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TRACE ELEMENTS IN OYSTER FARMING AREAS IN THE AMAZON

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ABSTRACT

Analyses of contaminants in sediments, such as trace elements, are fundamental to characterize normality or alterations in aquatic environments. These are important data to evaluate areas used for aquaculture, particularly those where there is a closer interaction between sediments and organisms cultured, as in oyster cultivation. The concentrations of trace elements (Al, Cd, Co, Cr, Cu, Fe, Ni, Mg) were assessed in the sediments of six oyster farming areas in the Amazon estuary. Sediment sampling was carried out in the months of November 2013 (dry season) and April 2014 (rainy season) and central areas were prioritized. Levels of trace elements were quantified by induced plasma optical emission spectrometry (ICP-MS). Possible effects on aquatic biota were evaluated by comparing the levels of metals in the sediments with the reference values proposed in the TEL and PEL protocols. The concentrations of trace elements in sediments in all evaluated areas were higher in the rainy season, except for copper. Following the order of concentration, in most of the locations Al> Fe> Mg> Cr> Ni> Co> Cu> Cd. Only Cd levels were higher than the TEL and PEL.

Key words: iron; aluminum; particle size; organic matter.

METAIS PESADOS EM ÁREAS DE CULTIVO DE OSTRAS NA AMAZÔNIA

RESUMO

Análises de contaminantes nos sedimentos, como os elementos traços são fundamentais para caracterizar normalidade ou alterações nos ambientes aquáticos. Estas são informações importantes para avaliar áreas usadas para aquicultura, em especial aquelas onde há uma interação mais próxima entre os sedimentos e organismos cultivados, como ocorrem na prática da ostreicultura. Foram avaliadas as concentrações de elementos traços (Al, Cd, Co, Cr, Cu, Fe, Ni, Mg) nos sedimentos de seis áreas de cultivos de ostras no estuário Amazônico. As amostragens de sedimentos foram realizadas nos meses de novembro de 2013 (seco) e abril de 2014 (chuvoso) e priorizaram pontos centrais nestas. Os níveis de elementos traços foram quantificados por espectrometria de emissão ótica com plasma induzido (ICP- MS). Possíveis efeitos sobre a biota aquática foram avaliados a partir da comparação dos níveis de metais nos sedimentos com os valores de referência proposto nos protocolos TEL e o PEL. As concentrações dos elementos traços nos sedimentos em todas as áreas avaliadas foram superiores no período chuvoso, com exceção do cobre. Seguindo a ordem de concentração, na maioria das localidades Al > Fe > Mg > Cr > Ni > Co > Cu > Cd. Apenas para Cd os níveis foram superiores em referência aos de TEL e PEL.

Palavras-chave: ferro; alumínio; granulometria; matéria orgânica.

INTRODUCTION

The State of Pará is formed by six mesoregions: Metropolitana, Marajó, Sudeste, Sudoeste, Baixo Amazonas and Nordeste. The Nordeste mesoregion has a geographical unit of 83,316 km^2, has 1.9 million inhabitants, and encompasses the main hydrographic basins of Araguaia-Tocantins and the Western Northeast Atlantic and covers 49 municipalities, comprising five microregions: Bragantina, Cametá, Guamá, Salgado and Tomé-Açu (PARÁ, 2015).

The Nordeste mesoregion of Pará concentrates the largest number of aquaculture ventures, withcontinental fish farming, marine shrimp farming and mariculture being the most developed aquaculture branches in Brazil. These activities are based primarily on non-native or exotic species, being held in various environments, modalities and production strategies (LEE and SARPEDONTI, 2008; MPA, 2013; BALLESTEROS *et al*., 2016; BRABO *et al*., 2016).

One of the main branches of mariculture is oyster culture, which is practiced exclusively in community projects managed by associations of producers, usually in suspended systems that adopt bags and racks installed on fixed tables as breeding structures (MPA, 2013; MIGNANI *et al*., 2013).

Oyster farming is considered less detrimental to the environment, especially when compared to shrimp and fish farming, for example, because it does not use artificial feed in the management of production, due to the filtering alimentary habit of these organisms. However, some authors point to the perceptible ecological impacts resulting from the implementation of bivalve mollusk farming. According to CALLIER *et al*. (2008), the main impacts related to this activity are: (1) depletion of phytoplankton and zooplankton biomass and decrease of seston in the water column due to the filtration practiced by bivalves and (2) increase of sedimentation rates related to biodeposition, which can cause organic enrichment and modifications in the geochemistry of the sediment.

The pollution arising from the sediments is related to the degradation of the quality of the waters that are significantly influenced from domestic and industrial effluent discharges, in addition to diffuse sources that reach the aquatic ecosystem (SALAMONI *et al*., 2009).

Trace elements in aquatic environments are found in abiotic and biotic resources. Geochemical characterization of the sediments show that this compartment acts as an environmental geoindicator due to its capacity to incorporate and accumulate contaminants (HORTELLANI *et al*., 2008; SANTOS *et al*., 2014; CAMPOLIM *et al*., 2017).

The identification and quantification of trace elements that are accumulated in sediments is of the utmost importance, because they are considered bioaccumulative due to their long residence time in aquatic environments. These can accumulate in humans through the food chain, causing health problems, such as the development of cancer associated with environmental exposure to cadmium (Cd), chromium (Cr), arsenic (As), selenium (Se) and lead (Pb) (BARROS and BARBIERI, 2012).

This information is important to evaluate in areas used for aquaculture, particularly where there is a closer interaction between sediments and grown organisms, as occurs in oyster farming. This also occurs in oyster farms located in the northeast of Pará in the communities of Augusto Corrêa (Nova Olinda); Curuça (Nazaré do Mocajuba); Maracanã (Nazaré do Seco); Salinópolis (Santo Antônio de Urindeua) and São Caetano de Odivelas (Pereru and Pereru de Fátima).

Thus, the concentration of trace elements (Al, Cd, Co, Cr, Cu, Fe, Ni, Mg) was assessed in bottom sediments of oyster farming areas oyster cultivation areas in the northeast of the state of Pará, Amazonas (Brazil), which can provide information about the environmental quality of these areas and provide technical knowledge of the environment where this cultivation occurs.

METHODS

The oyster farming oyster cultivation areas are located in the Amazonian estuary off the coast of the State of Pará, which along with the northwest coast of Maranhão was named by SOUZA FILHO (2005) as the Amazonian macrotidal mangrove coast, because mangrove areas exist on about 650 km of coastline in a straight line.

This area is characterized by a low relief (0 to 80 m), large coastal plain (with up to 70 km wide and extensive adjacent continental shelf (~200 km wide). Extremely irregular, indented areas, cut by several estuaries, are observed along the coast (SOUZA FILHO, 2005).

The climate is hot and humid, with a well-defined dry season (July to December) and rainy season (January to May), with average annual rainfall ranging from 2500 to 3000 mm per year and average temperature around 26 °C (MORAES *et al*., 2005).

The sampling of surface sediments was held in the central oyster farming area, during the low tide, in five municipalities (Figure 1).

The samplings were held manually and covered both seasons of the region, dry (November 2013) and rainy (April 2014). The sediment samples were stored in plastic bags and kept frozen until later analyses in laboratories.

The determinations of organic matter content were measured by the gravimetric method in triplicate and the granulometric composition was determined in a Shimadzu SALD-2201Laser Diffraction Particle Size Analyzer, at the Laboratory of Environmental Chemistry, Federal Rural University of Amazonia.

The samples were dried at room temperature and the fine fraction (<270 mesh) separated. A 200 mg portion was then weighed and nitric, hydrochloric, hydrofluoric and boric acids added. The sample solution was taken to the microwave. The analyzes were performed by ICP-MS (Bruker model 820 MS) installed in a clean room environment in the laboratory of the Evandro Chagas Institute in Pará.

Two standard reference material samples for trace elements in the soil (NIST 2710 and NIST 1646a, USA) were used to validate the results and the recovery results are listed in table 1.

The values of the trace elements determined in the sediments were compared to the sediment quality guideline values proposed by MACDONALD *et al*. (2000), which determine the Threshold Effect Level (TEL) and the Probable Effect Level (PEL) level.

A correlation matrix using Pearson's coefficient was drawn to examine the degree of affinity between the trace elements. This parameter measures the degree of association between the values.

RESULTS AND DISCUSSION

There was a predominance of sand in the sediments fromall sites during the less rainy month (Figure 2). It should be noted that in the rainy month there was an increase of silt and clay, with emphasis on the Community of Santo Antônio de Urindeua,

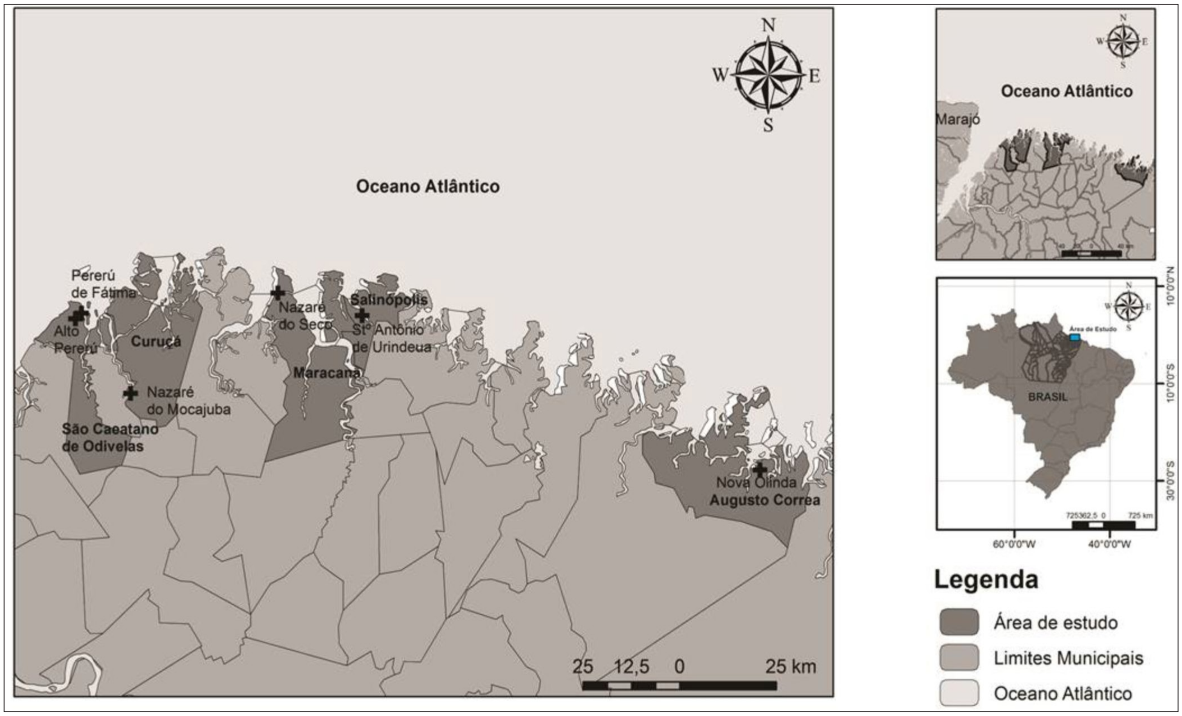

Figure 1. Location map of study areas. Source: The author.

Table 1. Recovery of trace elements.

Trace element	Reference sample NIST 2710 (%)	Reference sample NIST 1646a (%)
Al	83.4	88.9
Fe	94.5	100.3
Cr	94.0	91.2
Ni	78.6	76.4
Co	99.5	93.5
Cu	68.8	104.2
Cd	92.1	118.1

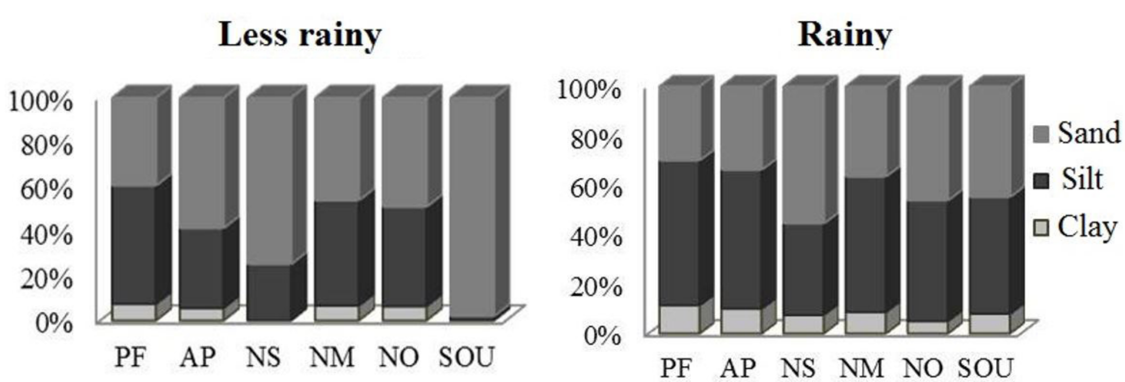

Figure 2. Granulometry of the less rainy and rainy periods. PF-Pererú de Fátima; AP-Alto pererú; NS-Nazaré do Seco; NM-Nazaré do Mocajuba; NO-Nova Olinda; SOU-Santo Antônio de Urindeua.

because the current was strong and turbulent in the farming environment during this period in this area.

Mangrove environments are usually composed of sediments of silt and clay fraction, due to low hydraulic power predominating in these places, however, according to FERREIRA *et al*. (2007), the velocity of the water flow is variable in mangroves, which can cause variations in the size and distribution of the grains. Moreover, the predominance of fine sediments also reflects the local hydrodynamics. According to BURONE *et al*. (2003), the deposition of fine sediments is not possible in areas with strong currents. This can be associated with the observed distribution in the Community of Santo Antônio de Urindeua, in the least rainy period.

The content of organic matter fluctuated between periods in all study sites (Table 2). The highest values were observed in the less rainy period, with exception to the sediments of the farms in Nazaré do Seco and de Santo Antônio de Urindeua which displayed the highest values in the rainy period, in accordance with granulometry observations with the increase of silt and clay fraction in this period.

The variation that occurred in the organic matter may have been a consequence of rain and the deposition of organic material. According to CRUZ *et al*. (2015) there is a large tendency to accumulate organic matter in oyster farming environments.

The levels of organic matter found may be a consequence of the collections of sediment carried out below the cultivation, where many fragments of oyster shells were observed. The same occurred in both low tides, locating near the edge of the mangrove, favoring the accumulation of organic matter of vegetable origin. According to BERREDO *et al*. (2008), the mangrove sediments are composed of significant mineral and organic fractions, the organic phase results from the accumulation and decomposition of vegetation, besides algae and animal remains, crustacean shells and small mollusks.

The mean concentrations of trace elements in the sediments oscillated at all sites (Table 3), with aluminum (Al) having the highest values, followed by iron (Fe) and magnesium (Mg).

The concentrations of Ca, Fe and Mg (Table 4 and Figure 3) were high for the region of northeastern Pará, when compared to other regions in Brazil (Table 4). These values can be related to the laterization, ferrification and deferrification processes that affected or affect the region, as these elements are found in clay-minerals that are aluminum, iron and magnesium silicates (COELHO *et al*., 2007). Kaolinite is one of the major clay-minerals species, the second largest of which is aluminum. It can be formed by the leaching and oxidation of rocks rich in Mg^{2+}, Ca^{2+} and Fe^{2+} (OLIVEIRA and BARBOSA, 2006).

For chromium, nickel and cobalt, the concentrations were higher in the rainy season (Figure 3). According to AZEVEDO and CHASIN (2003) large amounts of organic matter present in the ecosystem can reduce Cr^{+4} to Cr^{+3}, thusCr^{+3} may form low solubility polynuclear complexes and become part of the

Table 2. Organic matter (OM) content in surface sediments of oyster farms in the northeastern Pará.

Communities	Less rainy	Rainy
Pererú de Fátima	15.6	13.9
Alto Pererú	17.1	14.6
Nazaré do Seco	13.3	14.1
Nazaré do Mocajuba	17.4	8.2
Nova Olinda	16.5	14.0
Santo Antônio de Urindeua	14.2	16.0

Table 3. Mean concentrations (mg kg^{-1}) of trace elements (aluminum – Al, cadmium- Cd, cobalt - Co, chromium - Cr, copper - Cu, iron - Fe, magnesium - Mg, nickel - Ni) found in surface sediments of the communities in the study of northeastern Pará.

Metal	Nazaré do Mocajuba	Nova Olinda	Santo Antônio de Urindeua	Nazaré Seco	Alto Pererú	Pererú Fátima	*TEL	**PEL
Al	47469.4 ± 38081.0	44320.5 ± 247.1	71174.9 ± 3733.0	51984.9 ± 26370.8	70620.7 ± 1420.8	69810.3 ± 41251.9	_	_
Cd	5.4 ± 0.4	5.9 ± 0.3	5.9 ± 0.0	5.6 ± 0.0	5.6 ± 0.2	5.7 ± 1.2	0.7	4.21
Co	13.9 ± 0.2	13.2 ± 0.9	13.2 ± 0.2	12.5 ± 0.4	13.4 ± 0.3	13.9 ± 0.4	_	_
Cr	98.9 ± 7.1	97.5 ± 9.9	96.2 ± 4.9	90.8 ± 5.0	83.2 ± 1.1	82.7 ± 2.2	52.3	160
Cu	6.9 ± 1.8	4.4 ± 1.5	4.7 ± 1.5	5.2 ±0.6	9.4 ± 0.5	8.5 ± 2.1	18.7	108
Fe	29881.5 ± 1649.1	33233.9 ± 2022.5	30992.9 ± 240.3	29960.5 ± 509.4	30282.7 ± 891.5	29226.9 ± 2751.7	_	_
Mg	2924.7 ± 2151.5	3193.4 ± 80.6	4523.1 ± 107.4	3366.2 ± 1775.3	4530.4 ± 113.1	3812.1 ± 1074.2	_	_
Ni	24.1 ± 4.5	22.4 ± 1.9	21.5 ± 0.4	20.5 ± 1.5	22.2 ± 0.2	23.2 ± 2.7	15.9	42.8

*TEL (*Threshold Effect Level*), **PEL (*Probable Effect Level*). (-) no value for this element.

Table 4. Concentrations of trace elements (mg kg^{-1}) in sediments of estuaries of Brazil.

Places	Al	Cd	Co	Cr	Cu	Fe	Mg	Ni	Reference
Marapanim (PA)	-	-	8	82	10.7	-	-	22	Berrêdo *et al*. (2008)
Camamu (BA)		<1.0	-	29.2	20.2		-	-	Oliveira *et al.* (2009)
São Vicente (SP)	-	-	-	236.83	39.52	16571	-	32.45	Carmo *et al.* (2011)
Rio Saupe (BA)	3251.67	0.18	-	14.86	13.93	10125.0	2363.33	-	Reitermajer *et al*.(2011)
Porto de Suape (PE)		-	-	25	11		-	-	Marques *et al*. (2011)
Rio Passa Vaca (BA)	4725.0	-	-	-	45.6	13700.0	-	-	Andrade *et al*.(2013)
Rio Pará (PA)	-	<0.10	-	30.2	4.2	-	-	20	Vilhena *et al.* (2014)
Rio Mocajuba (PA)	-	<010	-	50	7.6	-	-	22.4	Vilhena *et al*. (2014)

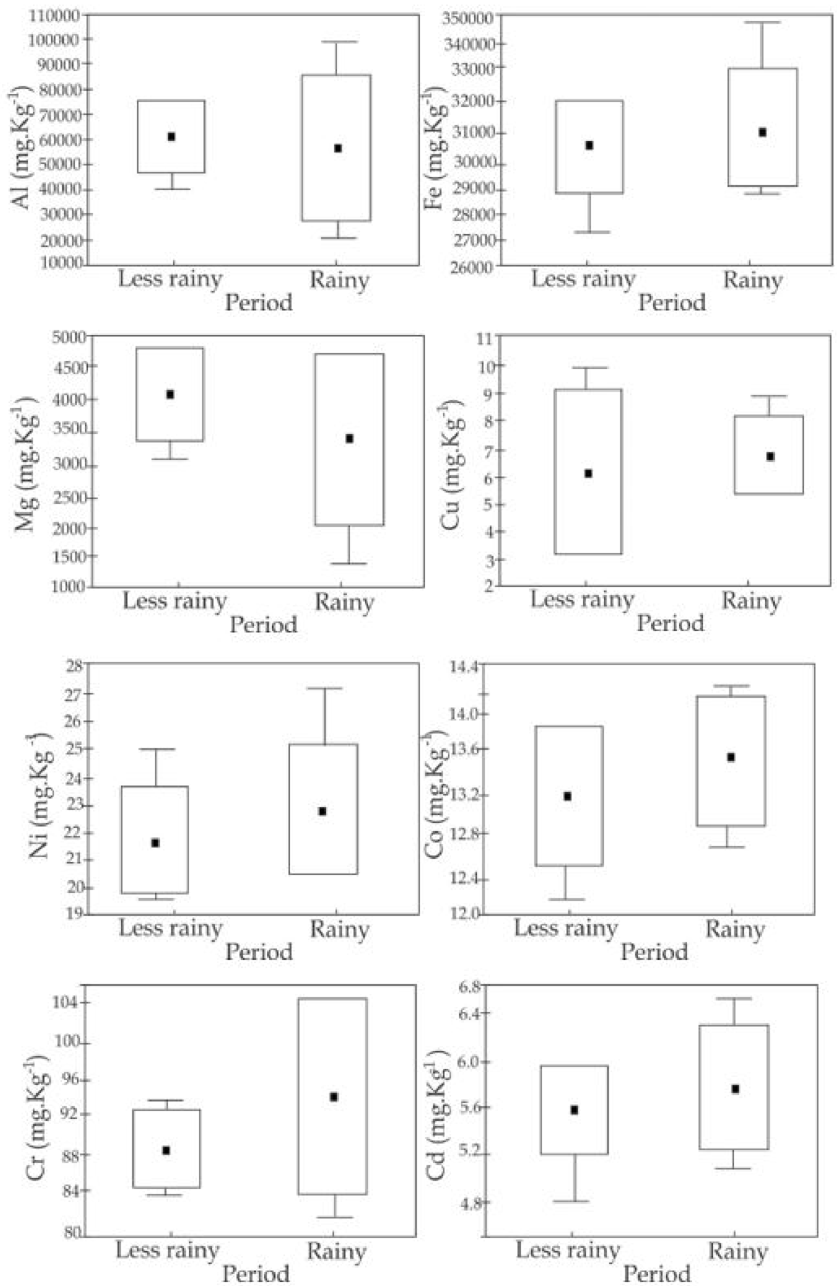

Figure 3. Box plot of the concentrations of metals, during the less rainy and rainy season of the region.

sediments. This can be expected for the studied areas, because they showed high values of organic matter, which results in low mobility of chromium for the environment and also low toxicity to aquatic biota.

Studies conducted in sediments in other areas in Brazil with chromium, nickel and cobalt showed values close to the present study (Table 4). However, the levels of chromium and nickel in sediment in all farming communities were above the reference TEL and below the PEL (Table 4), which can favor possible effects on the oyster culture.

For Nickel, the levels obtained in the present study may be of natural origin, since Ni is one of the five most abundant elements of the earth (AZEVEDO and CHASIN, 2003). As the levels were higher in the rainy season (Figure 3), it can be inferred that the leaching process favored the presence of this element in the sediments in these localities.

The concentrations of cadmium in sediments in oyster farming areasoyster cultivation areas are above the TEL and PEL reference limits (Table 4), i.e., the level of probable adverse effects to the biological community.

The mean levels of cadmium between the less rainy and rainy seasons were close (Figure 3), and above those described in the literature for other locations in Brazil (Table 4).

Copper was the only metal with a higher concentration in the least rainy season in comparison with the rainy season (Figure 3). However, it was also the only one in which concentrations were below the TEL and PEL references values (Table 4), showing that the environments under study are not contaminated by copper, nor an adverse effect is expected for oyster cultivation.

The levels obtained in this study were close to the levels observed (Table 4) in sediments from the northeast of Pará, indicating that the values of cobalt and copper obtained in the sediments of the oyster farming areas may be related with the characteristics of the localities.

Pearson correlation analysis was performed, showing the degree of affinity between the variables with a significance of 95% (0.05). The correlation matrix displays the pairs that obtained better correlations (r> 0.50) and statistical significance (p <0.05).

Al, Fe and Co in the sediment has been used as normalizing factors, due to significant affinities with other metals (ALOUPI and ANGELIDIS, 2001). The analysis revealed that the majority of positive correlations (Table 4) occurred with these metals (Al-Mg, Al-Cu), (Fe-Cd, Fe-Cr), (Co-Cu, Co-Ni, Co-Sil/Arg). According to ALOUPI and ANGELIDIS (2001), the necessary condition for these elements to be used as normalizers is probably because they are part of the natural constitution of sediments or result from natural climatic processes. It is possible to infer that in the present study the elements presented the highest concentrations during the rainy season.

There were positive and significant correlations between Silt/Arg-Cu and Silt/Arg-Ni (Table 5), which supports the idea that these elements are complexed to silt and clay of the studied environments.

The association of trace elements, like Al and Fe in estuarine sediments, such as what occurred in sediments of the oyster farm (Mg-Al, Fe-Cd, Cd-Al), indicates the presence of silicate minerals of terrestrial or allochthonous origin, such as biotite. This association is a strong indication of natural contributions of metals to the estuary. Positive correlations between Ni and Co can be explained, since these elements are mainly bound to clay minerals and sulfite minerals. According to BARBOSA *et al*. (2015), the sediments of the mangroves are silty clay and organic, with low permeability, favorable, therefore, for mineral dissolutions and mineralogical precipitations.

CONCLUSIONS

The trace elements evaluated were below reference values (TEL and PEL), with the exception of cadmium, which may cause some toxicity in oysters. However, it is necessary to investigate the bioavailability of this element for the biota in these oyster farming communities, as the toxicity in these organisms is related the specific physiology, associated with each metal.

The results suggest that the distribution of particles by size is one of the factors that affect the ability of the sediment to capture

Table 5. Correlation (Pearson) Matrix among the studied metals and organic matter and silt/clay of northeastern Pará.

Metals	Al	Cd	Co	Cr	Cu	Fe	Mg	Ni	Sil/Arg	Mo
Al	1.000									
Cd	0.113	1.000								
Co	0.204	-0.320	1.000							
Cr	-0.664	0.237	-0.144	1.000						
Cu	0.517	-0.628	0.596	-0.778	1.000					
Fe	-0.444	0.710	-0.273	0.542	-0.636	1.000				
Mg	0.915	0.299	-0.038	-0.507	0.319	-0.136	1.000			
Ni	-0.143	-0.414	0.937	0.097	0.441	-0.151	-0.353	1.000		
Sil/Arg	0.291	-0.393	0.982	-0.313	0.732	-0.359	0.042	0.891	1.000	
Mo	-0.823	-0.328	-0.201	0.140	-0.088	0.247	-0.774	0.083	-0.182	1.000

metals, since the oyster farming areas concentrate a higher amount of organic matter.

There is a need for continuous monitoring in oyster farming regions in the northeast of Pará, as these are used as a source of income and for consumption by the local population, so that guidelines can be provided for future programs to control environmental pollution.

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