquifer.

Chemical analyses were carried out by the INB team in partnership with CNEN (Brazilian Atomic Energy Commission) in the Poços de Caldas Laboratory (LAPOC/CNEN). The physical-chemical parameters were measured using a multiparameter probe coupled to a flow cell used in situ without filtration. Cations and anions were determined using plasma optical emission spectrometry (ICP-OES) in filtered samples that had been preserved with nitric acid. Sulfate was determined gravimetrically with barium chloride, and fluoride was determined using potentiometric analysis with an ion-specific electrode (Alberti, 2017).

Figure 2 *– Geophysical survey lines;; Location of monitoring wells PM-10 and PM-38.*

Results

A pseudo-3D model was generated by the interpolation of the seven geophysical sections using the physical parameters of resistivity and chargeability to facilitate spatial visualization. After data processing, images were smoothed, and visualization levels were defined using a specific depth range (Figure 3).

Resistivity models from 12 to 32 m in depth indicate a highly ionic effluent flowing from northeast to southwest and, therefore, a similar drainage pattern to the stream valley under the tailings pile. The contact between the tailings and bedrock is evident by a reduced number of saturated zones, with a resistivity > 537 Ω·m. The water content decreases in part due to the presence of an elongated feature with high electrical resistance and N/S orientation, which is interpreted as the top of the bedrock with increasing resistivity values with depth (> 2,000 Ω·m from 42 m in depth), interrupting the connection between the saturated and unsaturated zones.

Visualization levels also indicated specific locations where there is clear evidence of effluents flowing from the tailings towards the underlying fractured aquifer. The existence of a low resistivity zone in the southeast portion of the pseudo-3D model, towards the open pit area, is another indication that local aquifers could be hydraulically connected, with acidic effluent flowing through the fracture system. Another feature observed was a NW-SE alignment, highlighted at 42 and 62 m depths, indicating a fractured zone that transports water into the bedrock.

The DUCA residue inside the open pit area close to BF-08 is potentially problematic. Water from the old exploration areas and the local water table will interact with this residue and transport contaminants toward the fractured aquifer. Consequently, this contaminated effluent infiltrates into the deepest part of the aquifer through the local fracture system and, as the water table rises, it flows toward the porous portion of BF-08.

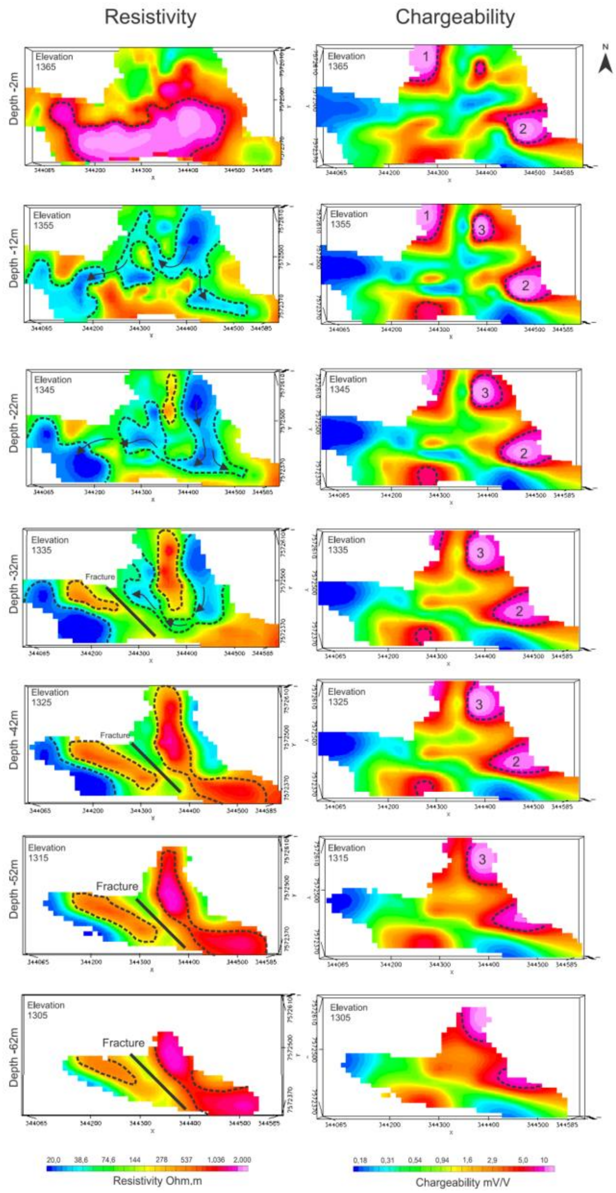

***Figure 3** - Visualization levels from 2m and 62m depth and geophysical interpretation based on the resistivity and chargeability 3D models*.

The hydrochemical data complemented geophysical interpretations to characterize the hydrogeological environment of BF-08 (Table 2). Data from monitoring wells PM-10 and PM-38 indicated distinct physicalchemical characteristics of local groundwater. These wells were installed at 8 m and 25 m deep, respectively. Thus, PM-10 reflects chemical conditions in the tailings, whereas PM-38 gives chemical information about the underlying fractured aquifer, assuming its well casing prevents influence from the overlying tailings.

The tailings water had an acidic pH (4.50 to 4.66), higher concentrations of DO (2.81 to 4.67 mg/L), mostly positive redox potentials (oxidizing environment), and low EC values (50 to 93 µS/cm). The deeper zones had a more reducing environment, with slightly acidic to neutral pH (5.37 to 6.86), lower concentrations of DO (0.45 to 2.2 mg/L), and higher EC values close to the open pit area (360 to 707 µS/cm), in addition to high sulfate concentrations.

The water sample from BIA Reservoir had the most acidic pH (3.76) and the highest EC (1,228 to 1,450 µS/cm) (Table 2). Analytical results indicated higher concentrations in the BIA Reservoir than in the monitoring wells. Sulfate dominates the sample, reflecting the oxidation of sulfide minerals in BF-08. The acidic water enhances the chemical mobility of metals and explains the higher Al, Fe, and Mn concentrations. Because groundwater sampling was in the upper layer of the local aquifer system, it was expected that the water quality would be similar to that observed in the shallow aquifer zone (B), as described in Table 1. However, the sample from the upper fractured aquifer (PM-38, Table 2) has higher SO4 and F concentrations and a slightly acidic pH compared to the background levels in the regional aquifer (Sample B, Table 1).

Although the tailings have favorable conditions for acid generation and considerable potential to modify contaminant mobility inside the tailings, the fractured aquifer had higher sulfate concentrations, which was unexpected (Baruah and Khare, 2010; Jardim, 2014). Thus, it is possible that external factors might affect the chemical composition of the tailings water, including the possible dissolution of DUCA residues from inside the open pit. It is believed that the DUCA residue dissolves, infiltrates into the bedrock through fractures, and is dispersed into the entire fractured aquifer. The DUCA contaminants apparently are not restricted to the fractured hydrogeological environment. As shown in pseudo-3D models, the tailings and underlying fractured aquifer are hydraulically connected, which allows effluent circulation. In other words, in addition to acid drainage generated inside BF-08, the tailings could be impacted by contaminants from the DUCA residue.

Table 1 – Physical-chemical parameters and ionic content of stream water (A), shallow zone (B), intermediate zone (C) and deep zone (D) of the local aquifer (Costa et al. 1998).

Parameter	A	B	C	D
Temperature (°C)	19.5	21.5	22.7	37.5
pH	6.6	6.00	7.5	9.8
Conductivity (µmho/cm)	25.7	49.8	110	801
Ca (mg/L)	2.14	6.23	6.07	1.13
Mg (mg/L)	0.88	0.60	0.98	0.23
Na (mg/L)	0.64	4.30	32.3	189
K (mg/L)	1.53	4.15	2.95	6.2
HCO3 (mg/L)	11.5	14.5	62.7	155
CO3 (mg/L)	-	-	12.0	133
Cl (mg/L)	1.65	2.60	1.7	4.7
SO4(mg/L)	-	0.93	13.7	51.9
Fe total (mg/L)	0.81	-	1.02	0.08
F(mg/L)	0.44	0.18	5.84	20.8
SiO2 (mg/L)	-	14.0	16.0	29.1

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Table 2 - Analytical results of groundwater samples collected from PM-10 and PM-38 and surface water from the BIA Reservoir provided by INB.

Sample	Date	pH	Turb¹	EC²	DO³	T_4	WT_5	Eh
PM 10	Apr/2015	4.5	3.2	83	3.85	19.2	691	409
	Oct/2015	4.62	5	93	2.81	18.9	715	256
	Oct/2017	4.66	48	50	4.67	18.8	728	240
PM 38	Jan/2016	6.86	15.6	707	0.49	20.8	838	59
	Oct/2017	6.83	21.1	620	1.18	19.3	897	-11
BIA	nov/15	5.52	5.5	1228	3.49	19.8	770	299
	Oct/2017	3.76	431	1450	1.16	19.9	740	308

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Table 2 (continuation) – Analytical results of groundwater samples collected from PM-10 and PM-38 and surface water from the BIA Reservoir provided by INB.

Sample	Al	Fe	Mn	Sulfate	Fluoride
(mg/L)					
PM 10	1.9	0.19	2.34	48.6	-
	2.1	-	2.08	37.7	1.61
	1.03	0.08	0.83	17.8	-
PM 38	0.1	0.01	0.3	139	2.44
	0.07	0.13	1.63	257	3.5
BIA	76.8	18.6	49.0	942	43.3
	154	31.4	86.4	1280	47.0

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[1]Turbidity NTU; [2]Electrical Conductivity (µS/cm); [3]Dissolved Oxygen (mg/L); [4]Temperature (ºC); [5]Water Table Depth (cm); - No results available

Conclusions

The results of the geophysical study provided a good understanding of the hydrogeological dynamics and underground flow of AMD inside the tailings impoundment at BF-08, as well as the identification of sulfidic zones. It was possible to distinguish less impacted saturated zones (< 74.6 Ω·m) from those with high ion content probably related to AMD (< 20 Ω·m), as well as three sulfide mineralization and high resistivity areas indicating the boundary between the base of the tailings in BF-08 and the top of the bedrock at ≈ 35 m deep (below 1,335 m in elevation). Besides, they suggest a possible hydraulic connection between the tailings and the underlying fractured aquifer that favors chemical exchanges between them, and also showed that the mineralized area of BF-08 is susceptible to chemical weathering and leaching and, hence, to generating AMD due to its interaction with saturated zones. The hydrochemical data corroborated the geophysical interpretation and indicates the existence of two hydrogeological systems. The tailings material has characteristics that allow chemical weathering and leaching due to high concentrations of DO and an acidic pH. The combination of these two parameters provide ideal conditions to dissolve and mobilize constituents within the pile, such as Fe, Al, Mn, and S, and indicates that the tailings are a source of groundwater contamination. Although the groundwater in the fractured aquifer does not have a chemical signature compatible with acid drainage, the higher concentration of sulfate could reflect impacts from the dissolution and transport of contaminants from the DUCA residue. This residue could release contaminants that infiltrate into the bedrock through fractures and affect the regional aquifer system. Using the combined geophysical and hydrochemical results, it was possible to identify a transition zone below the overlying oxidizing zone in the tailings where leaching of sulfide minerals is intensified, which could provide high concentrations of dissolved ions to the underlying more-reducing groundwater. The tailings areas with high sulfide content should be especially targeted during remediation because they have a high potential to generate acid drainage. Thus, the integrated analysis of DC resistivity and IP methods provided an improved understanding of in situ mitigation measures that could be used to more successfully minimize and control releases during the decommissioning and closure of mining complexes.

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