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A first evaluation of water resource conditions after an environmental reclamation effort at a former degraded coal mining area in Southern Brazil

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Abstract   In southern South America, Brazil, in the state of Santa Catarina, the neglect and lack of environmental supervision during coal mining caused the contamination of surface and groundwater by acid mine drainage (AMD). By the force of the local law, environmental reclamation actions in these abandoned areas have been carried. A scientific work of monitoring and assessment of the water resources within these areas has never been developed, as the efficacy of the reclamation strategies has never been discussed. This work aims to fill this gap by presenting and analyzing the environmental reclamation strategy of a former degraded coal mining area and its impacts on local water resources. The water monitoring plan in Area IV's was carried out in groundwater, and in lentic (ponds) and lotic (rivers) environments of surface waters, fourteen monitoring campaigns were conducted. The results showed that upstream and downstream river points have different water qualities, with the downstream points having poorer water quality, still affected by past mining activities. From the surface water perspective, the reclaiming method adopted was effective in three of the four ponds, presenting problems only in the downstream one. Two hypotheses were proposed; the first hypothesis is that contamination happens due to leaching of the material that still remains on the ponds' banks into the water. Another hypothesis is that the contamination comes from the upstream groundwater inflow into the pond, which runs through the entire area before reaching the pond. Those results serve to further access the actual monitoring perspectives as well as to better develop future reclaiming strategies.

Introduction

Mining is important for the economic growth of many countries; however, its pollution capacity is higher when compared to other activities. Coal mining in particular can cause a number of impacts on the environment (Bian et al., 2010; Qureshi et al., 2016; Rathore & Wright, 2007). The severity of these impacts depends on factors such as geological conditions, whether the mine is active or abandoned, and the mining methods (Bell et al., 2001).

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The main environmental impact of coal mining is the contamination of nearby water resources by acid mine drainage (AMD) (Cravotta, 1989; Cravotta et al., 1999; Gray, 1998; Yang et al., 2006). According to Gray (1996), AMD is the main environmental contamination problem in the world, capable of affecting both surface and groundwater.

AMD results from the oxidation of sulfide minerals, usually pyrite, being removed from the interior of the earth by mining activities and exposed to oxygen and atmospheric water (Cravotta, 1989). Pyrite oxidation involves several redox reactions and microbial catalyses; it is considered a complex biogeochemical process (Nordstrom, 1982; Xin et al., 2021), and is characterized by promoting low pH values and high concentrations of sulphates and dissolved metals (Ardejani et al., 2011; Sarmiento et al., 2018; Underwood et al., 2014).

AMD-related issues are studied worldwide, in Asia (Jung & Thornton, 1997; Lee et al., 2001; Lin et al., 2005; Ouyang et al., 2015; Park et al., 2019; Rambabu et al., 2020; Tao et al., 2012; Wang et al., 2021), Oceania (Lottermoser et al., 1999; Wright et al., 2018), Europe (Haigh & Kilmartin, 2015; Olías et al., 2006; Sáinz et al., 2002; Sracek et al., 2010), Africa (Ochieng et al., 2010; Rösner & Van Schalkwyk, 2000), North America (Burrows et al., 2015; Kimball et al., 2002), and Latin America (Prieto & Duitama, 2004). In Brazil, AMD affects multiple regions across the country, including the more southern states (Santa Catarina and Rio Grande do Sul), but also the Paraná and Minas Gerais states, respectively, located in the south and southeast regions (Campaner et al., 2014; Casagrande et al., 2019).

Among the published studies related to AMD impacts in water resources, we believe it is important to highlight the work of Olías et al. (2006) which studied the amount of metals transported to the ocean due to AMD contamination in the Tinto and Odiel basins in Spain. They reached transportation quantities of 7900 tons per year of iron, 5800 tons per year of aluminum and 3500 tons per year of zinc, all directly related to AMD. According to the authors, those values were considered to be 60% of zinc global gross flux and 17% of copper transported to the ocean by rivers. Wang et al. (2021) reviewed AMD-related publications about abandoned coal mines in Shanxi, China; the authors showed that the mines have aggravated the situation of water shortages, causing an ecological and environmental problem and a

drinking water safety crisis. In another study, Haigh and Kilmartin (2015) analyzed four reclaimed open-cast coal-lands in Southeast Wales, Great Britain, in order to verify if these sites continue to impact the quality of the region's water resources. The authors concluded that compacted surface layers contribute to flood peaks and the main impact on water quality was due to diffuse pollution caused by AMD reactions in the aeration zone above the water table within the spoils banks.

In the state of Santa Catarina, southern Brazil, coal mining began in the late nineteenth century. However, the escalation of the exploration and the aggravation of environmental contamination happened in the 1970s, with the mechanization of the mines (Alexandre, 1999). Neglect and lack of environmental supervision during this period caused serious repercussions to the local environment. This culminated in 1980, when the Federal government signed a decree recognizing the coal region of Santa Catarina as a critical pollution zone in the country, consequence of coal mining.

The first scientific studies on environmental pollution in the region date back to the 90 s, some featuring the participation of government agencies (Alexandre, 1999; Alexandre & Krebs, 1995; Gothe, 1993). Since then, some other AMD-related works in the region have been published: modeling of contamination sources (Gomes et al., 2011), water resource toxicity (Lattuada et al., 2009), sediment assessment and treatment (Freitas et al., 2017; Volpato et al., 2017), tailings desulfurization method (do Amaral Filho et al., 2017), adsorption of heavy metals from coal AMD (Núñez-Gómez et al., 2019), and background analysis of the water in the coal area (Simão et al., 2019). These studies have made significant contributions to the scope of knowledge concerning degraded areas and their impact on the regions' environment. However, none of these works has fully investigated the recovery of these degraded areas' local water resources.

By the force of the law, environmental reclamation actions in these abandoned areas have been carried out by different agents, including private companies and even Federal agencies. After the reclaiming process of the areas, the monitoring, and assessment of water resources are carried out by those responsible, the results are then forwarded to the State Environmental Agency. However, the real effectiveness, impacts, and critical analyses of these reclaiming strategies have not been scientifically assessed and discussed in the literature.

Given this scenario, the objective of this research is to work towards filling this gap, while presenting and discussing, for the first time, the environmental reclamation strategy of a former degraded coal mining area, reclaimed by the Federal Government, located in Southern Brazil, and its impacts on local water resources.

Materials and methods

Location of the study area

The study area, known as "Area IV," has 42.23 ha and is located in the municipality of Siderópolis, southern state of Santa Catarina, Brazil (Fig. 1). The area is situated in the sub-basin of the Fiorita River, which belongs to the Araranguá river basin, one of the most affected basins by acid mine drainage (AMD) in the state of Santa Catarina.

According to the Köeppen classification, the climate of the area is humid subtropical climate (Cfa); the average annual temperature is around 16 to 20 °C, rising from December to February. The local average annual rainfall is 1630 mm; its distribution varies according to the seasons; in autumn and winter, the accumulated average is around 350 mm in each season; and in spring, the accumulated average is 430 mm and 500 mm in summer (Gotardo et al., 2018).

The area is located in the vicinity of the urban center of the Siderópolis city. At the north-northeast limit is located an industrial sector, which includes industries within the polygonal area. At the southern limit is the Albina hill, where springs of a few streams originate. These streams flow into the ponds formed by the old open pit mines.

The surface water flows from the southern boundary, enters the area through pond 4, and moves into
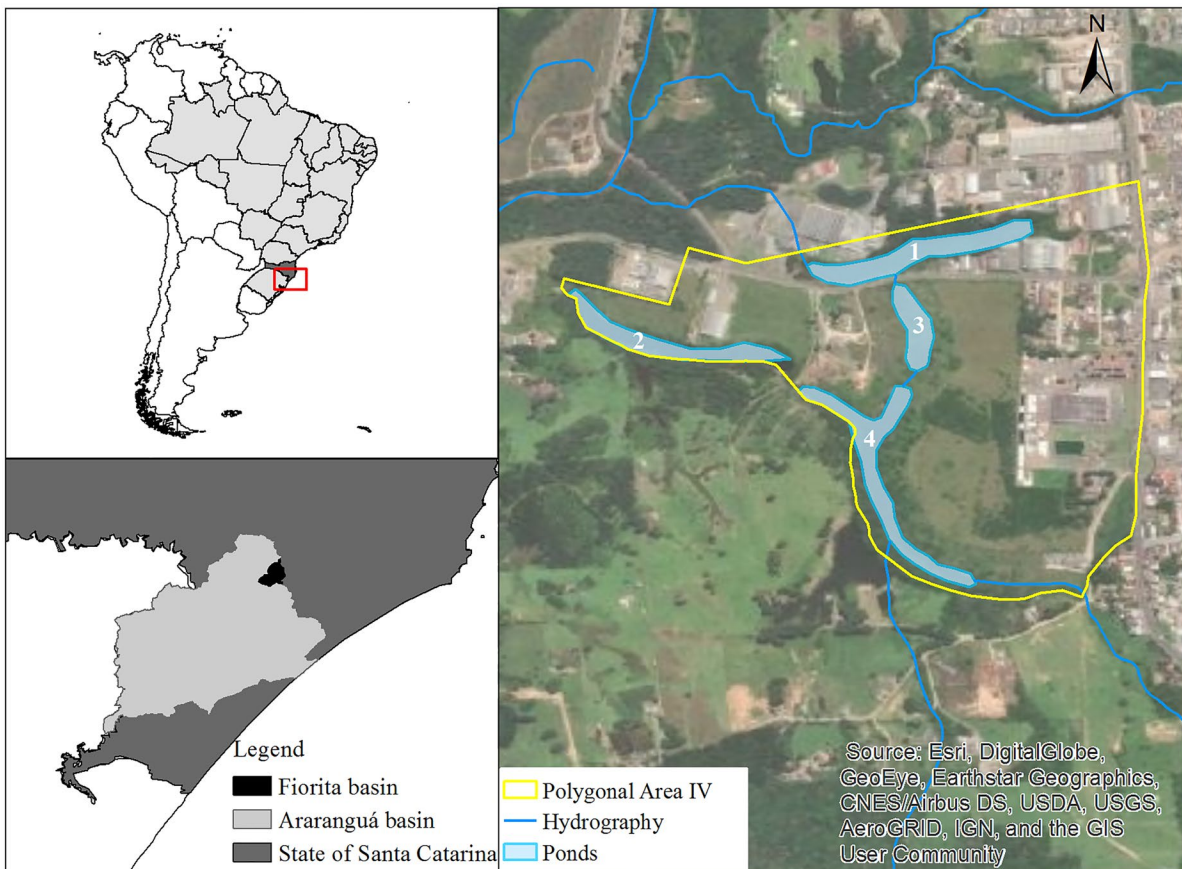

Fig. 1 Study area location

pond 3, then pond 1. The water flows through the out-flow from pond 1 towards the northwest of the area until it meets the Fiorita River. Pond 2 has no significant surface inlets or outlets.

Area history

Area IV is part of a region called "Vila Funil," one of the first areas mined using heavy equipment in the southern region of the Santa Catarina state. Research carried out in documents from the National Mining Agency of Brazil showed that the request for mining in the area dates back to 1936, but it was not possible to determine the date of the mine's closure. Based on information from local residents, we estimated that mining took place between the 1940s and 1950s.

Due to the proximity between the coal layer and the land surface, Area IV was mined using the technique called open cast mining. This technique consists of completely removing the upper lithological layers in order to access the coal layer. At the time this area was being mined, there was no environmental care in the removal of these layers and the reclaiming of the area. The removal of the rock layers above the coal was performed using explosives. After detonation, the mixed rock layers, including soil, were disposed of by huge machines, called dragline excavators, in piles from 10 to 20 m tall. This material, characterized by the mixture between lithological layers and soil, is referred to in this work by "waste rock."

Reclamation techniques

The reclamation technique which has been applied in this surface mined area includes (i) the constitution of a new stable topography conformation made from the waste rock, (ii) implementation of a drainage system in order to guarantee stability and minimize erosion processes, and (iii) construction of a clay cover over the waste rock to support Atlantic rainforest plant species and, mainly, to isolate the waste rock from the water infiltration, ensuring that the rainfall does not come into contact with the sulfides present in it.

Despite that clay cover was not introduced at the bottom of the pond, it was decided even so to carry out the treatment of the pond water. This treatment was applied to the acidity stored in the water ponds, and calcium carbonate ($CaCO_3$) was used to neutralize them.

It was not possible to perform interventions on pond 1's banks due to its margins already being occupied by industrial structures, as shown in Fig. 2.

Monitoring strategy (data and periods)

The water monitoring plan in Area IV's surface is carried out in lentic (ponds) and lotic (rivers) environments (Fig. 3). There are four ponds in Area IV, in each pond two samples are collected, one at the water surface level and the other at half of water column. The samples of the intermediate portion of the water column are collected using the *Van Dorn* sampler (Boiten, 2008).

Surveys are also performed at one point of groundwater and four points located in upstream and downstream area rivers (Fig. 3). Table 1 presents the label of the points, mean sea levels and pond depths, the number of campaigns performed, and their coordinates.

Fourteen monitoring campaigns were conducted. Campaign 0 was carried out prior to the start of the area reclamation, during the environmental diagnosis of the area, which took place in 2010. Throughout this research, the company hired by the Federal government to carry out the diagnosis used only a few points to characterize surface water. For a better understanding of the qualitative and quantitative hydrological processes in the area, the Geological Survey of Brazil team decided to insert additional monitoring points from the start of the reclaiming operation.

Campaign 1 was carried out after the beginning of the area's environmental reclamation, but before the ponds' neutralization.

Analytical data

In the field, the pH, dissolved oxygen (mg L^{-1}), electrical conductivity (μS/cm), and temperature (°C) parameters were measured with a multiparameter *Aquaread AP-800* probe. From the seventh campaign onwards, the oxidation/reduction potential (mV) parameter was also monitored. In rivers, streamflow is also measured using a Sontek acoustic current meter, *FlowTracker* equipment (Sontek, 2019). Groundwater sampling was conducted through low flow method (Puls & Barcelona, 1996).

In the ponds, the water level was recorded with staff gauges and the estimation of water turbidity was assessed with a Secchi Disk. Once collected, the
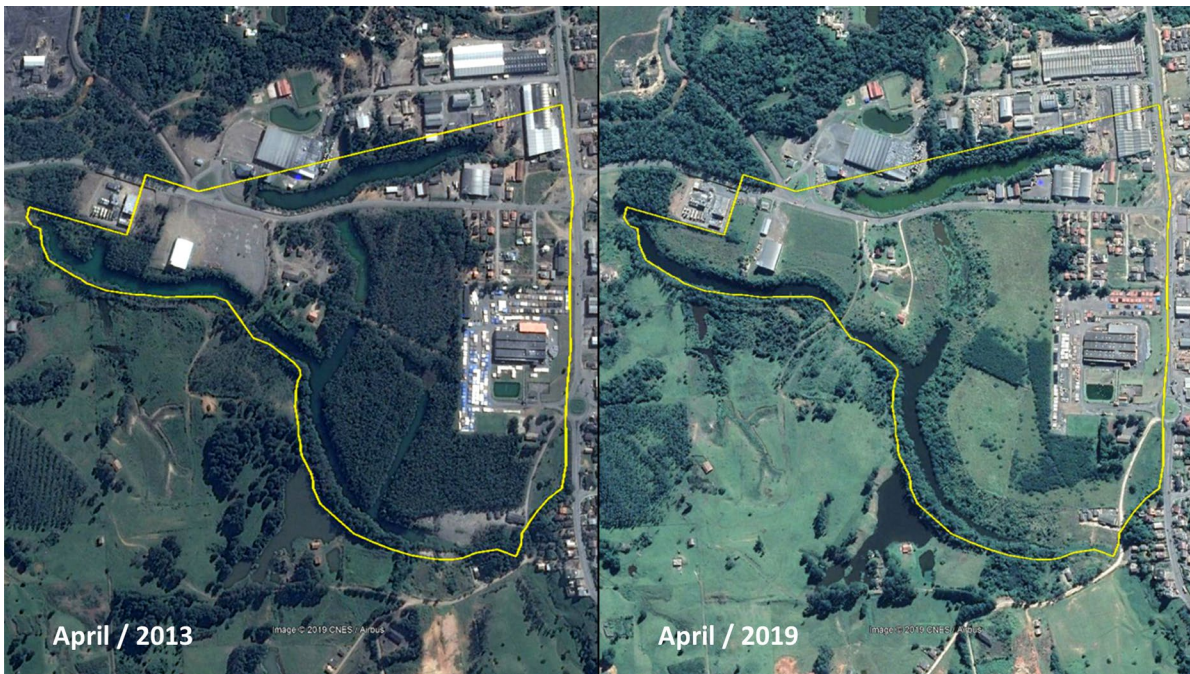
Fig. 2 Area IV before, April 2013, and after reclaiming process, April 2019 (Earth, 2019)

samples were sent to the Mining Pollution Control Center laboratory (*Centro de Controle de Poluição Mineral*—CECOPOMIN) of the Geological Survey of Brazil for analysis of the parameters listed in Table 2.

To check the laboratory data consistency, technical QA/QC procedures were performed following the Brazilian Geological Survey standards based on duplicates and comparative tests with data from similar areas, trend analysis, mass fluxes calculation, frequency distribution, and regression analysis.

Data analysis

The characterization of water resources was made through graphical and basic descriptive statistical analysis (mean, standard deviation, max, and minimum) of the monitoring data. Initial analysis was performed with data from these descriptive statistics, which presents the data of the entire series in a simplified and summarized manner (Mulholland, 2013). Time discretization of the most relevant parameters was also performed; at this point, the purpose was to track the evolution of the parameters motivated by the interferences conducted in the study area.

One hundred and sixty-nine (169) synoptic water samples were analyzed for this study, sampled between the years of 2010 and 2019. In total, 54 were collected in rivers (Table 3), 110 in ponds (Table 4), and 5 in the well (Table 5).

The pH and conductivity parameters were analyzed in duplicate, both in the field and in the laboratory. Due to the difficult sampling conditions and the unreliability of the field equipment calibration in the first campaigns, only values of both parameters obtained in laboratory were considered in this work. However, in a comparison between these two databases, one can see that there are no major differences, as showed in the Supplemental Material.

Regarding the parameters analyzed in the laboratory for this study's analysis, only those with mean concentrations equal or superior to 0.1 mg L^{-1} were used. Thus, the elements Cd, Pb, As, and Hg were excluded since they had an average concentration below 0.01 mg L^{-1}, as well as the elements Cu and Zn, which had average concentrations of 0.02 mg L^{-1} and 0.08 mg L^{-1}, respectively. With these eliminations, the only parameters that remained in the analysis were the ones that, according to Younger et al. (2004), are more

Fig. 3 Surface water sampling points

likely to be found in contamination by open pit mining: Fe, Mn, Al, and sulfate.

Results

The results from the river quality monitoring points (Table 3) showed that the upstream area sampling point, HIDS07, has quite different characteristics from those monitored downstream. At this point, the mean pH was in the neutral range, 7.1; the mean conductivity was around 74.15 μS cm^{-1}; and the other parameter values were below the other rivers' points. In the downstream area point (HIDS10) and in the points located in the Fiorita river (HIDS11 and HIDS12), the water quality was worse and the values of pH, conductivity, and sulfate showed little variation between them. At these three points, mean pH values ranged from 3.3 to 3.6, conductivity ranged

from 397.95 to 467.14 μS cm^{-1}, and sulfate ranged from 136.9 to 151.5 mg L^{-1}.

Although the areas in the most downstream point and the Fiorita river points have similar mean values of some parameters, the mean of manganese and iron concentration is respectively around 40.7% and 56.4% less in the HIDS10.

Figure 4 displays the rivers' temporal evolution of pH throughout the campaigns. These points are located outside the reclaimed area polygon and, for this reason, have not undergone any type of reclaiming intervention. These checkpoints are used to compare the water quality before and after passing through the area. Figure 4 shows that there was no change in the pH parameter trends over time for all points. It is also verified that the HIDS07 point presents pH values on average 3.7 units above the others.

The descriptive statistical results of the points located in the ponds are shown in Table 4.

Table 1 Sampling points, depths, sea levels, monitored period, and coordinates

	Point	Depth (m)*	Sea level (m)*	Sampling campaigns	*Longitude - UTM (E)*	*Latitude - UTM (S)*	Intervention
Rivers	HIDS07	-	-	0** to 13	653011	6835074	-
	HIDS10	-	-	0** to 13	652625	6835951	-
	HIDS11	-	-	1 to 13	652530	6836062	-
	HIDS12	-	-	1 to 13	652475	6835907	-
Ponds	POND01	7.89	95.14	0** to 13	653042	6835866	Neutralized
	POND01B			0** to 13	653042	6835866	Neutralized
	POND02	9.01	93.97	0** to 13	652577	6835685	Neutralized
	POND02B			0** to 13	652577	6835685	Neutralized
	POND03	1.71	95.74	1 to 13	653,008	6835719	Neutralized
	POND03B			1 to 13	653008	6835719	Neutralized
	POND04	8.27	96.25	0** to 13	652932	6835516	Neutralized
	POND04B			0** to 13	652932	6835516	Neutralized
Groundwater	WELL***	-	93.73	9 to 13	652956	6835791	-

For improved readability, following in the text, the pond's samples collected at surface level are in capital letter and the acronyms of the ponds with B at their end means that these are the points that were sampled at half water depth

*The sea level and pond reference depth were established based on the average of the measurements taken across all campaigns; **Campaign 0 refers to the samples taken during the environmental diagnosis of the area, prior to the beginning of the reclaiming process. This campaign was carried out in just a few sampling points; ***Groundwater monitoring started in campaign 9, after the construction of the well

Thirteen campaigns were carried out in pond 3, while fourteen in the others.

In ponds 1, 2 and 4, the samples collected at half depth exhibited poorer quality than the surface ones. In pond 3, however, these values are close, which is probably related to its low depth (Table 1).

When comparing ponds, it is perceptible that pond 1, downstream of the area, presents worse quality in its water samples, with low pH and high values of other parameters. Pond 2 has intermediate quality, while 3 and 4 stand out as having the highest quality.

Pond 1 is highlighted by its low mean pH values, 3.3 and 3.1, and by the greater values of the other parameters.

Pond 2 has the highest concentration of manganese; however, it is only slightly higher than the others.

Considering the ponds' pH values, it is noticeable that the standard deviation of pond 1 is considerably

Table 2 Parameters analyzed in the last campaign

Parameter	Minimum value detectable	Analysis method
pH (23 °C)	0.1	Potentiometric
EC—electrical conductivity (µS.cm^{-1} 23ºC)	0.001	Conductivity
Acidity (mg CaCO$_3$ L^{-1})	1	Potentiometric
SO$_4^{-2}$—Sulfate (mg L^{-1})	0.1	Gravimetric analysis
Fe—total iron (mg L^{-1})	0.01	Atomic emission spectrometry
Al—total aluminum (mg L^{-1})	0.01	Atomic emission spectrometry
Mn—total manganese (mg L^{-1})	0.002	Atomic emission spectrometry
Cu—copper (mg L^{-1})	0.002	Atomic emission spectrometry
Pb—lead (mg L^{-1})	0.005	Atomic emission spectrometry
As—arsenic (mg L^{-1})	0.002	Atomic emission spectrometry
Hg—mercury (mg L^{-1})	0.0003	Atomic emission spectrometry
Cd—cadmium (mg L^{-1})	0.002	Atomic emission spectrometry
Zn—zinc (mg L^{-1})	0.005	Atomic emission spectrometry

Table 3 Summary of descriptive statistics of streams

	Point	HIDS07	HIDS10	HIDS11	HIDS12
	n	14	14	13	13
pH	Mean	7.1	3.3	3.6	3.5
	S	0.4	0.2	0.3	0.3
	Min	6.6	3.0	3.1	3.2
	Max	7.7	3.6	4.1	4.1
EC	Mean	74.15	467.14	397.95	421.68
	S	10.22	150.35	127.40	139.68
	Min	56.00	178.10	210.40	231.50
	Max	89.50	782.50	631.00	700.80
Acidity	Mean	7	95	77	76
	S	6	35	20	22
	Min	1	25	51	35
	Max	24	166	111	111
SO_4^{2-}	Mean	9.4	136.9	143.4	151.5
	S	6.1	43.6	52.4	48.7
	Min	0.4	65.9	67.4	75.4
	Max	23.9	246.1	267.5	258.5
Fe	Mean	0.50	3.89	8.96	8.87
	S	0.34	2.51	4.00	3.25
	Min	0.00	1.40	3.10	3.67
	Max	1.25	11.10	17.70	15.20
Mn	Mean	0.08	0.99	1.63	1.71
	S	0.10	0.44	0.65	0.61
	Min	0.01	0.40	0.75	0.78
	Max	0.31	2.27	2.93	2.82
Al	Mean	0.10	5.24	4.79	4.62
	S	0.11	2.00	1.52	1.49
	Min	0.00	1.73	2.75	2.74
	Max	0.37	10.60	7.39	7.09

smaller than the one observed in the other ponds, 0.2, for the surface and subsurface samples. In regard to other ponds the standard deviation varied from 0.6 to 0.9, gradually increasing from pond 4 to pond 3, then 2. In Fig. 5, it is possible to observe the ponds' average pH values (bar graph) and the dispersion of the values around the average (points), as well as the sampling values of campaigns 0 and 1 (black filled circles), carried out before the ponds' neutralization.

Analyzing the temporal evolution of pH values in the ponds across the campaigns (Fig. 6), it is noted

that even after the neutralization, which occurred between the first and the second campaigns, pond 1 did not present changes in this parameter. This stagnation has not happened in the other ponds, which had high pH values after the application of the neutralizer. Figure 6 shows that ponds 3 and 4 had their pH elevated in the campaign following the usage of the neutralizer, while pond 2 had its pH gradually increased in the campaigns that followed the intervention.

Pond 1's pH stagnation behavior over time was also observed in the other tracked parameters, indicating that the environmental intervention in the area did not influence the water quality of this pond.

The results of groundwater monitoring are shown in Table 5. As the well monitoring began at campaign 9, it was not possible to compare the effect of pond's neutralization in the groundwater. Along the five groundwater campaigns carried out, quality parameters did not show substantial changes or trends in their results. The standard deviations of pH, EC, acidity, sulfate, and Mn were around 18% of the mean, while all Al values were lower than the detection limit of the analytical method. Fe values showed the greatest oscillation; the standard deviation for this parameter was close to 60% of the mean value, with Fe values ranging between 77 and 222 mg L^{-1}.

Discussion

The environmental monitoring and assessment revealed that the area's upstream and downstream river sampling points did not display any trend after the reclamation efforts. However, despite being subjected to the same reclaiming procedures, the surveyed ponds showed different results, particularly ponds 2, 3, and 4 whose water quality improved significantly after the reclaiming process.

Figure 7 shows the evolution of pH and acidity in the area combined with flow of surface runoff, ponds, and rivers (only the surface pond data was used for this analysis). An improvement of these parameters is noted in the graph after the neutralization in the three first ponds: 4, 3, and 2, while pond 1 and the

Table 4 Summary of descriptive statistics of ponds

	Point	POND01	POND01B	POND02	POND02B	POND03	POND03B	POND04	POND04B
	n	14	14	14	14	13	13	14	14
pH	Mean	3.3	3.1	5.0	5.1	6.2	6.2	6.6	6.2
	S	0.2	0.2	0.9	0.9	0.8	0.8	0.6	0.7
	Min	3.0	2.7	3.7	3.6	3.5	3.6	4.7	4.2
	Max	3.6	3.6	6.4	6.2	6.7	6.9	7.2	6.8
EC	Mean	425.97	668.86	207.44	235.95	123.41	125.21	97.51	105.85
	S	114.27	300.33	98.38	107.13	92.29	90.68	34.72	50.48
	Min	257.70	292.40	91.20	91.50	72.10	72.10	70.00	74.30
	Max	645.30	1282.80	389.50	437.00	426.30	420.80	204.80	265.30
Acidity	Mean	89	155	23	29	18	18	10	17
	S	33	95	19	22	18	18	7	8
	Min	58	78	1	5	4	5	2	8
	Max	175	409	70	77	70	73	32	29
SO_4^{2-}	Mean	121.3	198.8	83.4	87.7	31.8	36.5	24.8	26.4
	S	33.9	108.2	37.4	33.4	26.0	25.7	16.7	22.0
	Min	75.3	93.8	23.1	29.2	8.6	14.0	4.8	9.1
	Max	200.9	449.1	160.9	150.0	110.1	111.3	66.5	82.1
Fe	Mean	3.62	25.79	0.47	1.49	1.84	1.74	0.53	1.39
	S	2.26	41.82	0.26	2.92	1.84	0.94	0.41	1.79
	Min	0.92	2.17	0.00	0.00	0.00	0.35	0.00	0.00
	Max	8.60	159.00	1.11	11.06	6.97	3.88	1.50	6.72
Mn	Mean	1.01	1.20	1.16	1.64	0.45	0.48	0.33	1.29
	S	0.47	0.40	0.92	1.15	0.66	0.65	0.42	1.35
	Min	0.55	0.71	0.18	0.19	0.05	0.06	0.06	0.00
	Max	2.47	2.27	3.14	3.38	2.53	2.50	1.59	4.38
Al	Mean	4.68	7.46	0.48	0.42	0.39	0.39	0.25	0.32
	S	1.48	3.47	0.63	0.67	0.72	0.75	0.32	0.47
	Min	2.55	3.23	0.00	0.00	0.00	0.00	0.00	0.00
	Max	8.66	15.00	1.92	2.01	2.59	2.69	0.95	1.37

downstream point, HIDS10, remained with the same characteristics that were identified in the diagnosis period.

Table 5 Summary of descriptive statistics of the well

	n	Mean	S	Min	Max
pH	5	5.6	0.3	5.1	6.0
EC		454.40	35.77	430.00	516.00
Acidity		169	30	142	219
SO_4^{2-}		127.0	8.8	115.7	137.5
Fe		109.32	63.25	77.00	222.00
Mn		0.534	0.088	0.390	0.618
Al		<LD	<LD	<LD	<LD

$<LD$ lower than detection limit

It is possible to deduce that the employed environmental reclaiming method was efficient in neutralizing the water acidity in three out of four ponds. However, the method was not effective for neutralizing the downstream water course, as the stream that receives the contribution from the area persists with its water quality compromised in terms of pH and acidity.

Based on the presented data, it is possible to assess that the reclaiming problem occurs before the water arrives in pond 1. Through studies, we can make the following assumptions from this outcome:

The first hypothesis, Hypothesis 1, concerns the leaching of surface water around the pond. As the area around pond 1 was occupied by industries before the
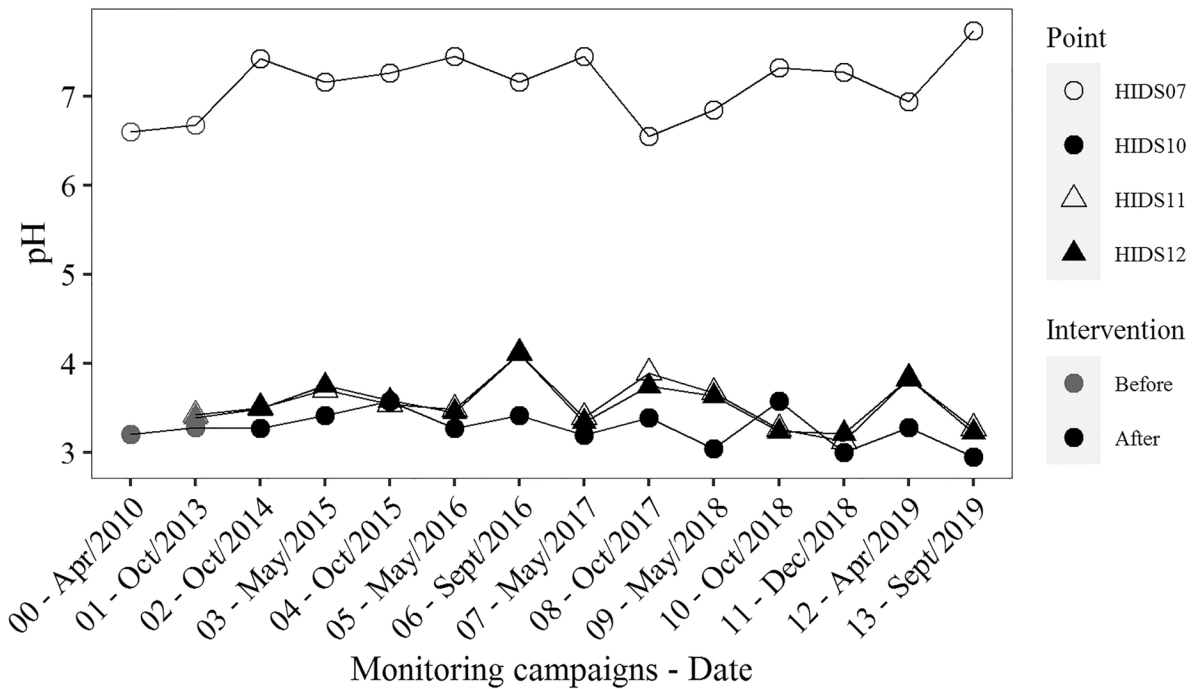
Fig. 4 Temporal evolution of pH at river sampling points

reclamation, there was no intervention in these places. The runoff from these banks would be leaching the material into the pond. Even though the presence of tailings was not found in the area, the diagnosis report (IPAT/UNESC, 2010) points out that the wast rock

found there have a high acid generating potential; the results showed negative neutralization potential ($\bar{x}$ $=-5.9$ kg CaCO$_3$/tonne), positive acid-producing potential ($\bar{x} = 7.3$ kg CaCO$_3$/tonne), and high levels of sulphur content ($\bar{x} = 23.3\%$). In this situation, as the reaction

Fig. 5 Mean (gray rectangle) and dispersion (circles) of pH between ponds. The black filled circles refer to campaigns 0 and 1, which respectively occurred prior to the ponds' intervention and prior to neutralization
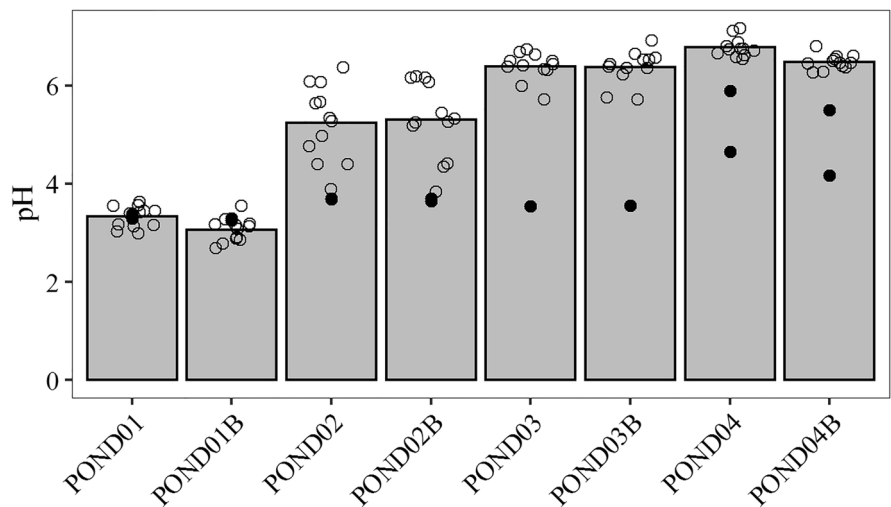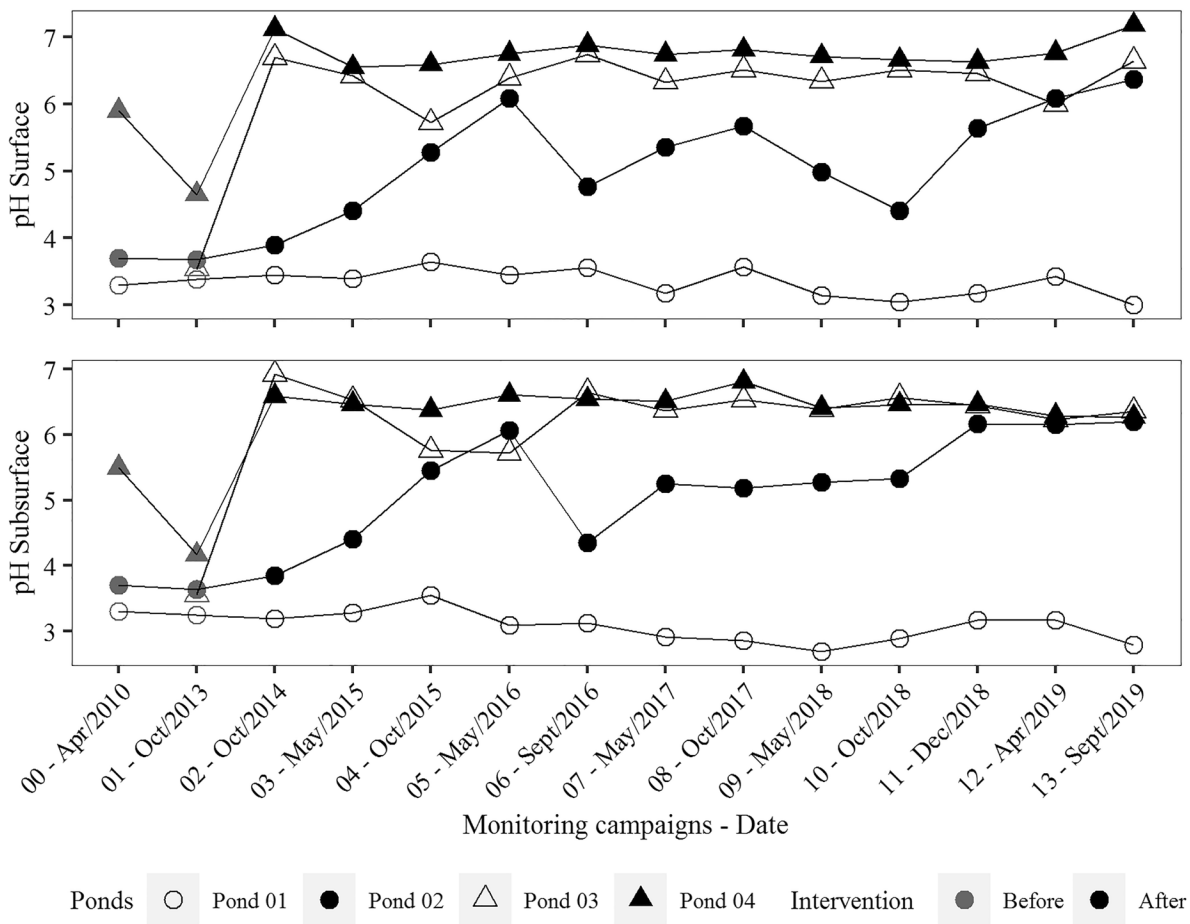
Fig. 6 Temporal evolution of pH in the ponds

takes place in the presence of atmospheric air, the pyrite is oxidized by oxygen, as detailed by Nordstrom (1982).

Another hypothesis, called Hypothesis 2, is related to contamination by groundwater. The groundwater in the area flows in the southeast-northwest direction and pond 1 receives this outflow. For this reason, the pond is affected by this subsurface flow after it passes through the entire area. Based on the data, a conceptual groundwater flow model of the area, shown in Fig. 8, was established. Mean and standard deviation of the parameters most related to AMD (pH, sulfate and iron) were calculated considering the monitoring values obtained after the ponds intervention. According to Haigh and Kilmartin (2017), the pollutants mobilized by the water inside the submerged mine

spoils, especially in the regions of water level movement, are one of the biggest impacts of past open cast mining. This fact becomes even more relevant when considering the high hydraulic conductivity found in this type of material; Younger et al. (2004) showed that slug and bail testing performed on this material found hydraulic conductivities ranging from 1 up to 10 m/day, 3 to 4 orders of magnitude above values typical for natural materials. With further aggravating factors such as lentic environments and little acid neutralization capacity, as is the case here, the impact of acidic groundwater intake is significant (Hayashi & Rosenberry, 2002). We believe that the quality parameters presented supports the contaminant transport model proposed (hypothesis 2), as a low
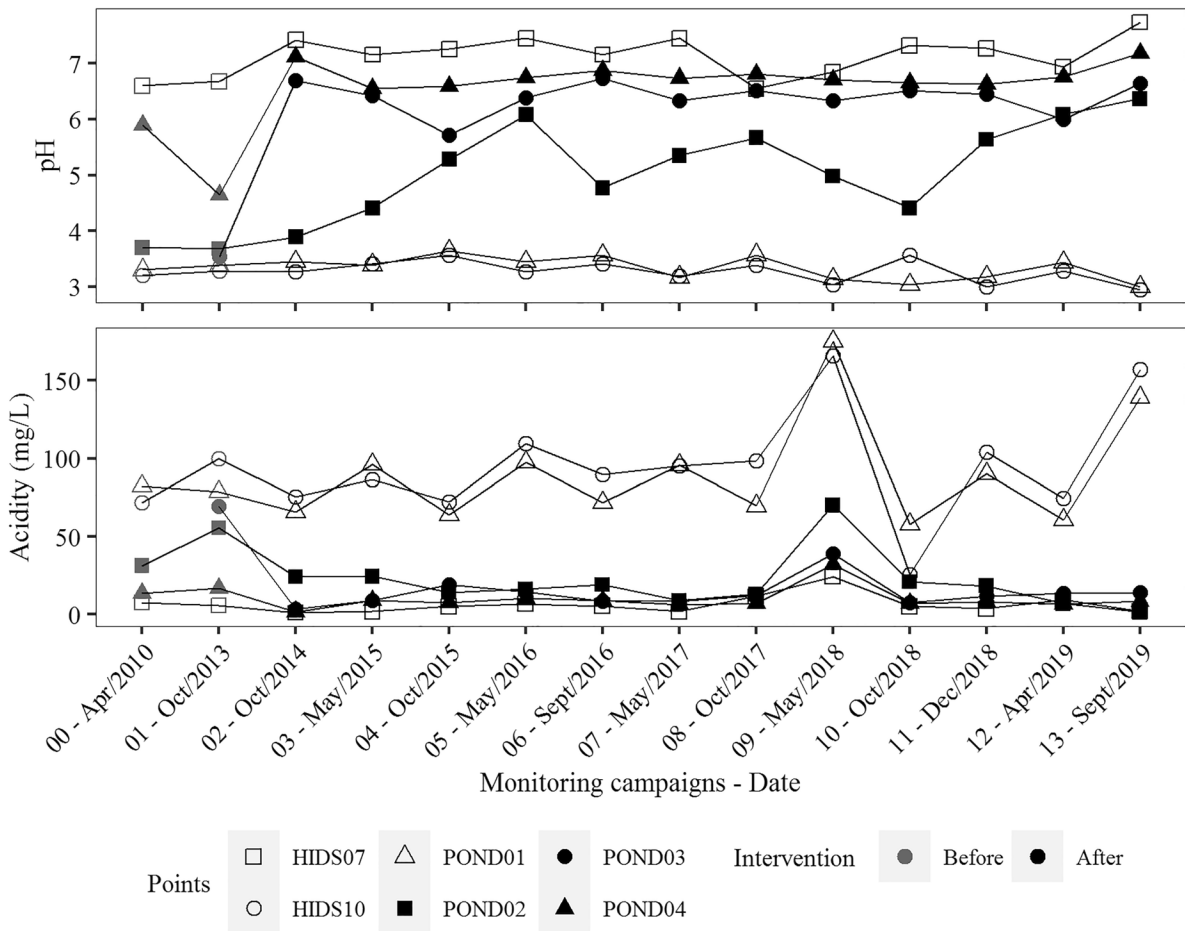
Fig. 7 Temporal evolution of pH and acidity in the combined runoff points of the area

concentration of sulfate and iron is observed in ponds 3 and 4, while in groundwater (WELL) and in pond 1, concentrations increase by a considerable amount, mainly for the samples collected at a half of water column (POND01B).

Considering hypothesis 2 it is possible that the other ponds have improved their water quality for the following reasons: (i) ponds 2 and 4, being located at the upstream limit of the area, do not receive inflow of contaminated water and (ii) pond 3, which is less than 2-m deep, does not receive a contribution from the groundwater flow, but, as observed through monitoring wells, contributes water inflow to this aquifer (Fig. 8).

At the moment it is not consistently possible to prove which hypothesis is correct, as it is also possible that both hypotheses work in conjunction.

The presented limitations in surface water quality improvements are directly related to the method used in the environmental reclaiming. As the main purpose of this method is based on surface isolation with the insertion of a clay cover and groundwater is still allowed to enter laterally in the subsurface regions and to run through the sulfide waste rock material. The urbanized sites are also a problem for the reclaiming strategy because if they are inside the area polygon, it is not possible to intervene in these places, as in the case of pond 1.
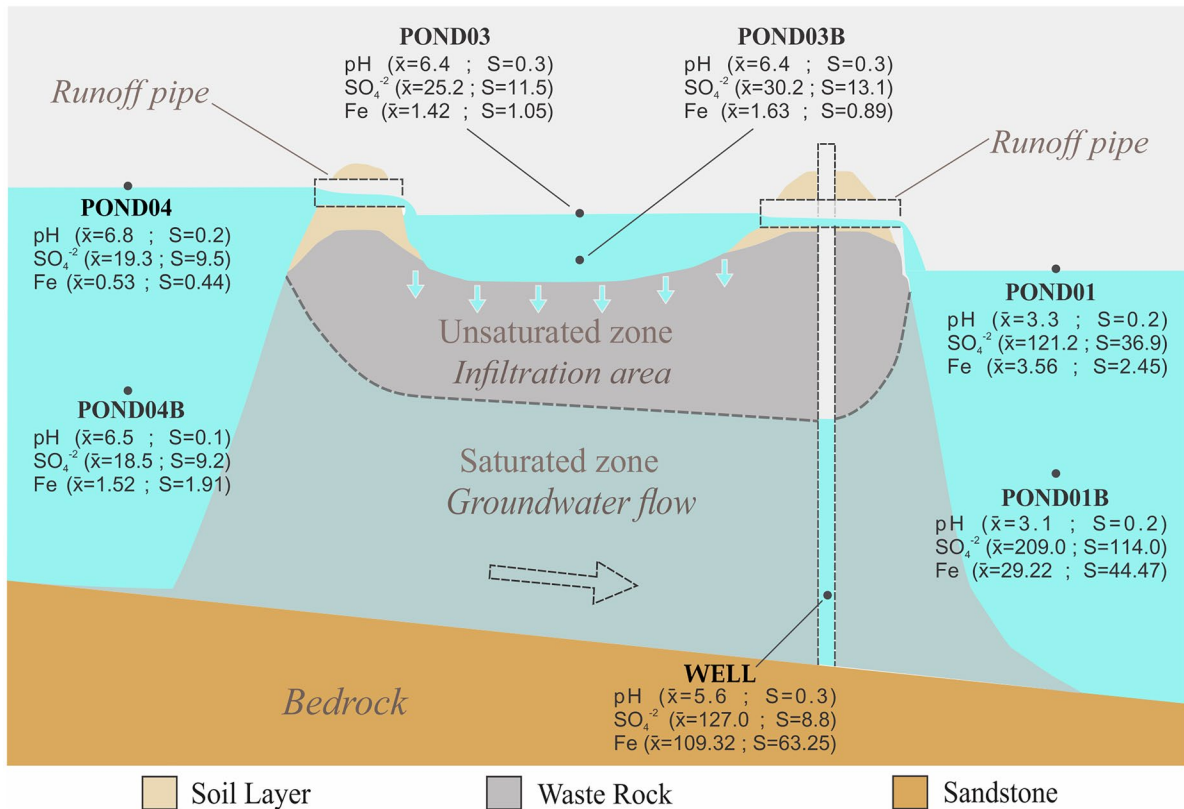
Fig. 8 Hypothesis 2, conceptual contaminant transport model

Conclusions

The study aimed to evaluate changes in water resource quality after an environmental reclamation process in a former open cast coal mining area. Fourteen monitoring campaigns were carried out, one before and thirteen after the reclaiming operation, taking place in the adjacent affected streams and in artificial ponds formed by open cast mining located within its area.

Descriptive statistics showed that upstream and downstream river points have different water qualities, with the downstream points having poorer water quality, still affected by past mining activities even after the reclamation. The temporal analysis also showed that the streams remained with similar concentrations and parameters and were not affected by the reclaiming effort.

Analysis of pond 1, located downstream of the area surface flow, showed that this pond presents the worst water quality of all. The data showed that pond 01 was the only one that did not react to the neutralization process executed during the reclamation endeavors.

Considering the surface flow in the area, it was found that the contamination problem probably occurs before the water enters pond 1. From this analysis, two hypotheses were proposed, operating separately or simultaneously. The first hypothesis is the contamination happens due to leaching of the material that still remains on the on the pond's bank. The other hypothesis is that the contamination comes from the upstream water table inflow into the pond, which runs through the entire area before reaching the pond. It is important that further research analyze these hypotheses, so they can be confirmed or refuted, promoting the new recovery projects that are being developed throughout the country.

From the surface water perspective, the reclaiming method adopted was effective in 3 of the 4 ponds, presenting problems only in the downstream one. The surveyed upstream pond exhibited good quality and steady parameters persisted across all campaigns,

serving as a reference point for the evolution of the other monitoring sites. As the downstream stream in the area is pond 1's outlet, its quality is similar to this one.

Finally, from the perspective of our data analysis, we believe that the reclamation strategies employed in the studied areas must be improved if they are to fully live up to the idea of eliminating environmental issues related to the consequences of past mining activities.

Data availability The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Conflict of interest The authors declare no competing interests.

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