

Water quality of the Gualaxo do Norte and Carmo rivers after the Fundão dam collapse, Mariana, MG

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Abstract

We investigated the effects of Fe-mining tailings of the Fundão dam on water quality, concerning physical and chemical parameters, aiming to infer the recovery process of the affected areas, and prospects of water quality in the coming years. Water quality data from the Gualaxo do Norte and Carmo rivers were evaluated in three moments: (i) before the dam rupture (1999–2000); (ii) one year after the rupture of the Fundão dam (2016); and (iii) after the implementation of recover mitigation activities (2017–2019). Concerning the variables evaluated, the mean concentrations of Fe and Mn were higher in the dry period, and increasing turbidity was detected in the rainy season. The turbidity, even after the recovery measures, peaked at 300 and 400 NTU in 2017 and 2019, respectively. The concentrations of dissolved Fe, Mn and Cr, closely related to the tailings, decreased with rainfall by a dilution mechanism. Conversely, the concentrations of Cd, Al, Zn, and Cu increased in the rainy season, probably by greater erosion. The implementation of the recovery actions at the Gualaxo do Norte headwaters are helping to reduce the Mn and Fe concentrations and turbidity downriver, all associated with the Fe-mining tailings.

1 Introduction

The Minas Gerais state, in southeastern Brazil, extracted more than 406 million ton of iron ore in 2017, which represented 68% of the Brazilian Fe production (DNPM 2019), besides having 81.6% of the country's iron reserves (ANM 2019). However, the volume of ore extracted results in a high environmental liability, due to the amount of tailings generated in the ore processing (Mechi and Sanches 2010; Schaefer et al. 2016).

In the 'Ore Treatment Units'; OTU (place that allocates the residue from the plants to the tailing's dams); consists of successive steps to concentrate iron ore, operate soon after extraction, with ore fragmentation, concentration for flotation, and drainage, in the wet procedure, as in the case of Fundão dam (Quaresma 2009). In this unit, tailings dams are the final destination of the silica rich material without market value (Ghose and Sen 2001; Srivastava et al 2001; Rao et al 2016).

Most mining operations in Minas Gerais state (MG) are categorized as large because the amount of iron ore mined per year exceeds one and a half million tons (ANM 2019). However, mining activities are considered by the environmental agencies as having medium pollution potential, depending on the scale. However, OTU with wet treatment of tailings are considered to have a large pollution potential, due to the presence of large volumes of disposed mot ends in dams.

The Fundão dam, located in the municipality of Mariana, MG, was characterized as a low-risk enterprise with a high associated pollution potential, according to the DNPM's Mineral Dams Registry (National Department of National Production). It was originally designed to receive more than 100 million tons of ore tailings, including fine and sandy tailings (SUPRAM 2008).

In November 2015, after successive adaptations to increase the containment volume, occur the Fundão tailing dam collapse in zone releasing 43.8 million cubic meters of mining waste, directly at Doce River watershed, reaching more than 600 km downstream to the mouth of the Doce River, and the Atlantic Ocean (Marta-Almeida et al. 2016; Golder Associates 2017). The mud plume had high turbulence and, dragged a mix of riverbed materials that had previously lain inert at the river bottom, causing changes in water quality parameters at different levels (Theis et al. 1988; Coimbra et al. 2019). The main characteristics of the plume were low amounts of clay and high silt and sand contents, in addition to low values of heavy metals, with the exception of manganese (Schaefer et al. 2016; Silva et al. 2016).

Since the Fundão tailing dam collapsed, the monitoring of water quality in the Doce River Basin has become a relevant factor in ensuring potability and restoring life in the affected areas. Such monitoring not only refers to the material pertaining to the Fundão dam tailing but also to the riverbed material that has been disturbed and transported in the affected rivers.

Several monitoring and recovery programs have been implemented in the Doce River Basin. The systematic qualitative and quantitative monitoring program (PMQQS); Springs program aiming out recovering more than five thousand springs; Program for the recovery of permanent preservation areas (APP) and water recharge aiming out recovering 40,000 hectares of degraded permanent preservation areas; and Revegetation, and backfill of the floodplains affected, to rehabilitate 2,000 hectares of floodplain and terraces directly affected by of tailings depositions at the Gualaxo do Norte, Carmo and Doce rivers.

It the know that some inert materials can affect water quality, depending on the volume suspended or dissolved in water, affecting riparian communities. In the present work we evaluated the water quality of the Gualaxo do Norte and Carmo rivers in the first zone, just below the Fundão dam burst. The effects the tailings on water quality with, regard to physical and chemical parameters, their interplay and to draw inferences about the recovery process of the affected areas.

2 Materials And Methods

Previous water quality data obtained in 1999 by Costa (2001) of the Gualaxo do Norte and Carmo rivers from was used on pre-collapse reference. For water quality data after the collapse, we used physical and chemical characteristics and spectral behavior of the contaminated waters in the Gualaxo do Norte and Carmo rivers obtained in 2016 by Foesch et al. (2020). It must be emphasized that the upper Carmo river sector before the Gualaxo do Norte mouth was not affected by any tailing deposition after the Fundão dam collapse, representing a reference for local background.

In addition, we collected water samples, monthly, out over the years 2017 to 2019 after the implementation of different recovery activities along the affected areas. Samples were collected at a maximum depth of 30 cm with the aid of a 2 m driller at the channel margin. In situ analyses of physical-chemical parameters (pH, temperature) were performed using a multiparameter probe (PCE Instruments pH Meter PCE-PHD) and collections of simple water samples were carried out according to APHA (2005) at 13 sites distributed in easily accessible places with low disturbances, along the Gualaxo do Norte and Carmo rivers (Fig. 1). In total, 325 collections were made in the 25 field campaigns in the study area.

In the laboratory, of the following physical and chemical characteristics were determined: total suspended solids (TSS), turbidity, and color; and water-soluble: chromium (Cr), copper (Cu), manganese (Mn), iron (Fe), cadmium (Cd), zinc (Zn), and aluminum (Al).

Turbidity was determined using a turbidimeter (model 2100 AN Turbidimeter-Hach; unit: NTU) and the apparent color was determined using an Aqua Color Cor-Policontrol instrument (unit: uC). The TSS analyses were performed using the gravimetric method (unit: mg L^{-1}), in glass fiber membrane (Whatman, 47 mm in diameter, particle retention greater than $1.2 \mu\text{m}$, thickness of $260 \mu\text{m}$). Water samples for the determination of dissolved metals were stored in sterile container. After collection, the samples were acidified with HNO_3 and refrigerated before analysis.

For analysis of dissolved metals, the water samples were filtered through a cellulose acetate membrane (particle retention $0.45 \mu\text{m}$), determine the dissolved metal contents according to EPA 3005A (EPA 1992) by Atomic Spectrometry Laboratory at UFV; using Agilent Technologies 200 Series AA Model 240 FS the contents Cr, Cu, Mn, Fe, Cd, Zn, and Al dissolved in water were determined. Before reading the samples, the spectrometer calibration curve was used. The detection limit was performed according to González and Herrador (2007), were determined based RDC n° 166 (Brasil 2017) on the inclination of the calibration curve and the standard deviation of the standard deviation of the intercept with the Y axis of three calibration curves. For each evaluated parameters, and all analyses were performed on a single sample.

In the study area there is less rainfall in winter when compared to summer, in the months of October to March the precipitation is more than 100 mm per month, already in the dry period that goes from April to September less than 100 mm per month, and the average annual precipitation is 1804 mm, for the history data (Pedreira and Souza 2011). For the study, period rainfall in 2017 was 1048,3 mm, 283,2 mm in dry and 765,1 mm in wet season; in 2018 was 1538,9 mm, 219,7 in dry and 1319,2 in wet season; in 2019 was 1049,65 mm, 206,05 mm in dry and 843,6 mm in wet season. Data analysis was performed considering the dry season (April to September) and rainy season (October to March). The 13 sampling sets were grouped into three sectors: the Gualaxo do Norte River (eight points); the Carmo River upstream (unaffected), the Barra Longa city (three points); and Carmo River downstream (affected) (two points).

Descriptive statistics (mean, standard deviation, and coefficient of variation) of the variables sampled in the period from 2017 to 2019 were obtained, considering the grouping by river sectors, and the averages were assessed by the Kruskal-Wallis non-parametric test at a probability of 0.001, since for this test assumptions such as the normality of the variable, homogeneity of variances between treatments are not required. To identify the impact of recovery activities, principal component analysis (PCA) was performed to select the quality parameters, grouping the collection sites according to their characteristics and seasonal period. In the PCA, 50 samples were used for the Carmo River downstream (affected), 34 in the dry period and 16 in the wet period; 200 samples for the Gualaxo do Norte, 136 in the dry period and 64 in the wet period; and 75 samples for the Carmo River upstream (unaffected), 51 in the dry period and 24 in the wet period. These analyses were carried out separately for the two periods evaluated: shortly after the collapse (Foesch 2020) and after the recovery actions.

The recovery activities implemented were the impediment of access to animals and people in the areas of springs and Permanent Preservation Areas (PPA), allowing natural regeneration, along with revegetation with native species, these actions took place throughout the area of the Doce river basin. The species used in revegetation started with the planting of legumes, such as *Crotalaria sp.*, *Mucuna aterrima*, *Cajanus cajan*; grasses: *Brachiaria* and *Pennisetum purpureum*, after this phase of biomass insertion, native tree species such as *Acacia mangium* were planted. The mud deposit areas, which comprise the areas of riverbed and banks of the Gualaxo do Norte, Carmo and Doce rivers, including its trainers and tributaries, in the municipalities of Mariana, Barra Longa, Rio Doce and Santa Cruz do Escalvado, not only the revegetation of the PPA was carried out, but also the revegetation in the entire deposit area of the dam sediment, also installed rocking of stones in the intricacies, drainages of the terraces with installation of bio blankets to reduce the speed of water and surface transport of sediment to the river (Renova 2019).

The evaluated parameters were compared with the Maximum Allowable Value (MAV) of the current legislation, which classifies water studied as class 2 (CONAMA 2005).

3 Results

3.1 Descriptive statistics

Among the physical and pH parameters evaluated, the temperature values had the lowest values of the coefficient of variation (COV), unlike turbidity, apparent color, and TSS, in sectors that received the tailing loads from the dam. At Fundão, Gualaxo do Norte, and downstream Carmo, COV values greater than 100 were recorded (Table 1). The mean water temperature in the rainy season was $28 \text{ }^\circ\text{C}$ the three sectors evaluated, while in the dry period it varied among the different sectors.

In general, the chemical parameters evaluated showed great variability, and differed between the dry and rainy seasons; in the rainy season, Cu, Mn, Zn, and Al had greater variability in the three sectors evaluated. The values of dissolved Cr were those that showed the least variability, with close values of COV in the dry and rainy seasons.

The mean concentrations of Fe and Mn were higher in the dry period, and decreased in the rainy period probably by dilution. The dissolved amounts of Cr, Fe, and Cd showed the least variability in both periods. However, in the most affected Gualaxo do Norte River and the downstream Carmo River sectors, the variability of Fe increased in the dry period.

The concentrations of Cr and Cu were higher in the dry period than in rainy period. Maximum dissolved Cr concentrations of 0.093, 0.137, and 0.130 mg L^{-1} and minimum dissolved Cr concentrations of 0.011, 0.001, and 0.004 mg L^{-1} , respectively, were found in the upstream Carmo River sectors (unaffected) in the rainy season and in the Gualaxo do Norte River and the downstream Carmo sector in the dry season.

Table 1

Mean, standard deviation (S.D.); coefficient of variation (COV); maximum (max.) and minimum (min.) values of the variables monitored from April 2017 to November 2019 in the upstream stretch of the Carmo River, the Gualaxo do Norte stretch, and the downstream stretch of the Carmo River after the confluence with the Gualaxo do Norte River

River	Season	Statistic	Temp.	pH	Turb.	Color	TSS	Cr	Cu	Mn	Fe	Cd	Zn	Al
			°C		NTU	mg P L ⁻¹	mg L ⁻¹							
Carmo upstream	Rainy	Mean	28.37*	7.66	55.53*	86.08*	47.69*	0.038	0.029*	0.093	0.197*	0.057	0.096	0.10
		S.D.	2.96	0.70	94.25	126.73	89.12	0.016	0.074	0.171	0.167	0.049	0.120	0.17
		COV	10.42	9.09	169.73	147.23	186.86	41.74	254.50	185.12	84.69	86.43	125.14	169.
		Max.	35.00	8.78	330.50	402.50	341.20	0.093	0.359	0.594	0.579	0.109	0.504	0.63
		Min.	25.00	6.62	2.26	6.40	0.100	0.011	0.001	0.001	0.015	0.001	0.004	0.00
	Dry	Mean	24.32*	7.22	6.67*	17.10*	5.80*	0.045	0.037*	0.058	0.089*	0.054	0.091	0.05
		S.D.	2.78	1.57	7.81	13.45	8.90	0.014	0.035	0.040	0.060	0.049	0.149	0.08
		COV	11.43	21.76	117.19	78.67	153.42	31.40	94.43	68.69	67.13	89.85	164.24	160.
		Max.	30.40	9.74	38.50	65.50	43.87	0.088	0.081	0.091	0.246	0.109	0.635	0.33
		Min.	19.10	3.30	1.60	4.05	< 0.001	0.007	0.001	0.001	0.001	0.001	0.001	0.00
Gualaxo do Norte	Rainy	Mean	28.20*	7.55	120.90*	127.18*	72.16*	0.040	0.027*	0.124	0.303*	0.054	0.096	0.08
		S.D.	2.42	0.74	157.58	139.60	131.59	0.018	0.030	0.254	0.221	0.048	0.091	0.18
		COV	8.58	9.82	130.33	109.76	182.35	45.05	113.05	204.28	72.97	89.50	94.60	232.
		Max.	33.70	8.86	659.00	420.50	646.75	0.082	0.081	0.989	0.786	0.109	0.296	1.38
		Min.	24.10	5.71	2.44	4.27	< 0.001	0.002	0.001	0.001	0.018	0.001	0.002	0.00
	Dry	Mean	23.25*	7.23	34.88*	44.27*	12.82*	0.043	0.039*	0.052	0.372*	0.059	0.090	0.09
		S.D.	2.90	1.66	47.96	46.69	19.02	0.021	0.035	0.038	0.528	0.048	0.152	0.17
		COV	12.46	23.03	137.49	105.47	148.28	48.11	89.35	73.09	141.94	80.70	169.47	179.
		Max.	29.60	10.67	349.00	363.00	131.80	0.137	0.081	0.091	3.013	0.109	0.703	0.99
		Min.	17.60	3.08	2.80	5.81	0.625	0.001	0.001	0.001	0.004	0.001	0.001	0.00
Carmo downstream	Rainy	Mean	28.84*	7.53	61.60*	100.15*	44.34*	0.042	0.027*	0.116	0.211*	0.041	0.102	0.07
		S.D.	2.59	0.77	78.25	132.43	69.47	0.016	0.033	0.245	0.187	0.048	0.108	0.12
		COV	8.98	10.17	127.04	132.24	156.69	38.14	123.13	211.59	88.79	118.24	105.73	166.
		Max.	35.00	8.78	330.50	402.50	341.20	0.093	0.359	0.594	0.579	0.109	0.504	0.63
		Min.	25.00	6.62	2.26	6.40	0.100	0.011	0.001	0.001	0.015	0.001	0.004	0.00
	Dry	Mean	25.29*	7.23	16.55*	25.40*	9.01*	0.046	0.039*	0.056	0.161*	0.056	0.099	0.05
		S.D.	3.16	1.82	15.25	16.92	8.50	0.021	0.035	0.041	0.199	0.049	0.165	0.10
		COV	12.50	25.23	92.14	66.60	94.27	46.11	89.68	72.66	123.38	86.82	166.45	186.
		Max.	31.40	10.59	54.00	71.80	37.80	0.130	0.081	0.091	0.765	0.109	0.710	0.44
		Min.	18.90	3.15	2.035	3.60	1.625	0.004	0.004	0.001	0.004	0.002	0.002	0.00

Temp. – temperature (°C); Turb. – turbidity (NTU, Nephelometric Turbidity Units); color – apparent color; TSS – total suspended solids. *Means differ statistically according to the Kruskal-Wallis test at a probability of 0.001. Limits established for Class 2 freshwater by CONAMA 357/2005: pH from 6.0 to 9.0; turbidity up to 100 NTU; dissolved Cu of 0.009 mg L⁻¹; dissolved Fe of 0.3 mg L⁻¹; dissolved Al of 0.1 mg L⁻¹.

Although manganese has the highest concentration in the dam tailings, its concentration in water was not much increased in the affected sectors, compared with the local background the (upstream Carmo River). There, the maximum mean value for Mn was 0.09 mg L⁻¹, while affected sectors Mn reached 0.12 mg L⁻¹. The maximum concentration was found in the rainy season (0.989 mg L⁻¹), and the concentrations of this element generally increased in the rainy season.

The maximum value of dissolved Cd was 0.109 mg L^{-1} and the minimum was 0.002 mg L^{-1} , and did not differ between the dry and rainy seasons in all three sectors. However, a mean of 0.041 mg L^{-1} was determined in the downstream Carmo River sector in the rainy season.

The content of dissolved Zn in water was above 0.05 mg L^{-1} on average, with higher concentrations in the rainy period with a maximum of 0.710 mg L^{-1} detected in the downstream Carmo River sector in the dry period. Zn was positively correlated with the rainfall 10 and 20 days before the collection of the samples; that is, the increasing precipitation directly increased the concentration of dissolved Zn, either by sedimentary load or remobilization of riverbed sediments.

3.2 Multivariate analysis

Grouping all variables by the rivers sectors, and classifying them into the dry and rainy periods, it appears that in the rainy period 53.5% of the variance is explained by the sum of the first two dimensions, confirming the correlation of the variables Zn, Al, Mn, Fe, color, and turbidity in this cluster (Fig. 3). For TSS, rainfall 10 days, rainfall 20 days (accumulated precipitation 10 and 20 days prior to collection), temperature, Cu, Cd, and Cr, explain 34% of the variance in dimension 1, showing that the effect of the accumulated precipitation values 10 and 20 days prior to sample collection affects the temperature and dissolved amounts of Cu, Cd, and Cr. In this case, the ellipses are very similar, which indicates that the three sectors evaluated are similar from the point of view of the evaluated parameters, consistently with the descriptive analysis of the data.

In the dry season (Fig. 3) the behavior of the upstream and downstream Carmo River sectors are similar, and distinguished the Gualaxo do Norte River. The variations of variables TSS, turbidity, Color, rainfall 10 days, and Cu are explained by dimensions 1 and 2. Fe and Al are explained by dimension 1, which distinguishes the Gualaxo do Norte River from the other sectors evaluated in the dry period. The other variables are explained by dimension 2, and for Cd and Zn, the vectors are smaller in both dimensions, showing lower load values. Two dimensions explain 49.8% of the variance.

When performing the PCA for the period from April to September 2016 (Fig. 4), for the same rivers evaluated, but at different points, it was possible to verify the relationship of the variables Cd and Cr, with their variances being better explained by dimension 2 than by dimension 1. We also observed high ratio values of some chemical parameters such as Fe, Cu, Al, and Mn with the physical variables of turbidity and color, explained by dimension 2.

4 Discussion

The reduction in the concentration of Fe and Mn in the dry period is attributed to lower metal dilution effect, increasing the concentration of metal dissolved in water (Dornfeld 2002), and can be influenced by mud tailings from the Fundão dam, since it did not affect the upstream Carmo River sector (Golder Associates 2017). The dissolved Cd, on the other hand, had a high variability in the rainy period in the downstream Carmo River sector, which may be associated with the greater release of domestic effluents by the Barra Longa city (Ulmgren 2000; Appiah-Effah et al. 2015).

Similar temperature values were reported by Marques and Barbosa (2001), who evaluated the water temperature in the Doce River, and Silva et al. (2018), who evaluated trace elements in water and sediment before and after the Fundão dam failure in the periods of April 2015 and April 2016.

High water temperatures can influence the aquatic plant and animal life. Abowei (2010) reports that the temperature of tropical waters generally varies between 25 and 35 °C. Thus, it can be said that this parameter was not directly affected by the rupture and deposition of tailings. The pH values also varied with the season, but it is worth mentioning that pH is an important factor in dissolved metal concentration in water (Costa 2001).

Other physical variables, such as turbidity, TSS, and apparent color, showed high seasonal variability. Turbidity values above 120 NTU were recorded for the rainy season in the Gualaxo do Norte River, above the limit (100 NTU) established by CONAMA 357/2005 for class 2 waters. However, a peak of 659 NTU was observed in the Gualaxo do Norte River sector, consistent with Fernandes (2017). In both upstream and downstream Carmo River sectors, the average values were below this limit in both seasons.

Regarding TSS, a value of 646.75 mg L^{-1} was recorded in the Gualaxo do Norte River in the rainy season, the maximum value found in the three sectors. CONAMA 357/2005 does not consider this parameter for water quality. Although there is a linear relation between TSS and turbidity, we observed lesser association in the dry season (Oliveira et al. 2018). Turbidity expresses, the apparent number of particles present in a water body, as determined by a beam of light that passes through the sample, quantifying the density (Toniolo 2016). Four classes are recognized: fairly turbid from 15 to 25 NTU, rather turbid from 25 to 35 NTU, turbid from 35 to 50 NTU, and very turbid above 50 NTU (Azis et al. 2015).

In recent study of Doce River, turbidity values above 400 NTU in the rainy period, and 250 NTU in the dry period were recorded by Oliveira et al. (2018), along with TSS values of 300 mg L^{-1} in the rainy period and 200 mg L^{-1} in the dry period. This highlights the natural high turbidity and TSS of the Doce River basin regardless of the Fundão dam contribution.

The three stretches have similar changes in physical parameters when comparing the measured mean values, however, they have different intensities. While in the Carmo Upstream stretch (unaffected), for example, in the rainy season Turbidity and TSS are 9 times higher than the dry season, in the affected stretches the intensity is lower, on average 3 times higher in the wet period when compared to the dry period.

In general, after precipitation variables such as Color, Turbidity and TSS are changed by increasing their values, this is because these variables are causally related to fine particles. Therefore, when precipitation occurs in the area increases the flow of the river allowing the entry of new sediments and the revolving of the bottom of the river. Even in tropical rivers that high physical variables (Color, Turbidity and TSS) are normal for a few months, changes in the concentration of these variables can occur due to rainfall, allowing the tailings deposited in the margin to be taken to the river trough, favoring resuspension processes (Hatje et al 2017). In the dry period, due to the lower velocity of the rivers, the suspended particles tend to sediment, with this the variables Cor, Turbidity and TSS reduce (Gong et al 2016).

Historically, the water courses of the Doce River basin have high turbidity, as in the case of the Santarém stream, located closer to the Fundão dam, where values of 35.65 NTU were recorded in the rainy season. In the Gualaxo do Norte River, turbidity values were not higher than 43 NTU in the rainy period of 2000 (Costa 2001). However, at the automatic station located in Governador Valadares, an extreme peak of 140,000 NTU was recorded on 11 November of 2015 when the mud plume from the Fundão dam passed through that sector (Maia and Pereira 2017). This confirms the influence of the fine colloidal tailings dam Fundão released, downstream river, below the Risoleta Neves Hydroelectric Plant and dam (Milanez and Losekann 2016; Schaefer et al. 2016).

In the rainy season, there is a natural increase in turbidity, which is a direct contribution of eroded and transported resulting from severe soil loss and reworking of tailings deposited on the floodplain affected by mud (Theis et al. 1988). With the deposition of mud on floodplain, forming technosols (Schaefer et al. 2015). These areas became prone to erosion soil exposure, surface aluvic and little structure of this sediment. This contributed to several increases in turbidity and constant modifications of the fluvial channel by the lag deposition (Schaefer et al. 2015; Santos et al. 2017).

Apparent color values above $127.18 \text{ mg PI L}^{-1}$ were observed in the Gualaxo do Norte River sector, during the rainy season. In both downstream and upstream Carmo River sectors, the mean values were 110.15 and $86.08 \text{ mg PI L}^{-1}$, respectively. These values are well above the maximum allowable value for the water potability standard, which is 15 mg PI L^{-1} according to consolidated Ordinance N^o. 5 of the Health Ministry (Saúde 2017). The increasing in value of apparent color is associated high concentration of suspended particles suspended ($> 1,2 \mu\text{m}$), and closely associated with turbidity, although humic acids and the presence of biofilm can also change the water color.

The dissolved Cr showed values higher than those established in the CONAMA Resolution 357/2005 (Cr to 0.05 mg L^{-1}). Cr may be present in water as trivalent Cr or as hexavalent Cr, the later form being extremely toxic (Kaczynski and Kieber 1993), but there is no speciation study that would allow conclusions on toxicity to be drawn.

With reference to Cu, the mean concentrations of dissolved Cu for the two periods and in the three sectors evaluated were higher than those established for class 2 waters (0.009 mg L^{-1}); an outlier of a dissolved Cu of 0.359 mg L^{-1} was found in the upstream Carmo River sector (Table 1).

With regard to Fe, which is the element with the second highest concentration in the tailings, the waters of the Gualaxo do Norte River showed mean values above the limit imposed by the CONAMA resolution (0.3 mg L^{-1}) for class 2. The maximum value found was 3.013 mg L^{-1} , 10 times above this limit. Overall, in the sectors most directly affected by the mud tailings, the Fe concentration was nearly four times higher than the concentration of unaffected sector. Also, higher concentrations of Fe were observed in the dry season, probably because, in the dry period, the waters have lower pH values. According to Nordstrom et al. (2009), the Fe solubility increases with lower pH values, and correlation analysis (Fig. 2) showed that the pH was negatively related to the values of the Fe metal, suggesting that acidity promoted Fe hydrolysis and release (Guedes et al. 2004), besides the diluting effect.

High Cd concentration in waters can be harmful to plants (Barceló et al. 1986) or human health (Silva et al. 2019). Cd remains in the environment for 10 to 30 years, and possesses slow excretion by humans (Nordberg 1996). On average, the three sectors had dissolved Cd, concentrations above the limit for the total Cd (0.001 mg L^{-1}) (CONAMA 2005) for class 2. In the dry season, the average concentration of dissolved Cd was lower in the downstream Carmo River sector.

Silva et al. (2018) found values of total Cd in the range of $0.65 \mu\text{g L}^{-1}$ at in the Gualaxo do Norte River in 2016, shortly after the Fundão dam collapse, whereas Costa (2001) found values below 0.001 mg L^{-1} in the Gualaxo do Norte River sector in the dry and rainy periods of 2000 (before the dam burst).

Average values of dissolved Zn were below the limit established by CONAMA 357/2005 for total Zn, which is 0.18 mg L^{-1} . Maximum values of total Zn of 0.077 mg L^{-1} were observed in the same Carmo River sector (Silva et al. 2018). Abnormal Zn concentrations were considered natural anomalies by Rodrigues et al. (2013), who detected concentrations between 28.7 and 85.6 mg kg^{-1} in sediments from the Gualaxo do Norte River, before the dam failure.

Regarding dissolved aluminum, the average concentrations in the three sectors and periods were within the allowable range for class 2 freshwater, according to CONAMA 357/2005 (Table 1). However, in the rainy season, an outlier 1.388 mg L^{-1} was recorded in the Gualaxo do Norte River sector. Al is generally present in water in the form of Al^{3+} (aq) under acid conditions, and as $\text{Al}(\text{OH})$ (aq.) in neutral or alkaline conditions. In acidic waters, Al is toxic to fish from 0.1 mg L^{-1} (Gensemer and Playle 1999; Ferreira et al. 2020). In all sectors and periods, the maximum values were above this limit.

Turbidity, color, and TSS are directly and highly correlated with each other and with the Fe content, which can be explained by the fact that Fe-oxides forms gives a reddish color to the suspended sediments present in waters (Moruzzi and Reali 2012), and it is the main element present in the tailings of Fundão.

The pH behavior in monitored period is consistent with that found by Costa (2001), and is poorly explained by both dimensions 1 and 2. In the period evaluated about eight months after the rupture of the Fundão dam, Mariana, MG, it is possible to distinguish three River sectors: Gualaxo do Norte, downstream and upstream Carmo River sectors.

In the dry period of 2017, two years after the accident, the Gualaxo do Norte River sector, closer to the source of tailings from dam, had turbidity, color, TSS, and Fe contents with vectors in the same direction, indicating a close relationship between these parameters in the dry period, unlike the upstream Carmo River sector which did not receive the same load from the dam rupture. The downstream Carmo River, after the confluence with the Gualaxo do Norte River, showed from the influence of tailings, but lower than the Gualaxo do Norte itself (Fig. 5).

When separating the rivers by dry and rainy periods over the three-year period from 2017 to 2019, the physical and chemical parameters with greatest correlation (Fig. 2), the change grouping of rivers and the dimensions of explanatory variables (Fig. 5).

Three years after the dam burst, improvement of water quality at unlike the Carmo River sector can be observed, the behavior of the Gualaxo do Norte, which is distorted affected by Fe, color, and to a lesser extent, turbidity (Fig. 5).

In the dry period in 2019, four years after the dam burst, it is possible to see a similar quality of waters upstream and downstream the Carmo River (Fig. 5). Closer to the dam, the Gualaxo do Norte River still shows abnormal Fe contents in the dry season.

In the rainy season in 2017, the Gualaxo do Norte River was the most affected by the TSS, turbidity, color, and Fe variables, explaining 77.4% of the variance. One year after (rainy period 2018), the discrepancy was smoothed, with the turbidity, color and Mn explaining 87.2% in dimension 1. In the following rainy period (2019), differences remained smooth but, the Fe content was associated with turbidity and color. Here, four years after the rupture, the influence of Mn on the parameters of turbidity and color was reduced.

The improvement in physical and chemical parameters highlights the ability of tropical rivers affected by mining tailings to naturally recover. After implementation of recovery activities at the affected areas, the levels of some parameters were reduced.

On the other hand, the variation on some parameters is strongly influenced by the seasonal accumulated precipitation (Fig. 6). For example, the mean concentration of dissolved Fe was higher in Gualaxo do Norte River at the beginning of the monitoring, while in the Carmo River sectors peaks of dissolved Fe and Mn occurred just after precipitation events, at the end of the first year of monitoring period.

Rainfall influence pH variability (Carvalho et al. 2000; Silva et al. 2008; Girardi et al. 2016), and on average the value increased by 0.5 points in the rainy period when compared to the dry period. However, in a limestone mining area, where CaCO_3 contribute to pH changes increasing pH by the dissolution of carbonates occurs in the dry season, when dilution is reduced (Fritzsos et al. 2009). The pH variation influences other physic-chemical variables (Yabe and Oliveira 1998; Girardi et al. 2016), and greater acidity leads to metal solubility, especially Cu, Mn and Fe (Yabe and Oliveira 1998). However, with greater rainfall, increasing concentrations Pb, Cd, Ni, Cr, and Cu were detected, attributed to soil erosion. Although Fe and Al showed reduced concentration after the implementation recovery activities, peaks in turbidity above 100 NTU continued recorded after the measures.

High, abnormal values of dissolved Fe were recorded before 2015 as contents above 2 mg L^{-1} were detected in 1999. In this stretch, even with the rupture of the Fundão dam, such values were not observed. After the rupture, the highest Fe content was 0.75 mg L^{-1} , and Fe decreased significantly after the recovery activities. Overall, the concentration of Fe increase in dry season with lower water volume (Correll et al. 1982), and occurred either before and after the Fundão dam burst.

The values of dissolved Al peaked in 2016, but decreased after recovery activities, reaching contents similar to pre-rupture values.

Turbidity also decreased after recovery activities, with seasonal peaks observed in early 2018 and early 2019. After the recovery activities, the turbidity values followed the same seasonal trend comparing the three sectors, with an overall reduction due to the implementation of vegetation at the recovery the margins and backfill.

5 Conclusions

The presence a sedimentary lag of tailings on the riverbed of the Gualaxo-Carmo rivers, affected by the Fundão dam burst, has no significant effect on the concentration of dissolved metals in water.

The physical parameters monitored (turbidity, color, and TSS) showed great seasonal variability, closely associated with rainfall.

After the Fundão dam burst, turbidity in the Gualaxo do Norte-Carmo River reached values unrecorded previously, and the Gualaxo do Norte had, turbidity values five times higher than that required for class 2 rivers. On average, the turbidity values were generally higher than those established for River classes 1 to 3 in the Gualaxo do Norte River, and reduced in the Carmo River sector, downstream.

Declarations

Acknowledgments, Samples, and Data

The authors declare that we have no real or perceived financial conflicts of interest.

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Figures

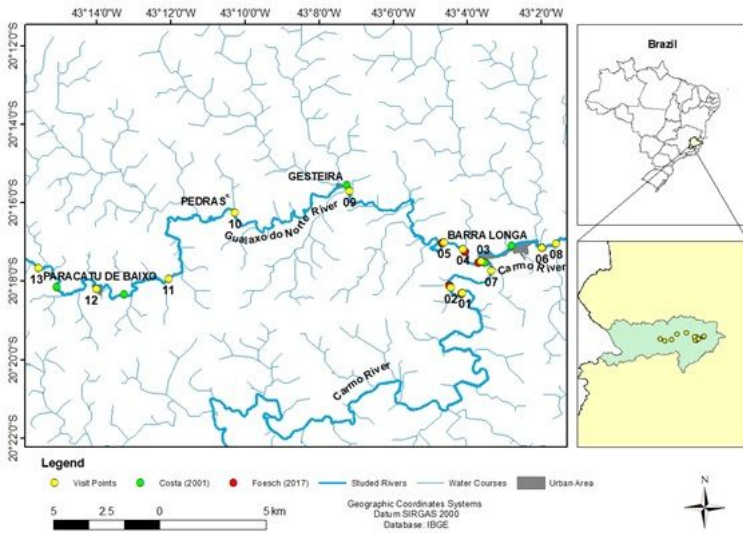


Figure 1
Location of water sample collection points evaluated between 1999 and 2000 before dam collapse by Costa (2001; green), in 2016 by Foesch (2017; red), and between 2017 and 2019 (yellow).

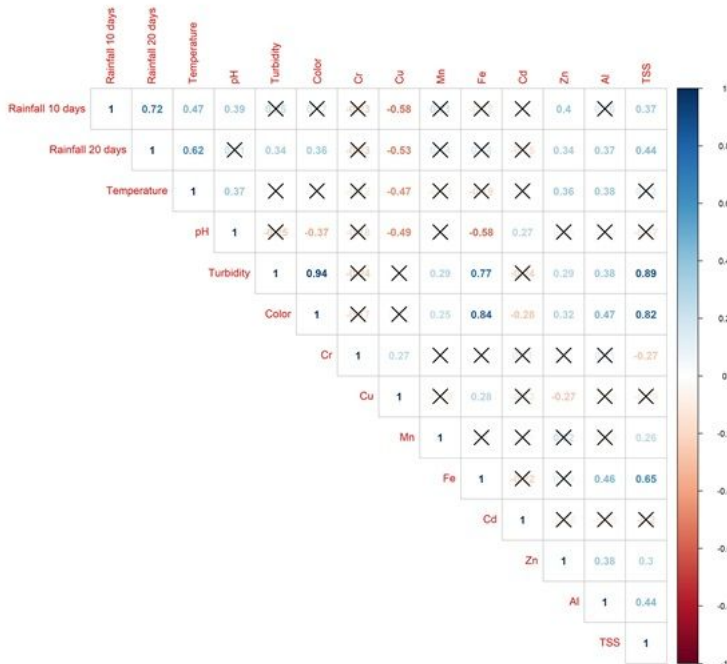


Figure 2
Spearman correlation of the analyzed variables. The X symbol distinguishes correlations that were not significant at 0.005 probability. Al – dissolved aluminum; Mn – dissolved manganese; Cd – dissolved cadmium; Zn, – dissolved zinc; Cr – dissolved chromium; Fe – dissolved iron; pH – hydrogen ionic potential; Sample turbidity is measured in Nephelometric Turbidity Units; TSS – Total Suspended Solids; COR – apparent color of the sample; Sample

temperature. The scale color represents the correlation between the variables, indicating that the closer to blue the more positively correlated the variables are, the closer to red, the more negatively correlated the variables are.

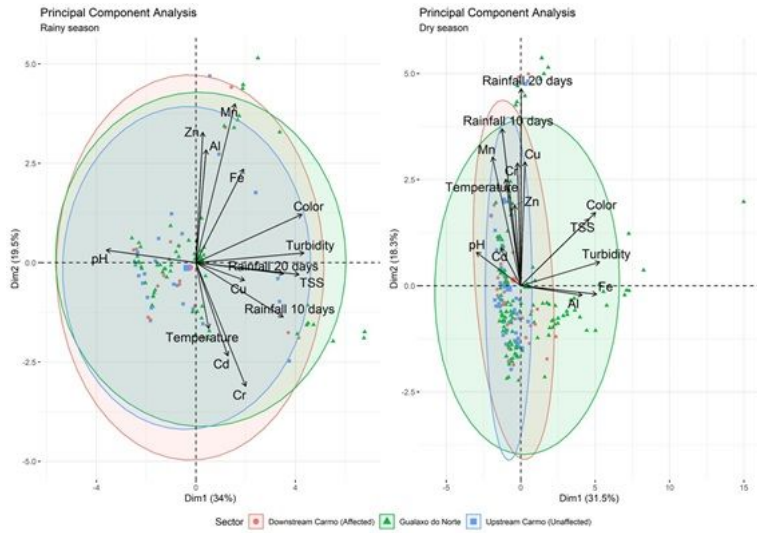


Figure 3

PCA for the rainy and dry season data evaluated from April 2017 to January 2019 grouped by River sectors.

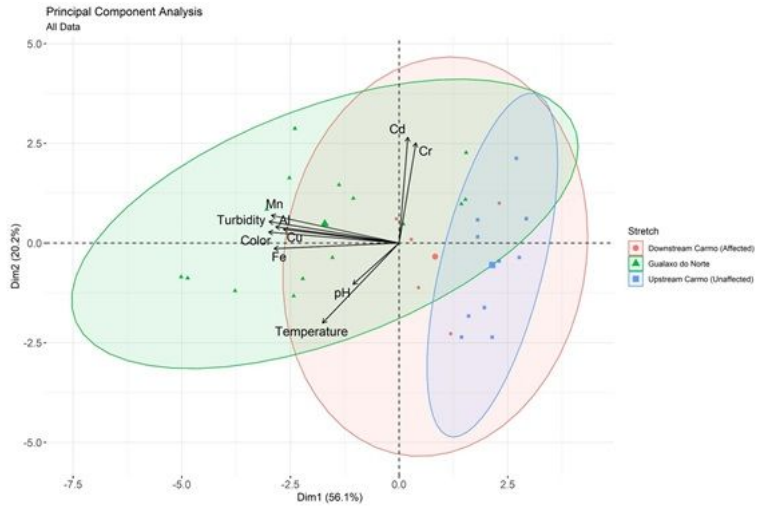


Figure 4

PCA for data collected in the dry period of the year 2016 by Foesch (2020) in the Gualaxo do Norte and Carmo Rivers.

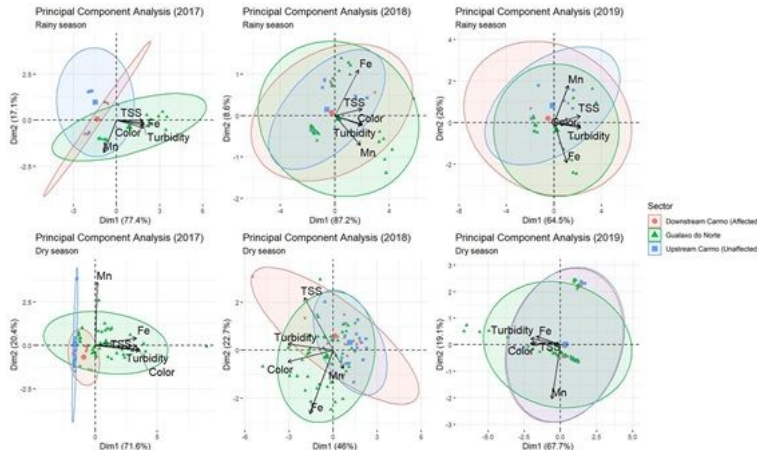


Figure 5

PCA of data collected in the rainy and dry period of the years 2017 to 2019 for only the variables evaluated throughout the period.

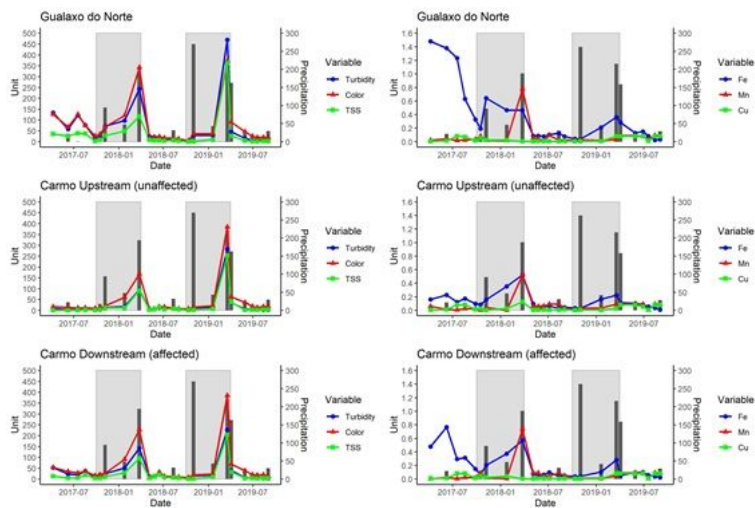


Figure 6

Time series for averages of the physical and chemical variables analyzed six months after consolidation of the recovery services. Precipitation – precipitation accumulated 20 days before sampling (mm); Fe – dissolved iron; Mn – dissolved manganese; Cu – dissolved copper (mg L⁻¹); NTU – turbidity (Nephelometric Turbidity Units); color – apparent color (mg PI L⁻¹); TSS – Total Suspended Solids (mg L⁻¹). Gray stripes distinguish the rainy season from the dry season. The first column shows the physical variables in relation to precipitation and the second column shows the chemical variables in relation to precipitation.

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