



Trace metals in Rio Doce sediments before and after the collapse of the Fundão iron ore tailing dam, Southeastern Brazil

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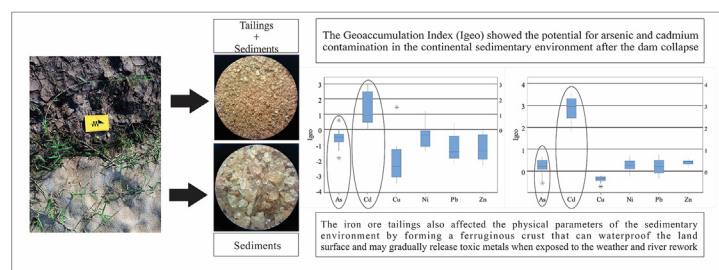
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HIGHLIGHTS

- The collapse of the ore tailing dam caused sediment disturbance and contamination.
- Iron ore tailing introduced cadmium while mudflow released arsenic.
- The results show that arsenic sources were present before the disaster.
- Fluvial sediment contaminants originated from the Fundão Dam and other sources.

GRAPHICAL ABSTRACT



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ABSTRACT

The collapse of the Fundão Dam, in Southeastern Brazil, caused about 50 million m³ of iron ore tailings to sluice down the mountain to Rio Doce, in what is considered the greatest environmental disaster in Brazilian history. The fluvial system received an intense and sudden mudflow that was transported for more than 650 km, before reaching the Atlantic Ocean. Because the area was already impacted by the mineral activities in the region, it becomes essential to evaluate the environmental conditions before the disaster to correctly assess the disaster real damage. This study compares the concentration of trace metals in the sediments of the Rio Doce alluvial plain, before and after the dam collapse, as well as the newly deposited iron ore tailings that became part of the sedimentary framework. The data indicate that the fine particles deposited have since been incorporated into the sandy river sediments. The cadmium and arsenic contents in the sediments increased to levels above the National Environment Council thresholds. The comparison between the levels of trace metals in the situations before and after disaster shows that the mining mud is the source of cadmium while the arsenic was present before the environmental disaster, and its concentration increased due to sediment remobilization. The iron ore tailings

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deposited on the alluvial sediments also affected the physical parameters since the formed ferruginous crusts waterproofed the ground surface and may, gradually, release toxic metals when exposed to weathering and river reworking.

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1. Introduction

The collapse of the Fundão Dam in Mariana, Minas Gerais, Southeastern Brazil, on November 5, 2015, has been deemed as the most significant and tragic environmental episode in the country's history. About 50 million m³ of mud containing iron ore tailing were dumped into the environment (Segura et al., 2016; Carmo et al., 2017). This disaster greatly impacted the equilibrium of the Rio Doce eco-system (Silva et al., 2016; Fernandes et al., 2016; Bottino et al., 2017) due to the intense and sudden mudflow (water and iron ore tailing) that was transported for more than 650 km until reaching the Atlantic Ocean (Marta-Almeida et al., 2016; Miranda and Marques, 2016; Queiroz et al., 2018; Rudorff et al., 2018).

Just over 3 years later, on January 25, 2019, this tragic episode was overshadowed by the collapse of the Brumadinho Dam, located in the homonymous city, also in Minas Gerais. Although the tailings volume released into the environment was much smaller (about 12 million m³), the consequences regarding the immediate loss of human lives were more severe (Passarinho, 2020; Vergilio et al., 2020). A mud wave as high as 30 m buried the administrative offices of the mining company (Cionek et al., 2019; Porsani et al., 2019), inappropriately built on the way of the spill, and left 270 victims among the identified and missing dead (Leocárdio, 2020).

Both dams belonged to the Samarco/BHP/Vale business association and their collapse had several negative economic and legal consequences, regarding criminal liability to the business partners (Carvalho and Almeida, 2018; Ruchkys et al., 2019; Morrison and Gomide, 2019; Lima et al., 2020; Scarpelin et al., 2020). Contrary to the evidence, the Brazilian Environmental Legislation has a sophisticated normative apparatus (Brasil, 2019) that needs, however, to be implemented by the environmental agencies, society itself and followed through by much stricter inspection.

An extremely relevant factor to this assessment is the knowledge of the prevailing environmental conditions before the accident. The region affected by the Fundão Dam tailings spill has a long history of mineral extraction in the State of Minas Gerais (Ecoplan-Lume, 2010; Hatje et al., 2017). Additionally, different land use and urban sewage discharge points have certainly been causing an impact on the environment in the Rio Doce Watershed for several decades. Therefore, for correctly assessing the impacts caused by Samarco/BHP/Vale tailings spill, it is necessary to determine the regional background before the spills. The differential of this work is that we present the metal content in the Rio Doce alluvial sediments before they been affected by the intense mudflow from the dam break.

In 2013, before the Fundão Dam disaster, our research group had sampled alluvial sediments in Rio Doce, sampling that resumed after the tragedy, in 2015, at the same points, seeking to determine the changes caused by the mining tailings deposition on the river sediments. This work determined the concentration of trace metals in the sediments from the Rio Doce alluvial plain, deposited before and after the collapse of the Fundão Dam, as well as the deposited

tailings that became part of the sedimentary framework of this fluvial system.

2. Materials and methods

The Rio Doce Basin in the Southeast Region of Brazil is partially located in the States of Minas Gerais and Espírito Santo, between the 17°45' and 21°15'S and the 39°55' and 43°45'W coordinates (Coelho, 2006) (Fig. 1). The basin is 853 km long with a large drainage area of approximately 83,465 km², comprising a total of 230 municipalities (Coelho, 2009; Guevara et al., 2018).

The sampling points are distributed in the watershed stretch inside the State of Espírito Santo (Fig. 1). In 2013, before the dam collapsed, alluvial sediments were collected between July 3rd and 6th. Then, about six months after the environmental disaster between May 2nd and 5th, 2016, alluvial sediments were sampled again to compare the results of the chemical analysis, textural standards, and the iron ore tailings deposited on the sediments just after the dam collapse (Fig. 2).

Undisturbed samples of fluvial sediments were collected from the alluvial plain of the Rio Doce by inserting 60 mm diameter PVC tubes at 1 m deep (Fig. 2) while sediment samples were gathered with a plastic-covered stainless-steel shovel. Overall, 35 sediment samples were collected, of which 11 are before disaster sediments (BS) and 12 after disaster sediments (AS) plus 12 samples of iron ore tailings (IOT) deposited on river sediments.

Pre and post environmental disaster, the grain size of sediments was determined by dry sieving using an agitator whereas that of the tailings was quantified by laser diffraction analysis using a laser granulometer. For imaging, the samples were metalized in a gold bath and submitted to a scanning electron microscope (SEM), at different magnifications.

The sample mineralogy was determined using the x-ray diffraction technique (XRD), in a goniometer equipped with a copper tube, with the reading interval angle 2θ between 4° and 70°, 0.02° step and 4°/min scan rate. The mineral phases were identified using a specific software for diffractograms and the associated database. The sediments are formed predominantly by coarse sand, rich in quartz and these mineral peaks tend to overshadow the other components, thus, the diffractometric analysis was performed in a fraction of less than 0.075 mm, seeking a better comparison between the mineralogical content of sediment and tailings.

The samples were also submitted to semi total chemical extraction following the EPA 3051A standard (Usepa, 1998). The elements extracted by this acid solution were quantified by optical emission spectrometry with inductively coupled plasma (ICP-OES). The As, Cd, Cu, Ni, Pb and Zn contents were determined and compared with the reference values recommended in Resolution 454/2012 of the National Environment Council, CONAMA (Brasil, 2012), which establishes guidelines for managing the material to be dredged in waters under national jurisdiction. This resolution defines the acceptable concentration ranges of potentially toxic

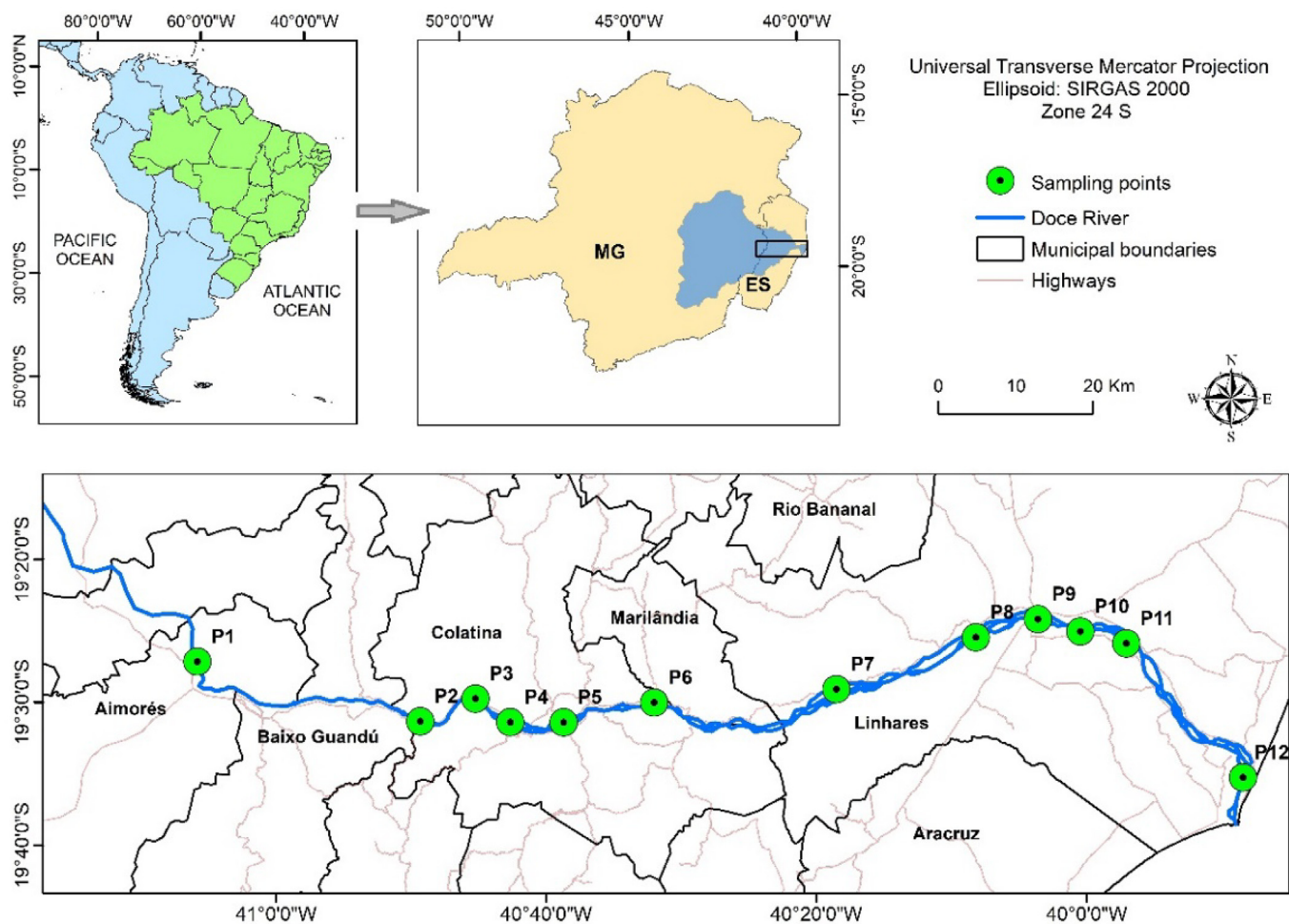


Fig. 1. Sampling points along the Rio Doce in the stretch between the Aimorés (MG) and Regência municipalities, in Linhares, Espírito Santo (ES).

elements when performing environmental impact assessment: Level 1 is the lower limit, below which the likelihood of adverse effects to the biota decreases, whereas Level 2 is the upper limit or the threshold above which adverse effects on biota are more likely.

Analysis quality control was performed using blanks and the certified sample ERM-CC141 - Loam Soil (Trace Elements), in quadruplicate. The recovery rate calculated for each metal varied between 85 and 115% (Supplementary Table S1), which is considered satisfactory for the 3051A Method (e.g. Souza et al., 2015; Guevara et al., 2018).

The geoaccumulation index (I_{geo}) was also calculated. This method assesses the environment contamination by metals and compares the local metal concentrations of the samples to either a reference value taken as a standard (Equation (1)) or a value background (Müller, 1969; Ghrefat et al., 2011) which, multiplied by 1.5, minimizes possible variations of background values for a given metal in the environment.

$$I_{geo} = \frac{\log_2([Me]_{am})}{(1.5 [Me]_{bg})} \quad (1)$$

where:

[Me]_{am} = metal concentration in the sample; [Me]_{bg} = metal background concentration in the study site.

The geoaccumulation index (I_{geo}) classifies metal accumulation

levels in seven different classes, varying from 0 to 6, correlated with the increasing degree of contamination (Supplementary Table S2).

3. Results

3.1. Physical parameters

When the mud and debris flow resulting from the dam collapse slowed down, the particulate matter that entered the Rio Doce alluvial plain formed a mud layer over the river sediments (Fig. 3a), after being partially mixed with the alluvial sediments, as shown below.

The predominantly silt-clayey granulometry of the iron ore tailing (Fig. 3b) entering the river system affected the sediment physical parameters. The Rio Doce alluvial sediments are essentially sandy and remained so after the accident, however, the coarse sand percentage decreased relatively while fine particles appeared in the silt fraction with increasing fine sand fractions (Fig. 3b).

SEM imaging of the alluvial sediment (Fig. 3c) reveals typical characteristics of sandy river sediments, due to intense rework, with rounded, equidimensional, and grooved grains. On the other hand, the mining iron tailing (Fig. 3d) has lamellar minerals, and several associated micro-aggregates in different forms, ranging from filled or hollow acicular habits to even rectangular-shaped features.

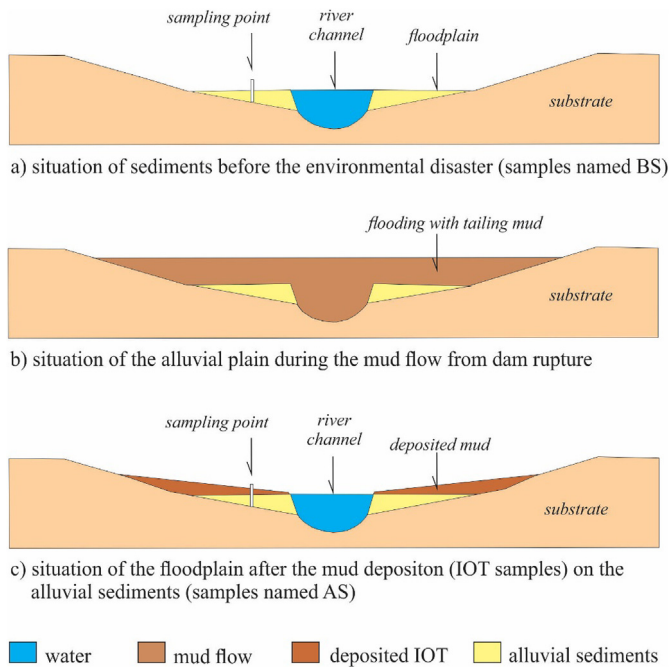


Fig. 2. Schematics of the Rio Doce alluvial plain showing alluvial sediment collection points (a) pre-environmental disaster (BS samples); (b) post-environmental disaster (AS samples); and (c) iron ore tailings (IOT samples).

3.2. Mineralogy

The fine sand fractions of the sediments before and after the collapse consist mainly of quartz, kaolinite, muscovite, and gibbsite (Fig. 4a, b). However, the relative intensity of kaolinite peaks increased in post-disaster sediments, indicating a kaolinite enrichment of the sedimentary material after the disaster.

Like the sediments, the iron ore tailings contain quartz, muscovite, kaolinite, and gibbsite, whereas hematite and goethite appear in the post-disaster samples (Fig. 4c). The relative intensity peaks associated with muscovite, kaolinite, and quartz also changed, similarly to post-disaster sediments.

3.3. Semitotal fraction of potentially toxic trace metals

The arsenic levels in the pre-disaster sediments were higher than Level 1 and lower than Level 2 established by CONAMA (Brasil, 2012) (Fig. 5a), indicating that the environment had already been impacted by the mining activities even before the Fundão Dam collapse. Likewise, arsenic levels in post-disaster sediments were above Level 1 and below Level 2, except for an outlier that exceeded 17 mg kg^{-1} , the limit concentration that indicates a high probability of occurring adverse effects to the biota. In the tailings, arsenic levels were also higher than Level 1, and even higher than the Level 2 threshold in certain locations, significantly exceeding the determined local background levels.

On the other hand, cadmium levels (Fig. 5b) in the riverbed

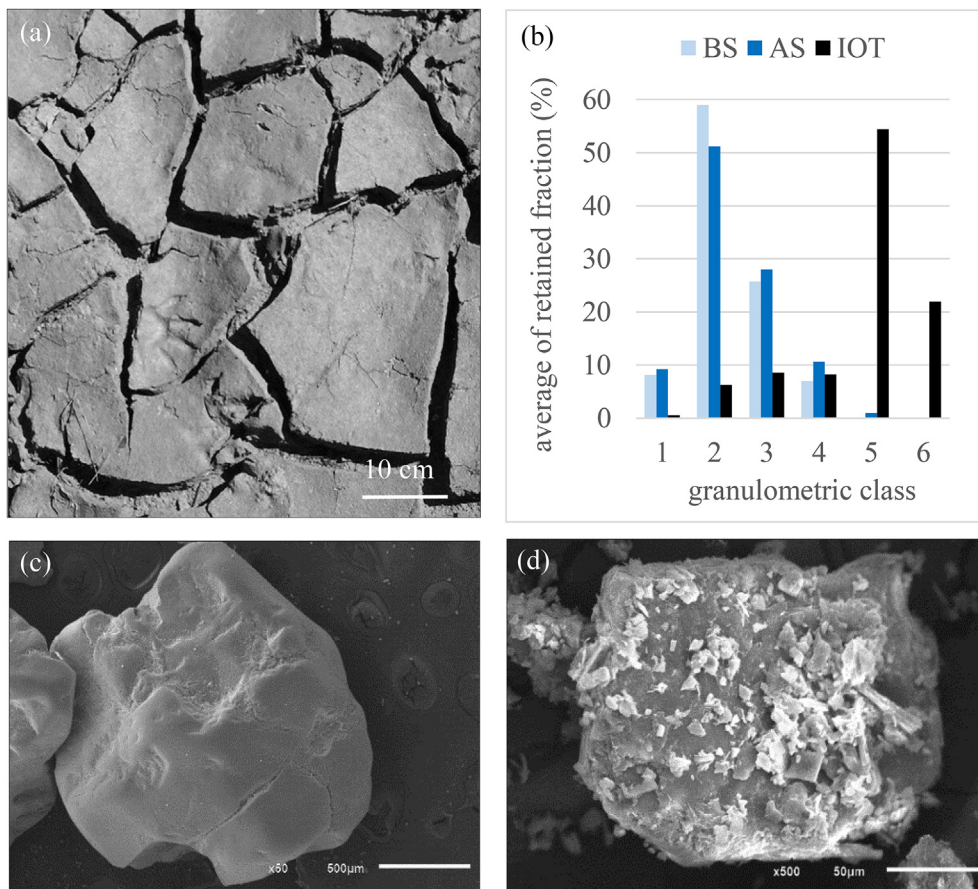


Fig. 3. (a) Iron ore tailing deposited on the sediments of the Rio Doce alluvial plain after the slowing down of the mudflow from the Fundão Dam collapse; (b) Grain classes of sediments before (BS) and after (AS) the Fundão Dam collapse and of iron ore tailings (IOT) deposited on the Rio Doce alluvial plain (1: granule, 2: coarse sand, 3: medium sand, 4: fine sand, 5: silt, 6: clay); SEM micrographs of the (c) alluvial sediment and (d) iron ore tailing.

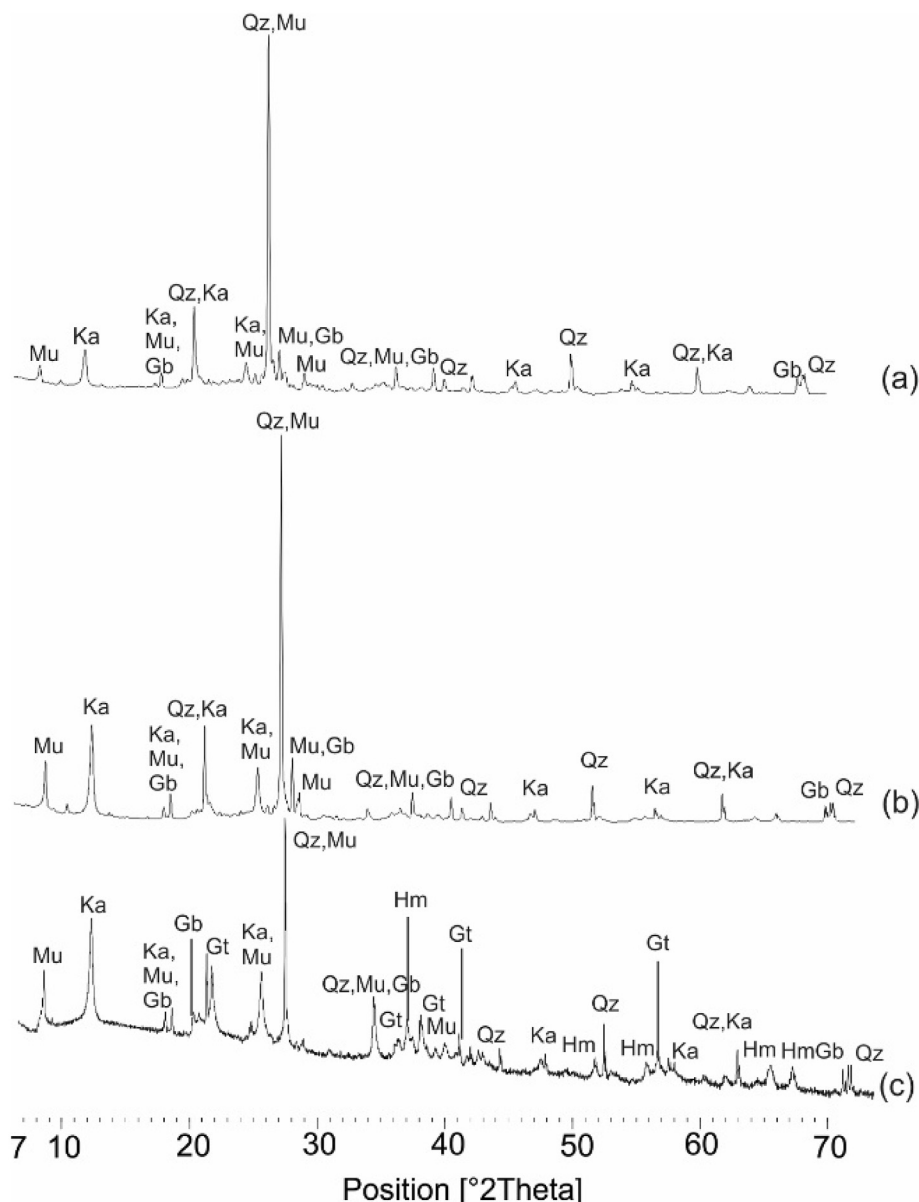


Fig. 4. Diffractograms of sediment samples (a) before the disaster, BS; and (b) after the disaster, AS and (c) iron ore tailings (IOT) deposited on the Rio Doce floodplain (Qz: quartz, Ka: kaolinite, Mu: muscovite, Gb: gibbsite, Hm: hematite and Gt: goethite).

sediments were considerably higher post-disaster compared to pre-disaster levels, exceeding the reference values of Level 1 and even those of Level 2. Cadmium levels in the iron ore tailings were also significant, that is, the determined levels could adversely affect the biota. This distribution pattern indicates that mining tailings contributed to cadmium contamination of sediments, post-environmental disaster.

Copper (Fig. 5c) levels in the sediments were relatively higher pre-disaster compared to post-disaster, sometimes exceeding the levels found in the tailings. Further, outliers were observed in the pre- and post-disaster sediments, with values that exceed the reference values of Level 1 stipulated by CONAMA (Brasil, 2012), however, never higher than the reference value of Level 2.

Lead levels (Fig. 5d) are quite similar in the pre- and post-disaster sediments and always below the limit concentrations of Levels 1 and 2. Nevertheless, the levels in the tailings exceeded locally the reference values of Level 1.

Pre and post environmental disaster levels of nickel (Fig. 5e) and zinc (Fig. 5f) were similar in sediments and deposited tailings and below the reference values of Levels 1 and 2, except for a few outliers.

The Principal Component Analysis (PCA) divided the samples into two (Fig. 6a) groups, PC1 and PC2. PC1 includes pre-environmental disaster sediments and a few post-disaster sediment samples. There is, however, a tendency for the post-disaster sediment samples to migrate to the mining tailings group, indicating the increasing chemical similarity between these samples after the changes caused by the incoming sludge. This migration was influenced by the increasing levels of arsenic and cadmium in post-disaster sediment samples. There is a positive correlation between the arsenic and cadmium levels (Fig. 6b), the metals that exceeded both reference values, Levels 1 and 2. PC1 also indicates positive correlations among zinc, lead and nickel levels, partly influencing the tailings group. Copper was not significantly

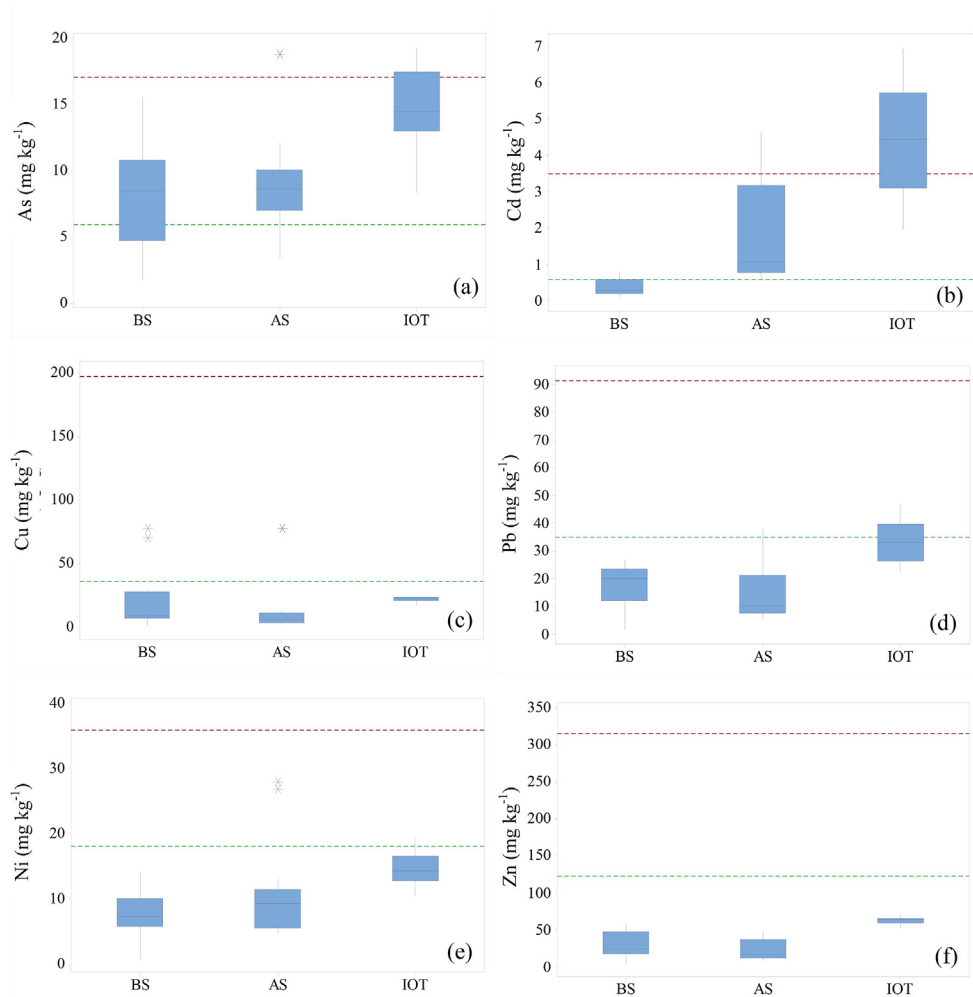


Fig. 5. Concentrations of trace metals in the alluvial sediments before (BS) and after (AS) the environmental disaster, and deposited iron ore tailings (IOT) in the Rio Doce. Green dotted line: Level 1 (threshold below which the likelihood of adverse effects to biota decreases, according to Brasil, 2012); red dotted line: Level 2 (threshold above which the probability of adverse effects to biota increases, according to Brasil, 2012). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

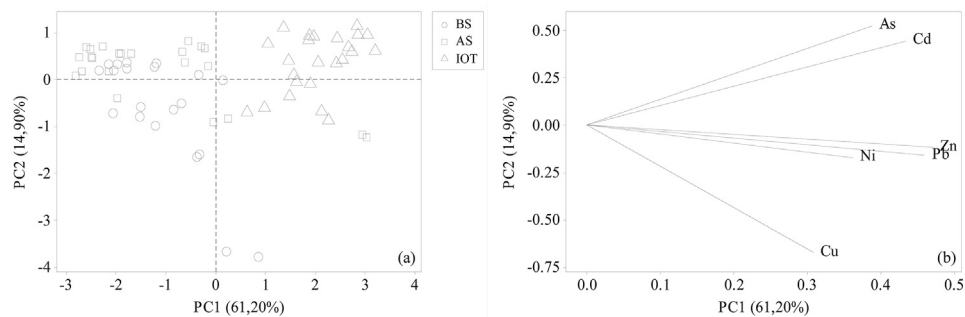


Fig. 6. Projections of (a) samples and (b) variables in the factorial planes PC1/PC2, concerning the trace metal contents according to EPA 3051A (Usepa, 1998), before (BS) and after (AS) disaster sediments, as well as iron ore tailings (IOT).

correlated with the other analyzed metals.

To calculate the geoaccumulation index (Igeo) (Fig. 7) and assess the environmental contamination degree, the pre-disaster sediment concentrations were adopted as background values. In the post-disaster sediments (Fig. 7a), the concentrations of cadmium reaches Class 3, indicating the emergence of moderate to severe

pollution in this metal, compared to background levels. By the other hand, the arsenic, copper and zinc remain in the Class 0, indicating no enrichment of these metals compared to the background values. Important to note that the alluvial sediments were already impacted by arsenic levels higher than the limits established by the CONAMA 454 standard (Brasil, 2012), as shown above.

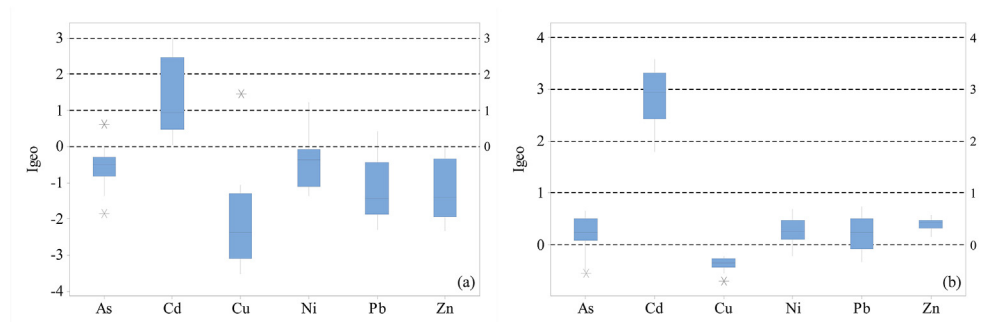


Fig. 7. Geoaccumulation Indexes (Igeo) referring to (a) post-environmental sediments (AS), and (b) iron ore tailings (IOT) deposited in Rio Doce after the Fundão Dam collapse (pre-disaster sediments are considered as the local background value).

In the iron tailings (Fig. 7b), arsenic and zinc levels changed from Class 0 to Class 1, where the highest concentrations of these metals were observed. Cadmium levels reach Classes 3 and 4, indicating a highly polluted condition compared to the background values while also exceeding the reference values of levels 1 and 2 established by CONAMA.

4. Discussion

The results of this study show that the mudflow containing iron ore tailings released after the Fundão Dam collapse impacted the alluvial sediments of the Rio Doce, by changing their physical and chemical parameters. The intense and sudden mudflow caused remobilization and redeposition of alluvial sediments, bringing and redistributing upstream sediments while introducing particulate matter from the mineral tailings that after depositing, formed residual ferrous crusts on the alluvial sediments.

Before tailings were deposited as a superficial layer, the sediment was reworked by the introduction of finer particles as evidenced by the changing grain size of the sediments after the disaster. The small percentage of clay fraction in the tailings demonstrates the grain selection during the material deposition after the mudflow, as observed in other affected areas (e.g. Davila et al., 2020). Most particulate matter smaller than 0.002 mm remained suspended in the mudflow until reaching the river mouth in the Atlantic Ocean. Some authors (e.g. Schaefer et al., 2016; Carmo et al., 2017; Queiroz et al., 2018) pointed out that the hydroelectric dams along the Rio Doce retained a considerable part of the iron tailing load, especially in the medium/coarse fraction, in areas upstream of the study region, whereas fractions smaller than fine sand were deposited downstream.

The mineralogy of iron ore tailings deposited in the Rio Doce sediments in Espírito Santo is mostly similar to the results reported for the sediments from different impacted locations analyzed by other authors (e.g. Silva et al., 2016; Schaefer et al., 2016; Almeida et al., 2018; Queiroz et al., 2018; Figueiredo et al., 2019).

Regarding the presence of potentially dangerous trace metals, the interpretations in the literature diverge as to the origin of contamination from either iron ore tailings or pre-existing sources. In the area closest to the Fundão Dam, Segura et al. (2016) detected Al, Fe, Mn, Ba, Sr, Pb, As and Cd in the mining tailings and pointed out, among them, Al, Fe, Mn, Ba, and Sr as of higher potential mobility, that is, they could migrate easily to sediments and seawater. Queiroz et al. (2018) consider that the ore tailings deposited in the estuarine portion of the Rio Doce constitute a “time bomb” with the potential for releasing heavy metals associated with ferruginous minerals. On the other hand, Davila et al. (2020) contested the statement, asserting that the toxic metals

were already present in the environment since most samples from unaffected areas had As, Ba, Co and Ni levels that exceeded the reference values of the environmental agency of the State of Minas Gerais. Nevertheless, most authors agree that the mudflow mobilized sedimented material and, therefore, changed the original environmental conditions.

Our study focused on the environmental standards determined by the Brazilian Environmental Law. The levels of trace metals determined in the sediments sampled “before” (BS) and “after” (AS) the accident and iron ore tailings (IOT), that became part of the sedimentary environment, were compared to the reference standards established in Resolution 454/2012 of the National Environment Council - CONAMA (Brasil, 2012). The reference values determine the threshold levels of trace metals that are released during sediment dredging and can, possibly, affect the biota. The resolution anticipates situations in which sediment matter is moved around, similarly to the passage of the mudflow released by the accident that changed the Rio Doce. Among the metals monitored by the regulations, the arsenic and cadmium levels determined in the sediments and iron tailings were outside the reference ranges. The levels exceeded Level 2 in some points, a threshold above which the probability of adverse effects to the biota increases.

The distribution pattern of arsenic levels indicates the presence of this metal source in sediments before the dam collapse, although it is also present in the mining iron tailings. Arsenic levels have been detected in soils and sediments not impacted by the mud from the accident by Guerra et al. (2017), Costa et al. (2018) and Davila et al. (2020), suggesting a previous environmental impact even before the Fundão Dam collapse. Cagnin et al. (2017) and Davila et al. (2020) associated this metal with the gold mining activities that exist upstream of the study area. However, it is noteworthy that the Geoaccumulation Index (Igeo) of arsenic ranks the iron tailings as a moderate pollution source compared to the environmental conditions before the accident.

Our study also indicate that cadmium levels in the ore tailings and post-disaster sediments were markedly above the reference values considered safe for biota. The geoaccumulation index (Igeo) ranks cadmium contamination levels in the tailings and post-disaster sediments, as reaching the range of an environment considered to be heavily polluted. This result contradicts the results reported by other authors for locations upstream (Silva et al., 2016; Figueiredo et al., 2019; Davila et al., 2020) and downstream (Gomes et al., 2017; Quadra et al., 2019) of the studied area.

The lead levels determined in alluvial sediments and iron tailings are similar and below the reference values of Levels 1 and 2 recommended by Resolution 454/2012 of the National Environment Council - CONAMA (Brasil, 2012). Likewise, the nickel and zinc

levels were similar in the sediments and tailings and comparable to those found by Silva et al. (2016) and Davila et al. (2020) upstream of the studied area. On the other hand, Queiroz et al. (2018) and Gabriel et al. (2020) reported relevant levels of these metals in estuarine sediments in regions close to the Rio Doce mouth. The Igeo determined for Ni, Pb and Zn were in the range between unpolluted and moderately polluted.

Furthermore, besides potential soil surface impermeabilization (Schaefer et al., 2016), the residual ferruginous layer deposited on sandy alluvial sediments can gradually release metals when exposed to weathering agents and the river dynamics itself (Hatje et al., 2017; Queiroz et al., 2018). Coimbra et al. (2019) monitored by remote sensing the suspended particulate matter along the Rio Doce between 2013 and 2016. These authors show the iron tailings entering the river system and arriving in the marine environment while clearly recording the resuspension of particulate matter related to the waste deposited along the river, up to one year after the Fundão Dam collapse.

The natural movement of sedimentary material occurs slowly, however, the release of mining effluents caused the sudden movement of sediments. Therefore, the natural and anthropic sedimentary materials, contaminated or not, that previously lay on the substrate and/or underground tending to attenuation or immobilization, were suddenly remobilized by the mudflow provoked by the dam collapse.

We consider that many of the diverging results reported in the literature result from differing methodologies, environmental matrices, and especially, different collection sites. Very careful considerations are required for comparing one of the largest mining areas in Brazil, the Quadrilátero Ferrífero, a region traditionally dedicated to mining, with an estuarine region and, even more, with the platform area adjacent to the Rio Doce delta, near Abrolhos, an environmentally protected region by law that is crucial for the maintenance and preservation of several marine species. Any environmental management guideline must consider as a priority the retention, in an extremely safe way, of the effluents generated by the intense anthropic activity existing in the mined area.

The tailings dumped by the mining industry in Rio Doce became part of the environment (Almeida et al., 2018), thus being able to gradually provide chemical input to the environment from the river rework. It must be highlighted, therefore, the several works studying the consequences of this impacting disaster (e.g. Segura et al., 2016; Guerra et al., 2017; Aires et al., 2018; Queiroz et al., 2018; Bernardino et al., 2019; Quadra et al., 2019; Gabriel et al., 2020), as well as the importance of environmental monitoring to assess its long-term effects on directly impacted soils, sediments, and the biota along the Rio Doce and the adjacent coastal environment.

5. Conclusions

The mudflow from the Fundão dam collapse remobilized the Rio Doce alluvial sediments and introduced fine particles in the fluvial deposit. The cadmium and arsenic content increased to levels above the National Environment Council thresholds for sedimentary materials to be dredged in waters under national jurisdiction. The increasing cadmium levels originated from the tailings, whereas arsenic originates from other pre-existing sources in the sedimentary basin. The iron ore tailings also affected the physical parameters of the sedimentary environment by forming a ferruginous crust that can waterproof the land surface and may gradually release toxic metals when exposed to the weather and river rework.

A long-term monitoring program should be developed along the Rio Doce to detect possible effects of the expected sporadic

resuspension of sediments, a natural result of climatic events and river dynamics.

Credit author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2020.127879>.

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