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RESEARCH ARTICLE

Dynamics of Heavy Metals in the Waters of Igarape do Quarenta: The Water Body that Crosses the Industrial Hub in the Brazilian Amazon

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Abstract:

This study aimed to characterize an important body of water that crosses the industrial pole of Manaus – Igarapé do Quarenta, in Amazonas, and to evaluate possible sources of heavy metals in these waters. It is known that the anthropogenic and industrial influence is very intense in these waters, which significantly increases the levels of heavy metals. The results showed that the waters of the Quarenta stream are close to neutrality (pH from 6.87 to 7.03), which contradicts the acid values characteristic of the region. The electrical conductivity values exceeded 300 µS cm 1, which places the electrolyte charge of these waters almost 20 times higher than the natural values. Among the heavy metals studied, the highest contents were for Fe and Mn, whose concentrations were higher than those established by legislation. Along the Quarenta stream, sinks of heavy metals can be seen, probably due to the phenomenon of dilution or precipitation of these materials. The PLI demonstrated that there is a large polluting load of Pb, Cd, Cu and Mn. The HCA study showed associations between Mn – Fe, Cu – Cd, Ni -Pb and Cr – Zn, which reflect biogeochemical processes being altered by anthropogenic factors. Finally, PCA informs that pH and electrical conductivity inversely influence the concentrations of Zn and Cd and that Ni is the most representative metal in the dataset.

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Introduction

The concern with the preservation of water resources is not from today and much is discussed about policies and technologies aimed at recovering water in various parts of the world. It is believed that at least 1/3 of the planet's population does not have access to treated water on the globe. It is estimated that many hospital internments are caused by the consumption of contaminated water[1].

Brazil is the country with the largest reserves of fresh water, and on the other hand, one that most wastes this valuable resource, without presenting policies to international organizations that will solve this situation. The country consumes millions of liters of water a day both in industry and agriculture, without, however, demonstrating technologies aimed at recovering many water bodies already compromised in the country, such as rivers, lakes and streams.

The Amazon is the largest holder of water in Brazil. It has seemingly inexhaustible reserves of water, both surface and underground. It is in the Amazon that the largest tropical forest on the planet is found. In addition to rivers that carry extraordinary volumes of water, the region has the largest water aquifer on the planet. This aquifer system is called by WHO as the Great Amazon Aquifer System, previously known as Alter do Chão. In this extremely humid biome, it also has flying rivers that are supplied under the action of evapotranspiration from trees, carrying water vapor to the rest of the country[2].

In fact, this system has undergone severe changes in recent decades. Many of these changes are consequences of the disorderly process of urbanization. This demographic/social expansion brought severe pressure on natural resources, above all, on water resources. Manaus is one of the cities with the highest GDP in the northern region and also where the largest industrial hub in the region is located. The city's water table is very close to the surface, which causes it to be cut by numerous streams. There are at least four water basins in Manaus, as follows: Tarumã-Açú, São Raimundo, Educandos and Purequaquara. The São Raimundo and Educandos basins are those which cross the urban center of Manaus. The first one is characterized by the discharge of domestic sewage, while Educandos basin, by both domestic sewage and industrial effluents[3,4,5].

Many works have already been carried out in the Educandos basin [6, 5, 7] and the vast majority of them are concentrated in its main draining body, the igarapé do Quarenta. This body of water crosses the city of Manaus in a southeast direction, carrying many effluents thrown into it by the industrial hub factories. The population of Manaus is estimated at 2,403,796 inhabitants, where at least 40% of it contribute to the release of effluents into these waters[7].

Manaus attracts many people to the northern region of Brazil, due to the opportunity of getting jobs in the industrial hub factories. There are approximately 600 companies operating in the industrial hub, in various sectors, such as electronics, electrical appliances, motorcycles, bicycles, chemical products and plastic injection. All processes of these companies generate effluents which are launched in the Educandos basin and taken to the Igarapé do Quarenta.

Literature research has found several studies which have evidenced the anthropogenic effects on the waters of the Igarapé do Quarenta, above all, regarding degradation by heavy metals [4,5,6,8]. Metals are not common in large concentrations in the environment. Generally speaking, high concentrations are the result of industrial activities. The most common metals present in the world industry are Fe, Cu, Cr, Ni, Zn, Mn, Cd, Pb. When in high concentrations in the environment they can pose serious risks to human health, which can lead to death, as many are responsible for the incidence of cancer [9]. Heavy metals have the potential to bioaccumulate in living beings and can sediment over long distances [10]. These elements are characteristic of industrial processes, since the atmospheric input of these metals is negligible.

Igarapé do Quarenta receives a large load of industrial effluents and, characterizing the distribution of heavy metal concentrations in this body of water, means understanding the sources that release these metals, as well as the deposition process along its bed. In addition, the spatial arrangement of heavy metals in the waters of the Igarapé do Quarenta can serve as a parameter to distinguish which is the most representative metal in this body of water and whether there is any leakage of these metals into the bottom sediments.

Thus, this study intends to characterize the waters of the Igarapé do Quarenta, taking in account the polluting load of heavy metals and thus identifying the possible contaminating sources, in addition to highlighting which metal is the most representative within this dataset.

Material and methods

Description of the study area

The city of Manaus is located on the Alter do Chão geological formation which is characterized by the scarcity of ions and for being a very weathered terrain. These conditions make the soil in this region chemically poor, and is reflected in the bodies of water that cross these terrains. Proof of this is that historically, the bodies of water that cross Manaus were extremely acidic in the past, with pH values not exceeding 4.5. The electrical conductivity values also did not exceed 20 µS/cm [11]. The Igarapé do Quarenta was a classic example, whose waters were recognized as important bathing spots in Manaus.

However, the waters of the igarapés that cross Manaus have not had these conditions for at least three decades. The example is the Igarapé do Quarenta, where this study was carried out, which presents pH values above 6.0 and electrical conductivity values above 200 µS/cm. Thus, natural values can only be observed today in the vicinity of Manaus and in forest reserves, such as the Adolpho Ducke Reserve, located in the northern part of Manaus[12].

The climate in Manaus is of the “Afi” type of the Köppen classification. According to this classification, the climate zone “A” corresponds to a tropical climate, with practically no winter and an average temperature for the coldest month always above 18°C. The climate type “f” indicates the occurrence of rain during the year, with precipitation above 60 mm in the driest month, which in Manaus corresponds to September. The climatic variety “i” (of isotherm) shows that there is no great difference between summer and winter, with annual variations in average temperature that do not reach 5°C.

Analytical procedures

Five water samplings were carried out between September and November 2019, along the Educandos basin, as shown in Figure 1. The samplings were carried out every two weeks, at five previously defined points: P1 (Detran East Zone of Manaus), P2 (Entry of the Conjunto dos Industriários), P3 (Exit of the Conjunto dos Industriários), P4 (Manaus 2000, Japiim) and P5 (Shopping Studio 5). It was expected to evaluate the sampling sites during a complete hydrological cycle. However, due to the pandemic caused by the novel coronavirus, the dynamics of water sampling and analysis was compromised.

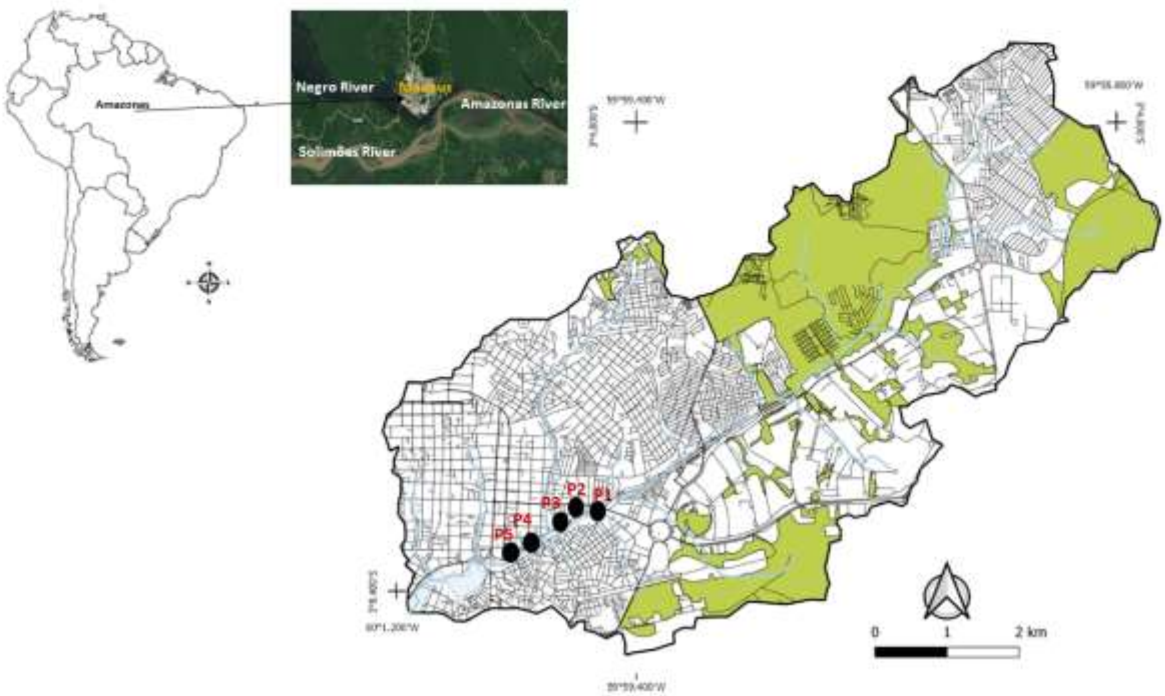

Figure 1: Location map of the points sampled in the Educandos hydrographic basin.

Thus, the collected samples were analyzed at INPA's Environmental Chemistry Laboratory. Samples were collected near the center of the water bodies and were immediately taken to the laboratory for preliminary analyses, such as pH and electrical conductivity - EC. The pH and electrical conductivity were determined by electrochemical methods. The pH was determined with an instrutherm pH meter - model pH - 2000 and electrical conductivity with a portable benchtop instrument from Mettler Toledo - seven Compact. Next, the samples were acidified with double distilled nitric acid for metal preservation up to pH 2.0 (2 drops of acid for every 100 mL of sample). Finally, the samples were sent to Samsung Eletrônica da Amazônia's Water Reuse Research Laboratory to determine heavy metals (Cr, Pb, Cd, Zn, Mn, Cu, Ni and Fe) by atomic absorption by flame analysis method (Shimadzu Spectrophotometer Model AA-7000). The samples were analyzed in triplicate and all results were tabulated in the R statistical package, version 4.01 - a robust free software that allows for multivariate statistical analyses.

Results and discussion

The concentration of heavy metals along the Igarapé do Quarenta was higher upstream. It was also noticed a phenomenon of decreased concentrations at the last point analyzed, probably due to the dilution or settling of these metals. The acidity of the waters was somewhat homogeneous, between P1 and P5, as shown in Figure 2. All the results presented below are based on the averages obtained from the five sampling points carried out between the months of September and November 2019.

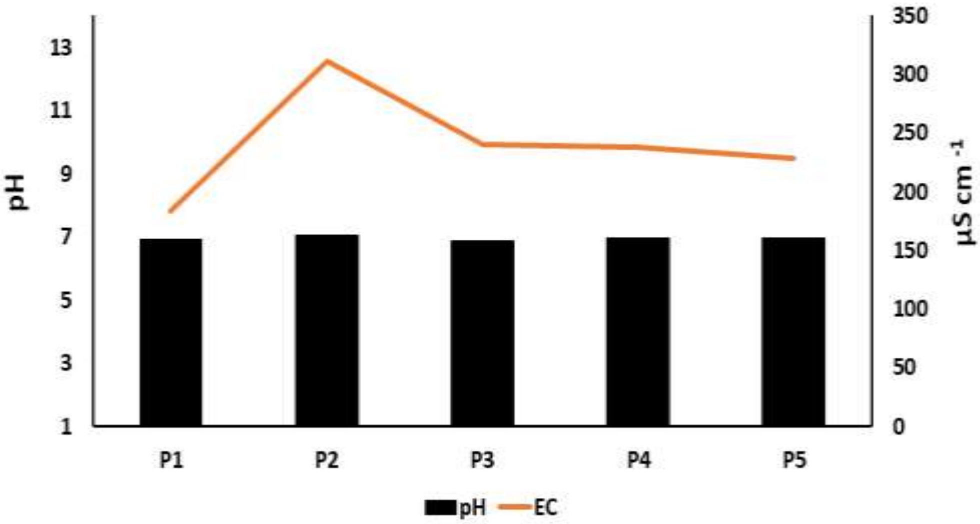

Figure 2: Variation of pH and Electrical Conductivity – EC, in the sampled locations of the Educandos basin.

The pH ranged from 6.87 at P3 to 7.03 at P2, which demonstrates the little variation in the acidity scale. However, it is known that the natural pH of the region is lower than 4.5, as shown by Bringel[6], Silva et al.[6], Torrezani et al.[13] and Calvo and Oliveira[14]. This shows how much these waters in the Quarenta basin are receiving alkalis resulting from urban activity, which certainly brings chemical and biological implications for this system.

Electrical conductivity values ranged from 227.0 to 311.0 µS cm^{-1}. The example of what is seen with pH, occurs similarly in the conductivity values, except for P2 which presents the highest values of electrical conductivity. Such observed electrical conductivity values are at least 10 or 20 times above the region's natural values [11,6,15].

Among heavy metals, the highest contents were observed for Mn and Fe. As this water basin extends across the industrial hub of Manaus, it was expected to find high rates of these materials, which was not confirmed. However, in comparison with CONAMA Resolution 357/2005 for class II waters, with the exception of the element Zn, these waters are contaminated by all heavy metals analyzed in this study. Another observation is due to the elements Mn and Fe, which presented concentrations above 1.0 mg L^{-1} (Figure 3), which suggests a low concentration of metals in these waters, which is an apparent contradiction. These concentrations, although low, are above what is recommended by current legislation and have serious implications for this body of water. These high levels of manganese and iron may be associated with the dark color and bad smell of the water[16].

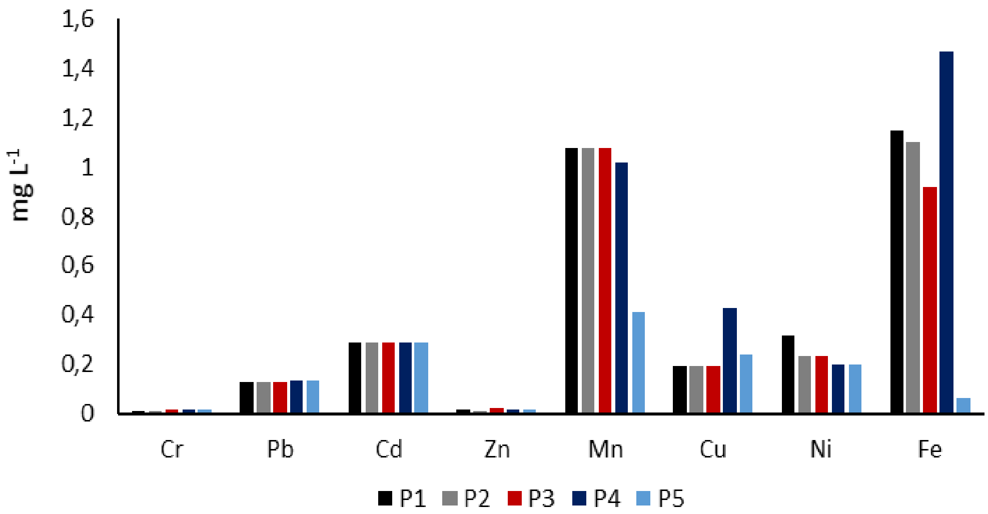

Figure 3: Contents of heavy metals analyzed as a function of sampling site.

It is important to highlight the dilution/sedimentation factor in this system, which seems to be efficient in "withdrawing" these metals from the water in P5. However, these metals must not simply disappear from the waters. What is likely to occur is the settling of these materials at the bottom of the Igarapé do Quarenta, as already demonstrated by Silva et al.[6] and Torrezani et al.[13]. This metal sink established in P5 is more evident when the more concentrated metals are analyzed distinctly, in this case Mn and Fe. Such phenomenon can be seen in Figure 4.

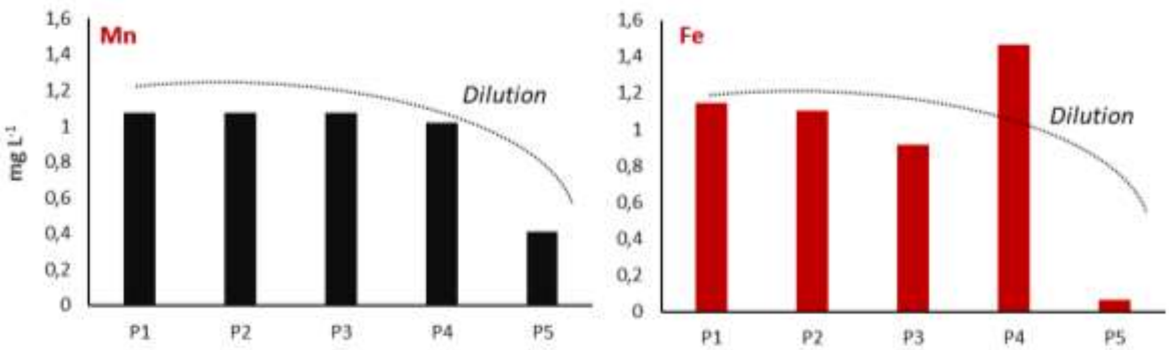

Figure 4: Phenomenon of the dilution of heavy metals at point P5, for manganese (A) and for iron (B).

In any case, although the effect of dilution can be perceived, the waters are contaminated by heavy metals and this polluting load is more dispersed in points adjacent to the Manaus Industrial Hub, which suggests the absence of public policies in the regulation and inspection of the release of effluents in these waters.

In fact, the acidity of the water does not clearly influence the distribution of the analyzed heavy metals. However, a trend is perceived in which all metals predominate in the alkaline pH of P2. Overall, metal values varied as a function of the most acidic point, P3, to the most alkaline, P2 (Figure 5).

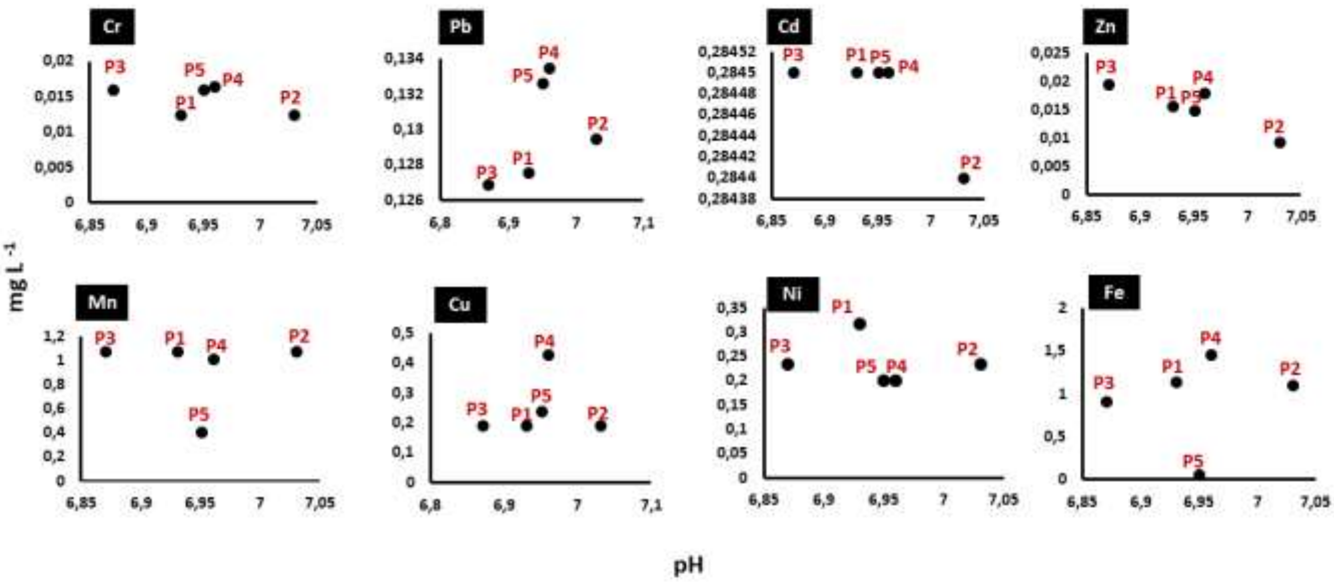

Figure 5: Influence of acidity on the mean concentration of the analyzed heavy metals.

It can be suggested that the decrease in acidity in these waters influenced the decrease in metal concentrations. This is very clear for the Cd and Zn elements. This condition can be explained through metal complexation phenomena, which can occur in environments with low acidity1,14.

What can be seen in the Manaus Industrial Hub is also seen in several countries around the world [17, 9,16,18,19] and one of the most important pollution markers is the PLI (Pollution Load Index). PLI is a water quality index aimed at the heavy metals polluting load. Next, there is a proposed model for the waters of the Educandos basin.

Pollution Load Index - PLI

PLI is an index of polluting loads aimed at heavy metals. Its mathematical definition is understood as a function of the ratio of metal concentrations to the natural background of that environment. The PLI equations can be described below:

$$PLI = \sqrt[n]{CF1xCF2xCF3x} \ldots \ldots \ldots CFn \qquad Eq\ (I)$$

Where CF is defined as:

$$CF = \frac{Cmetal}{Cbackground} \qquad Eq\ (II)$$

C_{metal} is the metal concentration observed in the medium, while $C_{background}$ represents the natural values for a region. Interpretation for CF values are

provided below. FC < 1: No contamination; 1 ≤ CF ≤ 3: Moderate Contamination; 3 ≤ CF ≤ 6: High contamination; CF > 6: Very High Contamination. As for the Polluting Load Index - PLI, when below 1: no contamination or unidentified contamination; when above 1, heavy metal contamination.
In this study, we sought to search for background data from the past two decades, in order to establish naturality markers for the CF and PLI results. What is noticeable is that, in fact, the waters of the Igarapé do Quarenta are very compromised by the presence of heavy metals (Table 1).

Table 1: CF* and PLI values for sampled locations.

	CF_{Cr}	CF_{Pb}	CF_{Cd}	CF_{Zn}	CF_{Mn}	CF_{Cu}	CF_{Ni}	CF_{Fe}	PLI
P1	0.21	42.53	568.92	0.19	35.86	9.52	1.58	0.15	4.09
P2	0.21	43.18	568.89	0.12	35.86	9.52	1.17	0.14	3.68
P3	0.27	42.32	568.95	0.24	35.86	9.52	1.17	0.12	4.06
P4	0.27	44.52	568.93	0.22	33.93	21.43	1.11	0.19	4.64
P5	0.27	44.21	568.98	0.19	13.69	11.90	1.11	0.00	1.89

*Background values of Guedes[20] and Viana[21]

According to Table 1, Cr, Zn and Fe do not represent contamination for the medium. There is moderate Ni contamination, while Pb, Cd, Mn and Cu represent a very high contamination for these waters. All determined PLI values place the five sites sampled in this study as contaminated.

These results for CFPb are much higher than those observed by Abdullah et al [17], but are lower than those of CFZn in Malaysia. Ahmad et al [18] observed in waters and sediments in Pakistan, CF values below 9, although with PLI above 1 in all sampled points. Buhari et al.[19] also observed PLI values lower than 2 in Peninsular Malaysia, while Khan et al [16] noted that the highest values of FC were obtained for Ni (30.22) and the lowest for Pb (2.42) in an environment where the maximum PLI reached the value of 17.47 in Bangladesh.

As can be seen, there are many works around the world that use PLI markers, and the waters of Igarapé do Quarenta are as much or more compromised than many waters on the Asian and African continents. The use of PLI in Brazil is still incipient, but it manages to measure, at the level of analyzed loads, how much that environment is impacted by the action of heavy metals.

Multivariate statistics

The results obtained for the concentrations of heavy metals in conjunction with the pH and electrical conductivity results were tabulated and standardized in the statistical package R, version 4.04. The purpose of this tabulation and statistical standardization was to classify the metals and discriminate the most representative metal within this data set.

In the classification, the Hierarchical Cluster Analysis - HCA technique was used, which aims to group the variables according to affinity, and, above all, by the established correlation. Next, in Figure 6, the groups classified by HCA of heavy metals analyzed in Igarapé do Quarenta can be seen:

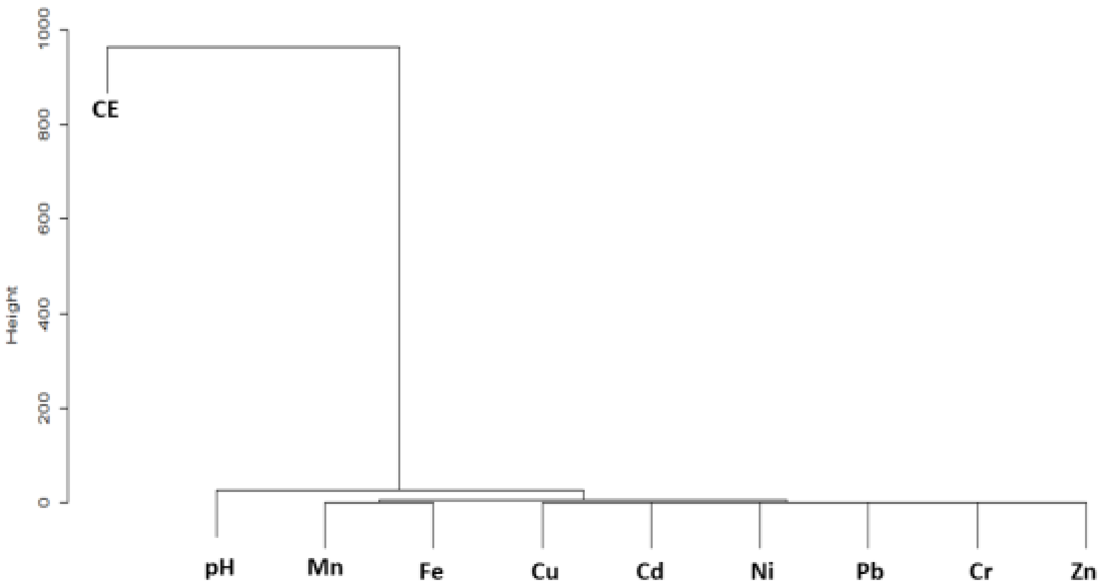

Figure 6: HCA for the variables analyzed in the Igarapé do Quarenta, using the Ward method and Euclidean distances.

The proposed HCA brings four groups of metals that are influenced by pH dynamics and electrical conductivity. The observed associations are Mn – Fe, Cu – Cd, Ni – Pb and Cr – Zn. Normally, these metals make associations depending on their valences and their ionic radii. Possibly, the physicochemical interactions in the waters of the Igarapé do Quarenta are likely to occur in pairs, influenced by the sources of contamination along this body of water.

With the premise of identifying the sources of these metals, the Principal Component Analysis – PCA, can serve to discriminate the most representative variable, and also trace common sources of that variable in the medium. According to Reis [22], the application of PCA must include: i) The variables included in the analysis; ii) the percentages of variance explained by each of the principal components; iii) The number of retained components and the proportion of total variance explained by them; iv) a table with the contribution of each variable for each component (factor loadings), before and after a factor rotation method is applied and; v) perform the interpretation of each main component retained.

Thus, a PCA was proposed for the results of this study, standardizing the variables and using the unit correlation circle, as shown in figure 7. PC1 carries 39.79% of information about the data set and PC2 31.22%. Table 2 brings the contributions of the first four PCs that correspond to 100% of the information about this dataset.

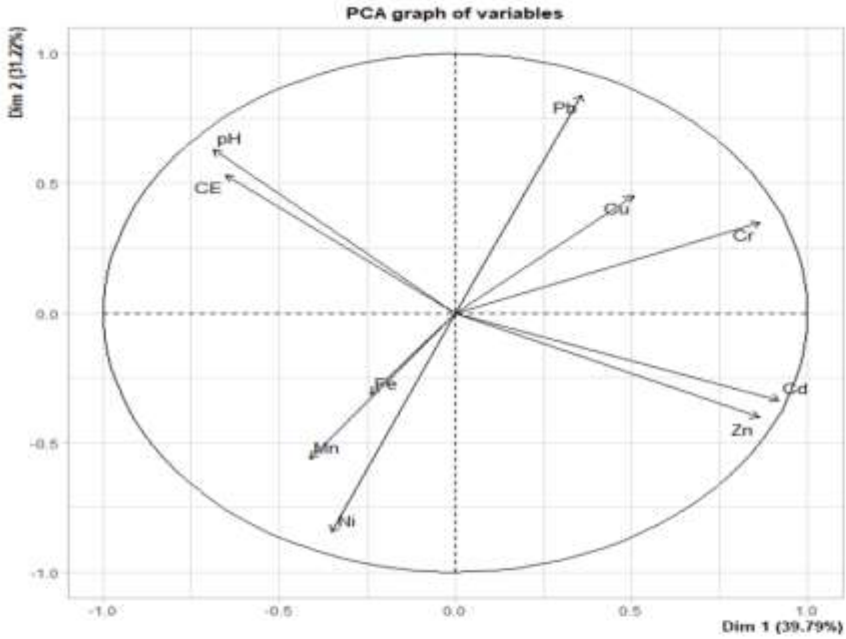

Figure 7: PCA of results obtained in the waters of the Igarapé do Quarenta.

The PCA discriminates that pH and electrical conductivity inversely influence the concentrations of Zn and Cd. It also highlights that Ni is the most representative element among the analyzed variables, with an important inverse contribution from Pb. This corroborates the results expressed by HCA, which brings these two elements together as pairs in this system. In fact, PCA demonstrates that when Ni concentrations increase, Pb concentrations decrease and vice versa.

Table 2: Retained components for the construction of the PCA

Eigen values	Dim 1	Dim 2	Dim 3	Dim 4
Variance	3,979	3.122	1,936	0.963
% of var	39,789	31.217	19,362	9,632
Cumulative of var	39,789	71,006	90.368	100

PCA also brings important correlations, such as Zn- Cd ,/ Pb -Cu-Cr/ and Fe-Mn-Ni. These interactions perceived in PCA suggest that these elements come from the same source of contamination, as shown in Figure 8 below. Therefore, some considerations about these metals can now be made.

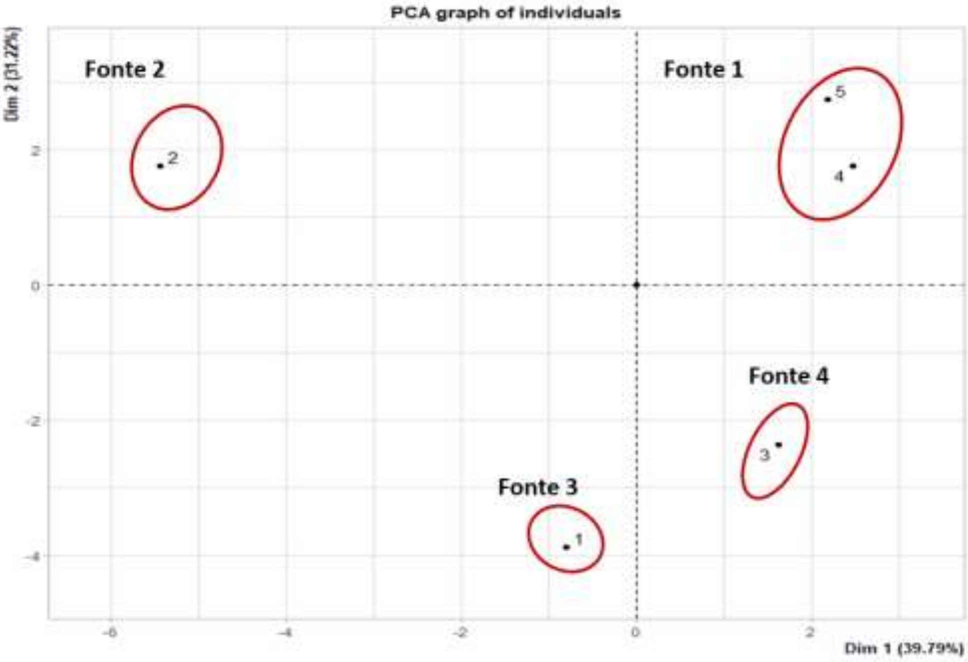

Figure 8: PCA with the sources of contamination by heavy metals in the waters of the Igarapé do Quarenta.

Figure 7 shows Pb, Cu and Cr with equivalent contributions in PCA and the most abundant source of these metals is at points P4 and P5 (Figure 8); Cd and Zn have their sources at P3 and Fe, Mn and Ni settle at P1. This point even stands out for the high dark coloration of the waters, which may be associated with these high fears of Fe, Mn and Ni.

Chemometrics still is not widely used in environmental analyzes in Brazil, especially in the Amazon. Papers dealing with chemometrics in the waters of the Quarenta are still scarce. Calvo and Oliveira [14], in a pioneering work, associated the contribution of Zn, Fe and Al in the waters of the Igarapé do Quarenta to companies in the industrial hub of Manaus.

As there are different industrial segments in the Manaus Industrial Hub, this can be explained by the different sources of heavy metals in these waters, in addition, of course, to the important contribution of domestic sewage in these waters.

Final considerations

The waters of the Igarapé do Quarenta are highly impacted, as a result of the city's polluting pressure and, above all, the discharge of effluents from the industrial complex. Some of the observations made can be listed here:

- The pH and electrical conductivity values are severely altered. The pH moves towards basicity, contrary to the acidic values of the region. The electrical conductivity is 10 to 20 times above the natural values, which certainly brings implications for the balance of this system;

- Concentrations of heavy metals do not exceed 1.0 mg L-1, except for Fe and Mn. However, although low, these concentrations are above what the current legislation recommends, and this is probably due to the release of effluents without technical care in these waters;
- Point P5, located on the Shopping Studio 5 bridge, represents a sink of heavy metals, either by dilution or sedimentation, given the sudden decrease in concentrations in that location;
- It was observed, in general, that metal concentrations tended to decrease with decreasing water acidity. This fact was more evident with Cd and Zn, which can be explained by the phenomenon of complexation in less acidic media;
- With contamination indices, all CF values indicated contamination, except for Ni and Cr. However, PLI values showed heavy metal contamination in all sampled locations;
- HCA showed paired associations, according to the radius and valence of the elements: Mn – Fe; Cu - Cd; Ni – Pb and Cr – Zn. Everything indicates that these elements are in balance in this environment, supplying each other's demand, as seen in the PCA;
- PCA showed that Ni is the most representative metal in this dataset. That Pb behaves inversely to Ni, its pair demonstrated by HCA. The PCA also discriminated the possible common sources of these elements: Cd and Zn have P3 as their main source; Pb, Cu and Cr come mainly from P5 and P4 and Fe, Mn and Ni come from P1, which largely explains the dark color of the water in this location.

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