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Diffuse sources of pollution and their influence on the nature of the sediments in Água Preta Lake

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ABSTRACT. Sediments in aquatic systems are indicators of contamination levels. Therefore, they play an important role in the assessment of the nature and intensity of human impact on aquatic environments. The objective of this work was to assess the levels of pollution by determining the concentrations of various metals and phosphorus forms in the sediments of Água Preta Lake, which supplies water to the Metropolitan Region of Belém, in north-eastern Brazil. Our data indicate non-uniform sedimentation with a large granulometric fraction of silt. The predominant form of phosphorus was inorganic phosphorus, to the detriment of the organic form. Observed mean concentrations of nickel, copper, lead, and zinc were lower than the PEL (Probable Effect Level). However, the highest observed concentrations of nickel and lead exceeded the prescribed TEL (Threshold Effect Level), which suggests pollution from an anthropogenic source.

Keywords: trace elements, phosphorus, contamination.

Fontes difusas de poluição e sua influência na natureza dos sedimentos do Lago Água Preta

RESUMO. Os sedimentos desempenham um importante papel na avaliação da intensidade e das formas de impacto sobre um meio aquático, porque funcionam como indicadores de níveis de contaminação. O objetivo deste trabalho foi avaliar as concentrações de metais e formas de fósforo em sedimentos do Lago Água Preta, que fornece água para a Região Metropolitana de Belém. Os dados obtidos indicam um reservatório com sedimentos não-uniformes, e uma maior fração granulométrica de silte. A forma predominante de fósforo foi o inorgânico, em detrimento da forma orgânica. As concentrações obtidas para o níquel, cobre, chumbo e zinco foram menores do que o PEL ("Probable Effect Level"). O níquel e chumbo representado níveis mais elevados do que os sugeridos pelo TEL ("Threshold Effect Level"), o que sugere uma influência antropogênica.

Palavras-chave: elementos traços, fósforo, contaminação.

Introduction

Urban growth in Brazil intensified throughout the 1960s and 70s (RIBEIRO; SCALON, 2001). The urban population of all the states that form 'the Legal Amazon' increased significantly during this period. The State of Para, for example, grew rapidly in the 1960s due to the implementation of target plans of the federal government, and the construction of the Belém-Brasília high way (PRATES; BACHA, 2011). The municipality of Belém has a population of 1,280,614, of which 1,272,354 live in the urban zones, and 8,260 in the rural zones (SÁ et al., 2005).

Urban growth and lack of basic sanitation result in the pollution of aquatic ecosystems. The major sources of pollution that affect the water quality of lakes are agriculture runoff, effluents from animal husbandry, untreated or semi-treated domestic effluents, and industrial wastewater, mainly from food, dairy, and dye industries (FYTIANOS; KOTZAKIOTI, 2005).

Among the several lakes affected by pollution are the Bolonha and Água Preta reservoirs, located in the Utinga State Park in north-eastern Brazil. The environmental problems that threaten this park are linked to the increasing urbanization of the nearby municipalities of Belém and Ananindeua. Located approximately 100 meters from the watersheds that supply Belém (FYTIANOS; KOTZAKIOTI, 2005), the Aura River is also a major conduit of pollution to the Park. Studies to determine possible contamination of Água Preta lake via the underground migration of leachate derived from waste deposits, using slingram electromagnetic measurements from the area located between the Belém Solid Waste Deposit (Aura river basin) and the lake, report that the detected conductivity changes are better explained by lithological changes than by leachate (BAHIA et al., 2004).

Although several research studies have addressed various aspects of the Bolonha and Água Preta reservoirs (BEZERRA; PEREIRA, 1995; SECTAM, 1997; MELO et al., 2006; COSTA et al., 2010), very few have looked into the sediments of these lakes in terms of the assessment of pollution levels. Sediments are important for the evaluation of contamination levels of aquatic ecosystems because of their capacity to accumulate trace elements (MULLER et al., 2010). The objective of the present work was to characterize the spatial distribution of granulometry, organic matter, phosphorus forms (total phosphorus, organic, and inorganic phosphorus), and trace elements (nickel, copper, lead, chromium, cadmium, and zinc) in sediments from the Água Preta reservoir. Although other studies have characterized the water quality of the Água Preta reservoir, few have focused on surface sediments. The importance of the Água Preta reservoir as a source of fresh raw water to the metropolitan region of Belém, makes it essential to understand the factors that threaten this aquatic ecosystem and its ecological services.

Material and methods

Surface sediment samples were collected from the Água Preta reservoir with a Van Veen-type point sampler at 10 points, georeferenced into coordinates, in November 2007. The samples were stored in polyethylene pots with lids, and kept frozen at the Laboratory for Control of Residues of UFPA (Figure 1).

Sediment samples were dried at a temperature of 50°C, after which 20 g of each sample was weighed out for further analysis. Next, the samples were separated into fractions < 2 μm, 2–62 μm e, > 62 μm, corresponding to clay, silt, and sand, respectively. The sand fraction was obtained by wet sieving using a 63-μm sieve, while the silt and clay fractions were separated by centrifugation at 1000 rpm for two minutes. Granulometric analysis was performed on these fractions, as described in Suguio (1973).

For the analysis of the percentage of organic matter (OM), 0.5 g of the fine (silt-clay) fraction was weighed out. It was then subject to oxidation by potassium dichromate in an acidic medium (concentrated $H_2SO_4 + K_2CrO_7$). Excess potassium dichromate (mixed oxidant), left over after the oxidation, was titrated with a standard solution of ammonium sulfate (LORING; RANTALA, 1992) to obtain the organic carbon content (OC %). Finally, percent organic matter was calculated as: OM% = OC% × 1.724.

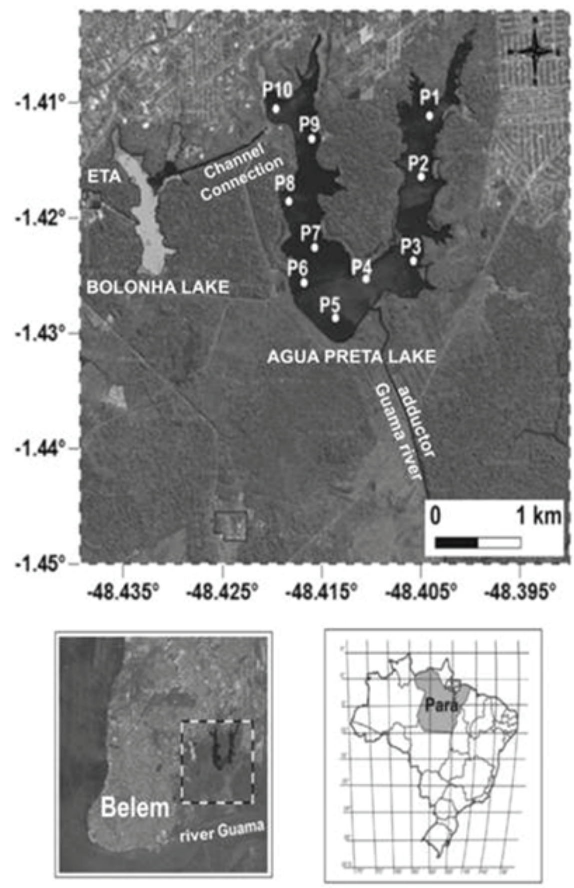

Figure 1. Location of sampling points in the Água Preta reservoir (Belém, Pará State).

For the analysis of the phosphorus forms in the sediments, we followed the method described in Protazio et al. (2004); it involved quantification of total phosphorus and organic phosphorus via the sequential extraction of phosphorus in sediments. After the extraction of ash-free sediment with 1 mol L^{-1} HCl, total phosphorus was determined spectrophotometrically as molybdate reactive phosphorus. Inorganic phosphorus was also estimated in the same way, except that the sample was not calcined. After the digestion, extraction was done and concentration was determined by the ascorbic acid method. After both total phosphorus and inorganic phosphorus were calculated, organic phosphorus concentration was determined by the difference between the two.

For the analysis of trace elements (Ni, Cu, Pb, Cr, Cd, and Zn) the samples were sieved through a stainless-steel 62-µm mesh in a medium moistened with deionized water. Next, the fine fractions (silt and clay) were dried in an oven at 50°C, and disaggregated in an agate mortar. These samples (approximately 10 g each) were sent to the ACME BRASIL commercial laboratory for the determination of heavy metal content, which involved acid digestion with 10 mL $HClO_4$-HNO_3-HCl-HF, and finally determining the content by inductively coupled plasma mass spectrometry (ICP-MS).

Results and discussions

Characterization of the sediments

There was a predominance of silt in the surface sediments of the Água Preta Lake. At all 10 sampling points, the silt fraction constituted the largest proportion of the sample (minimum of 67% at point 10; maximum of 96% at point 5, mean of 83±8%), and the sand fraction constituted the lowest proportion (minimum of 0.10% at point 5, maximum of 21% at point 10, mean of 5±6%). The clay fraction, considered important with regard to the adsorption of metals, had a mean value of 12±4% (minimum value was 4% at point 5, and maximum value was 17% at point 8). The particle size of metallic species in sediments affects their interactions with sediments. Owing to their surface area and adsorptive capacity, particles in the silt-clay fraction are important with regard to the retention of metallic species in the sediment (MARCHAND et al., 2011).

The low proportion of the sand fraction, together with cohesion in the fractions of silt and clay, characterize the Água Preta reservoir as a low energy environment, with no waves (BULHÕES; ESTRADA, 2011). Similar patterns (21 clay, 69 silt, and 10% sand) were reported from the floodplain sediments of Grande of Curuai Lake, in the State of Para, Brazil (AMORIM et al., 2009).

The lowest proportion of organic matter (OM) in a sample was 1% (at point 5), and the highest was 6% (at point 2), with an average of 3±1%. Sediments with less than 10% organic matter content are classified as mineral sediments (ESTEVES, 1988). By this definition, the Água Preta reservoir may be said to have mineral sediments. Highest values were found in the sediments of the Guama river and Guajara Bay, with values of 9% on the right bank and 8% on the left bank (SANTOS et al., 2012).

Evaluation of the phosphorus concentration

Regarding the phosphorus forms (total, organic and inorganic) in sediments of the Água Preta reservoir (Table 1), there was an increase in the concentration of total phosphorus (average of 172±37 $\mu g g^{-1}$).

Table 1. Concentrations of phosphorus forms (total, organic and inorganic) in sediment samples from Água Preta.

Phosphorus forms	Minimun ($\mu g\ g^{-1}$)	Maximum ($\mu g\ g^{-1}$)	Average ± standard deviation ($\mu g\ g^{-1}$)
Total phosphorus	90	220	172 ± 37
Organic phosphorus	8	174	71 ± 51
Inorganic phosphorus	20	213	101 ± 53

In anthropogenically disturbed environments, such as the Bacanga River/ MA,average concentration of total phosphorus (438 $\mu g g^{-1}$; PROTAZIO et al., 2004) was higher than those detected in the Água Preta reservoir (172 $\mu g g^{-1}$). The high value of total phosphorus in our study can be explained by phosphorus from sewage (industrial and domestic) that is released into the lake.

At six out of 10 points (points 2, 3, 4, 5, 8, and 9) the predominant form of phosphorus was inorganic, indicating that these points may experience higher rates of demineralization of organic matter. On the other hand, at points 1, 6, and 7, organic phosphorus was the dominant form, suggesting that these points experience lower rates of demineralization of organic matter, as well as phosphorus remobilization. Principal component analyses verified the positive correlation between organic matter and organic phosphorus.

Metals determination

Regarding the trace elements, the study was conducted for seven chemical elements (Ni, Co, Pb, Cr, Cd e Zn) that are important for the environmental quality of aquatic ecosystems. The total concentration of metals present in the sediment samples from Água Preta lake is shown in Table 2.

Table 2. Concentrations of metals found in sediment samples from Água Preta.

Metals	Min. Conc. ($\mu g\ g^{-1}$)	Max. Conc. ($\mu g\ g^{-1}$)	Average ($\mu g\ g^{-1}$)	TEL ($\mu g\ g^{-1}$)	PEL ($\mu g\ g^{-1}$)	Pre-Industrial Era ($\mu g\ g^{-1}$)
Ni	21	37	30±5	18	36	70
Cu	13	27	23±4	36	197	50
Pb	20	44	37±7	35	91	70
Cr	107	47	86±16	37	90	90
Cd	0,1	0,4	0,2±0,1	0,6	3	-
Zn	62	126	98±20	123	315	175

Unreferenced; TEL and PEL; Pre-Industrial Era for European lakes, USA and Canada (HAKANSON, 1980).

Mean concentrations of most metal species were below the reference values of the pre-industrial Era of European lakes. They were also within the acceptable limits laid down by the National Council for the Environment in Brazil; values of these metal concentrations that exceed this prescribed range may cause adverse effects on aquatic biota, and humans. The TEL indicates the level below which no adverse effects on the biological community are expected, and the PEL is the level above which adverse ecological effects are most likely (HAKANSON, 1980; SANTOS et al., 2013). The range between the TEL and PEL represents values that could have a variety of adverse ecological effects. Table 1 shows the benchmarks established by the National Environment Council for the metals Ni**,** Co, Pb, Cr, Cd e Zn**.**

In Água Preta lake there were vast differences between the average concentrations of metals found in the sediments of the sampling points. Point 5 had the lowest concentrations of metals, while point 9 had the highest, with values for two species of metals falling between the TEL and PEL indices, suggesting that the point might be associated with negative environmental impacts. However, metal concentrations at all other points, and for all other metals at point 9 found were found to be lower than values from pre-industrial era European lakes

The average values of copper and cadmium were lower than TEL and PEL standards, suggesting that these elements are not linked to pollution. This metal can be strongly associated with the crystalline structures of the sediment. Copper concentrations found in our study are similar to those found in the stretch of the Mogi River that passes through São Paulo. Amounts of both metals observed in the surface sediments of the Uberabinha river, in Uberlândia (SILVA et al., 2011), sediments of the Sergipe River Hydrographic Basin (SILVA et al., 2012), and sediments of the Billings Reservoir, in the state of São Paulo (HORTELLANI et al., 2013) were also below the PEL benchmark.

Mean concentrations of chrome and nickel present in sediments were above TEL, but below PEL, limits. However, maximum concentrations of both chrome and nickel were above the upper limits of TEL and PEL, indicating possible contamination by these metals, which were the only two to have concentrations that exceeded environmental safety standards (Figure 2).

The amounts of chrome found in the present study are higher than those reported from Billings reservoir (HORTELLANI et al., 2013) and the sediment basin of the Mogi Guaçu River (SILVA et al., 2005). Nickel concentrations reported from the sediments of Billings Reservoir (HORTELLANI et al., 2013), and the superficial sediments of the Rio Sergipe Hydrographic Basin (SILVA et al., 2012) were also above the PEL.

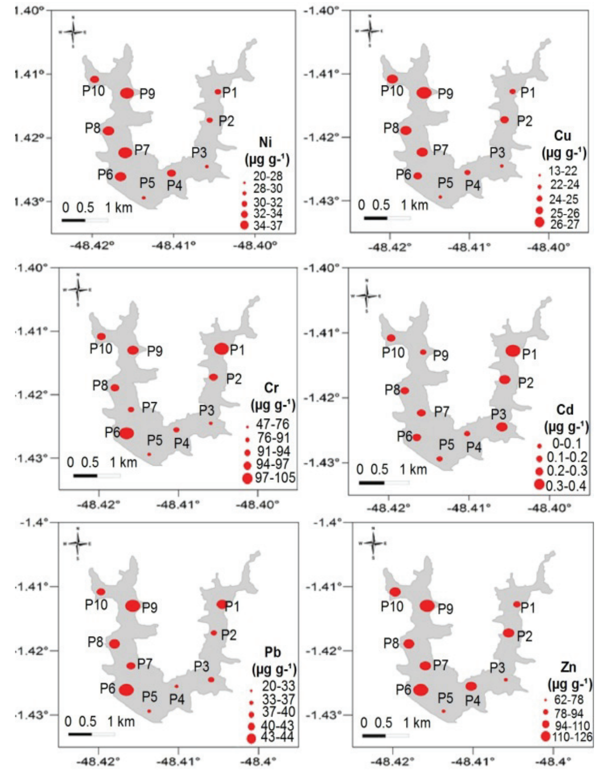

Figure 2. Surface distribution of Ni, Cu, Pb, Cr, Cd and Zn in the sediments of the Água Preta lake.

As in the case of copper and cadmium, zinc and lead also were at low concentrations at point 5. Although the highest zinc concentration in our sample exceeded the TEL limit, mean zinc concentrations were below both PEL and TEL limits, suggesting that this metal is not associated with anthropogenic influences. Furthermore, although values from Água Preta were below the reference indices, they were higher than those found in the sediments of Billings Reservoir, (HORTELLANI et al., 2013), the Uberabinha river (SILVA et al., 2011), and the Mogi Guaçu river basin (both in the São Paulo State portion as well as the stretch that flows through Minas Gerais State; SILVA et al., 2005).

Like nickel and copper concentrations, lead concentrations were also lowest at point 5 and highest at point 9. Mean lead concentrations from our study were below the PEL limit, but exceeded the TEL limit, indicating the influence of human activity on levels of lead in Água Preta. Lead concentrations reported for other fresh water bodies in Brazil varied; whereas in the sediments of the Guam River mouth, and both right bank and left banks of Guajara bay (SANTOS et al., 2012) lead concentrations were below PEL and TEL limits, in the sediments of Billings Reservoir (HORTELLANI et al., 2013), and the surface sediments of Uberabinha River (SILVA et al., 2011), they exceeded the TEL limit.

Across Água Preta Lake, sampling point 5 had the lowest concentrations of all the studied elements. This is likely because of its proximity to the Guama River, which brings in fresh water and dilutes the lake's water at this point. On the other hand, sampling point 9 had the highest concentrations of most of the studied elements, likely because of its proximity to sources of urban pollution. The close association between heavy metals and the residual fraction suggests that the concentrations of these metals are largely controlled by the movement of fine particles, which act as transporters for diffuse sources of pollution.

The principal component analysis explained 92% of the overall variance in the data, of which the first component (PC_1) explained 70%, and the second component (PC2), the remaining 22%. PC1 indicated a positive relationship between organic matter (0.46), clay (0.86), nickel (0.91), copper (0.89), lead (0.94), zinc (0.89), total phosphorous (0.85), and organic phosphorus (0.42; Figure 3). The positive correlation between the trace elements, total phosphorus, and clay, indicates the adsorption capacity of the granulometric clay fraction.

PC2 indicated a negative relationship between inorganic phosphorus (0.79), organic matter (-0.54), cadmium (-0.78), and organic phosphorus (-0.62; Figure 3). The negative correlation between organic and inorganic phosphorus implies a transformation between forms within the cycle of this element. Cadmium, which did not figure in PC1 but was part of PC2, appeared to be positively correlated with organic matter and organic phosphorus, suggesting that even at low concentrations, this trace element may play a role in the environmental chemistry of the reservoir.

Our findings also include a positive relationship between organic matter and nickel, copper, lead, and zinc. Organic matter, which has a proven adsorption capacity for heavy metals, may keep metals contained in the environment. An important physical component of organic matter is a fine-textured material, which is a product of the natural break-down and decomposition of plant, and animal wastes (VIDAL; BECKER, 2006). The material occurs as discrete particles or as a layer of particles, because anthropogenic material increases the affinity of the sediment for metals

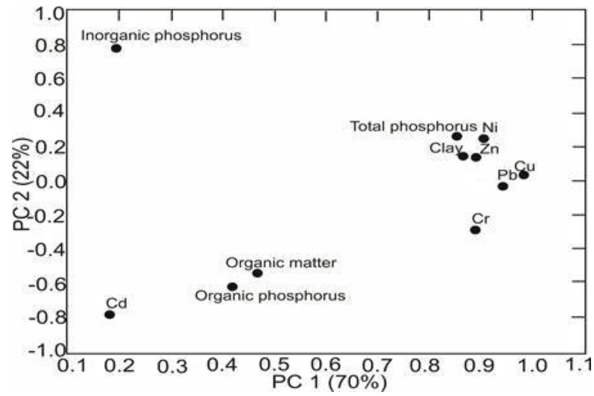

Figure 3. Weights of the variables of the principal distribution components of the phosphorus forms (TP = total phosphorus, OP = organic phosphorus and IP = inorganic phosphorus), the sediment size fractions (clay, sand), organic matter (OM), and trace elements (Ni, Cu, Pb, Cr, Cd and Zn).

Conclusion

The predominant form of phosphorus of the Água Preta was inorganic phosphorus. The obtained concentrations for nickel, copper, lead and zinc were lower than the PEL. The nickel and lead represented higher than those suggested by TEL, what suggest an anthropogenic influence. The investigation of the phosphorus forms and trace elements in sediments of the reservoir can be used as supplement in making decisions about the quality of the sediments of this aquatic environment in relation to its social importance.

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