



## Research Paper

# Determination of metals in estuarine fishes in a metropolitan region of the coastal zone of the Brazilian Amazon

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## ABSTRACT

The aim of the present study was to determine concentrations of cadmium, copper, chromium, manganese and iron in fishes in the São Marcos (SMB), São Jose (SJB) and Arraial (AB) Bays. Metal concentrations were determined using inductively coupled plasma optical emission spectrometry. Mean Cd and Cr levels were above the permissible limits set by different international or national guidelines in all three bays, whereas copper levels were well below the maximum acceptable limit. High concentrations of iron were found in all species analyzed, whereas high concentrations of manganese were found, especially in specimens caught in SJB. Spatial analysis indicated significant differences among the elements investigated. Copper was correlated more with SMB and AB as well as the species *Macrodon ancylodon* (carnivore) and *Sciades herzbergii* (omnivore). Cadmium and iron were strongly associated with AB and SJB, while manganese was only associated with SJB, mainly in carnivorous and herbivorous species.

## 1. Introduction

Human activities have led to an intense increase in the occurrence of metals in aquatic ecosystems (Asante et al., 2010; Nriagu and Pacyna, 1988; Squadroni et al., 2016b), especially estuarine-coastal systems (Delgado et al., 2010; Spencer et al., 2003), where elements transported from land-based sources, river output and runoff are often deposited (Gu et al., 2017). Researchers have investigated the effects of these elements and their accumulation in the aquatic biota (Alves et al., 2014; Bonai et al., 2009; Fu et al., 2014; Haimovici et al., 2014; Squadroni et al., 2016a). Metals in natural waters are toxic, persistent, non-biodegradable and can bioaccumulate in organisms, leading to the phenomenon of biomagnification (Adel et al., 2016; Hossain et al., 2018; Rahman et al., 2013; Seixas et al., 2014; Zafarzadeh et al., 2018). Metals

can enter the metabolic pathways of diverse members of the aquatic biota, leading to increasing levels due to bioaccumulation through ecological relationships, especially those related to feeding (e.g., the food web) (Gu et al., 2017; Lavoie et al., 2010; Liu et al., 2018; Luoma and Rainbow, 2008; Ofukany et al., 2014; Tao et al., 2012).

Fishes are recognized accumulators capable of biomagnifying metals on different trophic levels (Ahmed et al., 2015; Islam et al., 2015; Saha et al., 2016; Taweel et al., 2013; Tyokumbur, 2016). Thus, these organisms are widely used as bioindicators for the monitoring of metals in aquatic environments (Burger et al., 2002; Gu et al., 2017; Zhong et al., 2015). The accumulation of metals in fishes depends on the physico-chemical properties of the water as well as ecological and physiological characteristics of individuals (Jayaprakash et al., 2015). Several studies have demonstrated that the bioaccumulation pattern is influenced by

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physiological condition, trophic position, age, body size, feeding behavior, sex and a high metabolism during the reproductive period (Amundsen et al., 1997; Chi et al., 2007; Farkas et al., 2003; Singh et al., 2007; Yilmaz, 2003).

Trace elements can bioaccumulate in vital organs, such as the liver, kidneys, gills and muscles, to concentrations hundreds of times higher than those detected in the surrounding environment. Thus, levels of metals accumulated in different tissues can differ significantly due to distinct metabolic mechanisms (Chua et al., 2018; Varol et al., 2022). However, most studies investigating metal accumulation in fishes have focused mainly on muscle tissue, which is the main edible portion consumed by humans (Ge et al., 2020; Varol et al., 2020). Studies on metal contents in other tissues are scarce and further research is needed to reveal the tissue-specific bioaccumulation of metals in fish species (Ge et al., 2020; Vetsis et al., 2021).

Fishes constitute the major source of animal protein for numerous human populations. Coastal communities are major consumers of fishes due to practical issues (easy access) as well as cultural and socioeconomic factors (Lopes et al., 2016). Therefore, studies on the contamination of muscle tissue in fishes by metals have intensified. Determining levels in the edible portion is critical to protecting human health, as the consumption of contaminated fish is the primary transfer route to humans and poses serious a health problem for consumers (da Silva Rabitto et al., 2011; Franco-Fuentes et al., 2021; Gusso-Choueri et al., 2018; Kalantzi et al., 2019; Pérez-Domínguez et al., 2012; Rajeshkumar and Li, 2018; Renieri et al., 2019; Souza-Araujo et al., 2021; Sow et al., 2013; Tang et al., 2013; Töre et al., 2021; Varol and Sünbül, 2020; Wosnick et al., 2021).

The Gulf of Maranhão is located to the south of the Amazon River delta in a region with dense mangroves and high river discharge. This region is also home to several business enterprises and a port complex with intense industrial activity (da Silva Castro et al., 2019; Silva et al.,

2018). However, information regarding the possible impact of human actions on components of the aquatic biota is scarce and there is a need for studies that evaluate the occurrence of metals in the muscle tissue of fishes of different trophic guilds as biomarkers in estuarine environments near urban-industrial centers. Therefore, the aim of the present study was to determine contents of cadmium, chromium, copper, iron and manganese in fishes of different trophic levels caught in the three bays (São Marcos, São José and Arraial) that form the Gulf of Maranhão.

## 2. Material and methods

### 2.1. Study area

The Gulf of Maranhão is located in the northernmost portion of the state of Maranhão and is composed of the São Marcos, São José and Arraial Bays, which are separated by Maranhão Island (Fig. 1). São Marcos Bay lies to the west of the island and constitutes an active estuary where the Mearim and Pindaré Rivers converge. The most intense tides in the gulf are recorded for the mouth of this bay. São José Bay lies to the east of the island, has shallow depths and receives waters from the Itapecuru and Munim rivers (Silva et al., 2018). Arraial bay lies to the southeast of the island and is formed by several tributaries of the Itapecuru river basin. It also exchanges masses of water with São Marcos Bay through the Mosquitos strait. The Gulf of Maranhão is classified as a high-energy region and is constantly submitted to the combined effects of coastal currents generated by different hydrodynamic forces, such as tides, waves and trade winds, as well as the discharge of several rivers, forming a large estuarine complex.

### 2.2. Sampling

Fish species were caught from eleven estuaries located in the São

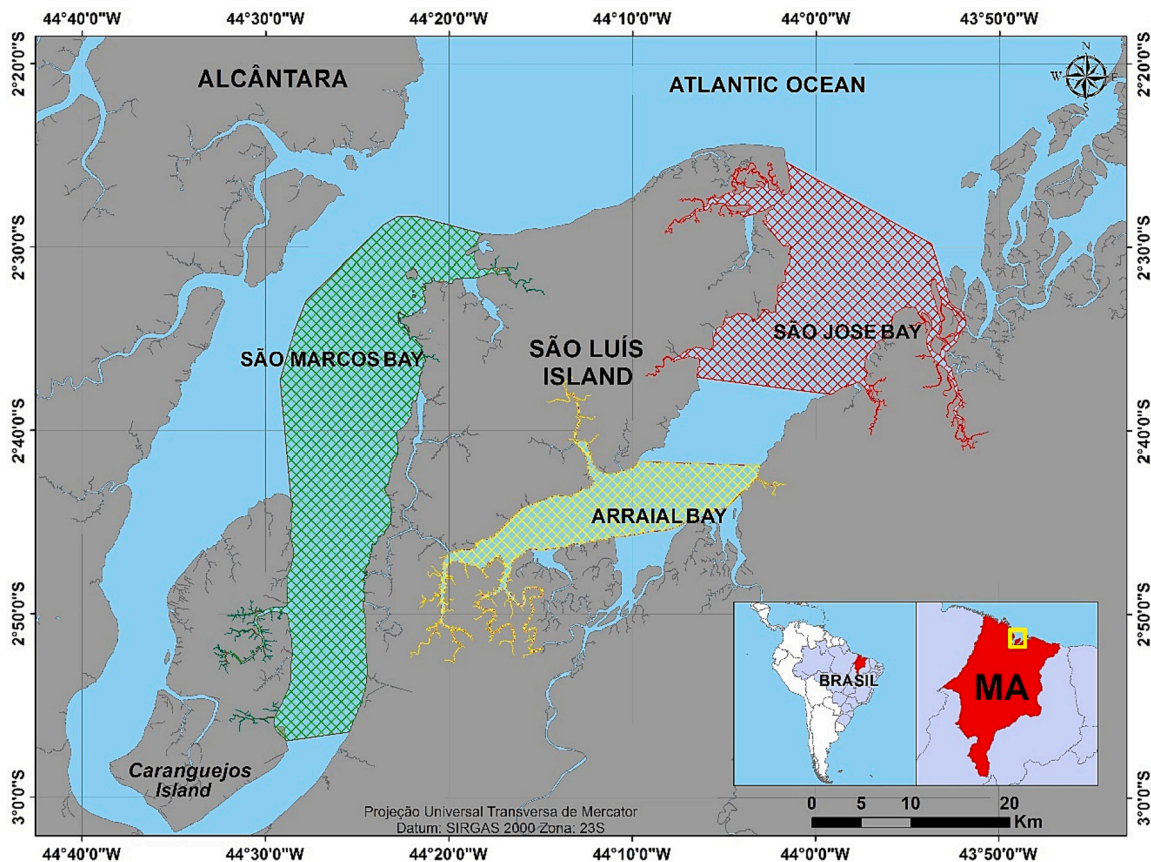


Fig. 1. Estuarine complex of Gulf of Maranhão and bays studied in coastal zone of Brazilian Amazon.

Marcos, Arraial and São José Bays with the aid of a trawnet measuring 6.0 m in length and 3.0 m in width with a mesh opening of 1.0 cm. All specimens were placed in plastic bags, stored in ice and transported to the lab for identification using specialized literature (Carpenter, 2002; Harrison, 2002; Marceniuk, 2005; Marceniuk et al., 2012). Five species with different eating habits were analyzed: *Sciades herzbergii* (omnivore), *Cetengraulis edentulus* (herbivore), *Macrodon ancylodon* (carnivore), *Pseudauchenipterus nodosus* (detritivore) and *Stellifer rastrifer* (carnivore).

### 2.3. Muscle digestion

The individuals were measured. Part of the edible portion (muscle tissue) was removed and was placed into Falcon tubes (15 mL). All samples were macerated and portions of approximately 0.5 g were weighed. Triplicate samples were prepared from each of the specimens. The samples were dehydrated at 60 °C until reaching a constant weight (approximately two weeks), then ground with a pestle and mortar and stored in Teflon tubes until digestion. A Mars 6 system (CEM Company, USA) was used for microwave-assisted digestion. Approximately 0.1 g of muscle tissue was placed into a Teflon microwave digestion vessel. About 2 mL of HNO<sub>3</sub> (65 %) were added to each tube, which were heated to 200 °C for 20 min, with a 20-min ramp-up time in a microwave oven (Batch/Mars 6/CEM), followed by a 20-min cooling time following the protocol for animal tissue samples. After digestion, each sample was transferred to clean volumetric flasks and diluted with deionized water to 50 mL. The samples were stored in a refrigerator at 5 °C until analysis.

### 2.4. Decontamination of flasks and filters

The decontamination of the labware followed the recommendations of APHA (1998), with adaptations. The material was maintained in an Extran® bath (20 %) for 24 h, rinsed with ultrapure water, placed into an acid bath (HNO<sub>3</sub> at 10 %) for 24 h, rinsed again and dried in a laboratory oven (SP Labor - SP-100/180-A) at 60 °C. The fiberglass filters (0.45 µm and 47 mm; Merck Millipore) were first washed with 60 mL of ultrapure water, followed by 20 mL de HNO<sub>3</sub> (10 %) and 20 mL of ultrapure water.

### 2.5. Analytical procedures

Concentrations of cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe) and manganese (Mn) in the muscle tissue were determined at the Analytical Chemistry Multiuser Center of the Department of Chemistry of the Federal University of Maranhão using an inductively coupled plasma optical emission spectrometer (ICP-OES, Spectro Analytical Instruments, model 9800, ICPE-9820, Shimadzu). Emission lines were selected based on sensitivity. Analyses were performed four times using the axial view with plasma condition 1 and an exposure time of 30 s. Calibration curves were created from six analytical standard multi-element solutions (SpecSol, Brazil) in the range of 0.001 mg/L to 1 mg/L and the final concentration of metals was expressed as mg/kg. The operational conditions of the device are listed in Table 1. Limit of detection (LOD) values were calculated as the concentrations

**Table 1**  
ICP-OES operating conditions for analysis of metals in muscle tissue of fishes.

Elements	Wavelength (nm)	LOD	LOQ	r <sup>2</sup>
Cd	226.502	0.10	0.30	0.99997
Cu	324.754	0.50	1.70	0.99998
Cr	267.716	0.30	1.00	0.99996
Fe	238.204	0.10	0.30	0.99977
Mn	257.610	0.03	0.01	0.99998

LOD = limit of detection. LOQ = limit of quantification; r<sup>2</sup> = coefficient of determination.

corresponding to signals equal to three times the standard deviation of the blank solution signal at a plasma flow rate of 7.0 L min<sup>-1</sup>.

The limit of detection (LOD) and limit of quantification (LOQ) were calculated according to the Shimadzu specifications. The exactness of the method was calculated using reference materials (TORT-3: lobster hepatopancreas; ERM®-CE278k: Mussel). Average recovery rates were 80 % for Cd, 88 % for Cr, 89 % for Cu, 91 % for Fe and 83 % for Mn.

### 2.6. Data analysis

Statistical analyses were performed with the aid of the PAST 3.14 program, with a significance level of 0.05 (Hammer et al., 2001). One-way analysis of variance (ANOVA) was used to test possible differences in the concentrations of metals between species and bays when the presuppositions of normality and equalness of variances were met (Zar, 2010). When these presuppositions were not met, the Kruskal-Wallis test was used, followed by the Mann-Whitney test (when significant results were found) and Bonferroni correction for the pairwise comparisons of the magnitude of difference in the variables between bays. The metal concentration data in estuarine fishes were log-transformed before further multivariate analysis. Principal component analysis (PCA) was performed using the correlation matrix as the basis to analyze the origin and associations of metals between species and bays.

## 3. Results

A total of 45 samples were analyzed from five marine-estuarine species (*Sciades herzbergii*, *Cetengraulis edentulus*, *Macrodon ancylodon*, *Pseudauchenipterus nodosus* and *Stellifer rastrifer*) caught in estuaries located in the São Marcos, Arraial and São José Bays, which form the Gulf of Maranhão.

The concentrations of metals determined in the muscle tissue were compared to maximum permissible limits for edible fish tissues established by ANVISA (Brazilian National Sanitary Vigilance Agency) Decree n° 55.871/65 and Ordinance n°685/88, the European Commission Regulation (EC, 2006) and the Food and Agriculture Organization (FAO, 1983) (Table 2).

Concentrations above the maximum permitted limit were found for cadmium (Cd) in individuals of *Cetengraulis edentulus* and *Macrodon ancylodon* and chromium (Cr) in individuals of *Macrodon ancylodon* and *Pseudauchenipterus nodosus*. Copper (Cu) was not detected above the maximum limit in any of the specimens. Although there are no national laws regulating concentrations of iron (Fe) and manganese (Mn) in the muscle tissue of fishes, concentrations of these elements were found in all species analyzed, with some specimens exhibiting very high concentrations in comparison to the overall average of the study.

Concentrations of Mn and Fe were highest in *C. edentulus* and *M. ancylodon* and lowest in *S. rastrifer*. Concentrations of Cd were highest in *M. ancylodon* and lowest in *S. herzbergii*. Concentrations of Cu were highest in *S. herzbergii* and lowest in *S. rastrifer*. *P. nodosus* and *C. edentulus* had the highest and lowest concentrations of Cr, respectively (Fig. 2).

Regarding the sampling locations, São Marcos Bay had the highest mean concentration of Fe and the lowest means for Cr and Cu. São José Bay had the highest mean for Mg and the lowest mean for Cd. Arraial Bay had the highest means for Cd and Cu and the lowest mean for Mn (Fig. 3).

The ANOVA results indicated a significant difference between the São José and Arraial Bays for Cd concentrations (Fig. 4). Different concentrations of Cr, Cu and Mn were found between São Marcos Bay and São José Bay. In contrast, no significant differences among the bays were found for Fe.

The first two axes of principal component analysis (PCA) of the concentrations of heavy metals for each fish species per sampling location and trophic level explained 68.3 % of the total variability in the data (Fig. 5). The significance of the axes was tested using a random Broken

**Table 2**

Concentrations of metals in estuarine fishes caught in Gulf of Maranhão. Bold type indicates values above maximum limit permitted. LD = limit of detection. O = omnivore; H = herbivore; C = carnivore; D = detritivore.

Location	Species	Concentration in mg/kg					Feeding habit
		Cd	Cr	Cu	Fe	Mn	
São José Bay	<i>Sciades herzbergii</i>	<b>0.12</b>	<b>0.19</b>	<LD	4.92	0.16	O
		0.01	0.07	0.09	<LD	0.05	
		<LD	0.03	<LD	5.25	0.12	
		0.01	0.01	0.03	4.23	0.08	
		0.01	0.06	0.15	3.46	0.05	
		0.01	0.01	0.01	0.15	0.01	
	<i>Cetengraulis edentulus</i>	0.01	0.05	0.16	0.16	105.53	H
		<LD	<LD	<LD	4.88	0.27	
		0.01	0.06	0.05	4.05	84.53	
		0.01	0.05	0.12	4.53	0.04	
		0.03	<b>0.13</b>	0.24	3.30	0.17	
		0.01	0.07	0.08	2.08	0.04	
	<i>Pseudauchenipterus nodosus</i>	<LD	0.05	0.08	1.96	0.03	D
		<LD	0.40	<LD	8.98	0.23	
		<LD	0.05	<LD	10.65	0.13	
		<LD	0.02	0.05	2.68	0.19	
		<LD	0.01	<LD	2.99	0.02	
		<LD	0.01	<LD	2.08	0.12	
<i>Macrodon ancylodon</i>	<LD	0.01	0.01	1.37	0.10	C	
	<LD	<LD	<LD	1.16	0.03		
	<LD	0.01	<LD	3.93	0.02		
	<LD	0.01	<LD	0.91	0.00		
	<LD	<b>0.11</b>	<LD	4.55	<LD		
	<LD	0.07	<LD	5.48	0.03		
São Marcos Bay	<i>Pseudauchenipterus nodosus</i>	<LD	<LD	<LD	2.76	0.01	D
		<LD	0.01	<LD	2.55	0.01	
		<LD	<LD	<LD	1.01	<LD	
		<LD	0.01	<LD	16.97	0.63	
		0.58	0.01	0.14	2.65	0.02	
		<b>1.17</b>	<LD	<LD	5.78	0.20	
	<i>Cetengraulis edentulus</i>	<b>1.39</b>	<LD	<LD	6.24	0.35	H
		0.96	0.01	0.02	2.21	0.06	
		0.58	0.04	0.12	22.05	0.14	
		0.72	0.02	0.44	2.94	0.02	
		<b>1.83</b>	<b>0.14</b>	<LD	3.05	0.01	
		0.57	<LD	<LD	2.31	0.01	
Arraial Bay	<i>Stellifer rastrifer</i>	0.58	<LD	<LD	2.55	0.20	D
		0.57	<LD	<LD	3.43	0.08	
		0.57	<LD	<LD	2.26	0.08	
		0.57	0.05	0.017	3.93	0.115	
		<LD	0.01	<LD	2.52	0.04	
		<LD	<LD	<b>6.58</b>	1.64	0.05	
<i>Sciades herzbergii</i>	<LD	<LD	<LD	1.27	0.27	O	
	<LD	<LD	<LD	1.82	0.10		
	<LD	<LD	<LD	1.77	0.06		
	<LD	<LD	<LD	1.77	0.06		
Limit established by legislation (mg/kg)		1.0 <sup>a, b, d</sup>	0.1 <sup>b</sup>	30.0 <sup>b, c</sup>	–	–	

<sup>a</sup> ANVISA, Ordinance no 685, 27 August 1998.

<sup>b</sup> BRAZIL, Decree n° 55.871, 26 March 1965.

<sup>c</sup> FAO, Food and Agriculture Organization, 1983.

<sup>d</sup> EC, European Commission.

Stick model with 9999 bootstrap replications (Jackson, 1993), which indicated that Components 1 and 2 alone were sufficient to represent the factorial variance.

Component 1 (41.44 %) was positively correlated with Cd and Fe, indicating higher concentrations of these elements in the São Marcos and Arraial Bays associated with the two carnivorous species (*S. rastrifer* and *M. ancylodon*) and the herbivore (*C. edentulus*). Mn in the species *C. edentulus* and *M. ancylodon* was negatively correlated with Component 1, with a tendency toward a greater concentration in São José Bay. These species have herbivorous and carnivorous habits, respectively.

For Component 2 (26.79 %), Cu was the element with the strongest correlation, being negatively correlated with the axis, with greater concentrations in São Marcos Bay associated with *M. ancylodon* (carnivore) and Arraial Bay associated with *S. herzbergii* (omnivore). Cr was positively correlated with Component 2, with higher concentrations in São José Bay associated with all trophic guilds analyzed in the present study.

#### 4. Discussion

The present results revealed the occurrence of Cd, Cr, Cu, Fe and Mn in all species collected from the Gulf of Maranhão. Moreover, concentrations of metals above the maximum limit permitted were found in all three bays studied. Similar results were reported by Silva et al. (2021), who found average metal levels above the limits established by ANVISA in most fish species caught in the municipality of Salvador in the state of Bahia, Brazil, and Franco-Fuentes et al. (2021), who investigated food security and the risk of toxicity in commercial fishes from the Galapagos Marine Reserve on the coast of Ecuador, the results of which indicated concentrations higher than the maximum concentrations permitted by the European Commission and the Food and Agriculture Organization. Fishes constitute an important structural component of the food chain in aquatic environments and are an essential food source, including for the human population. Therefore, knowledge on contamination levels in fishes is particularly important to monitoring the potential effects of

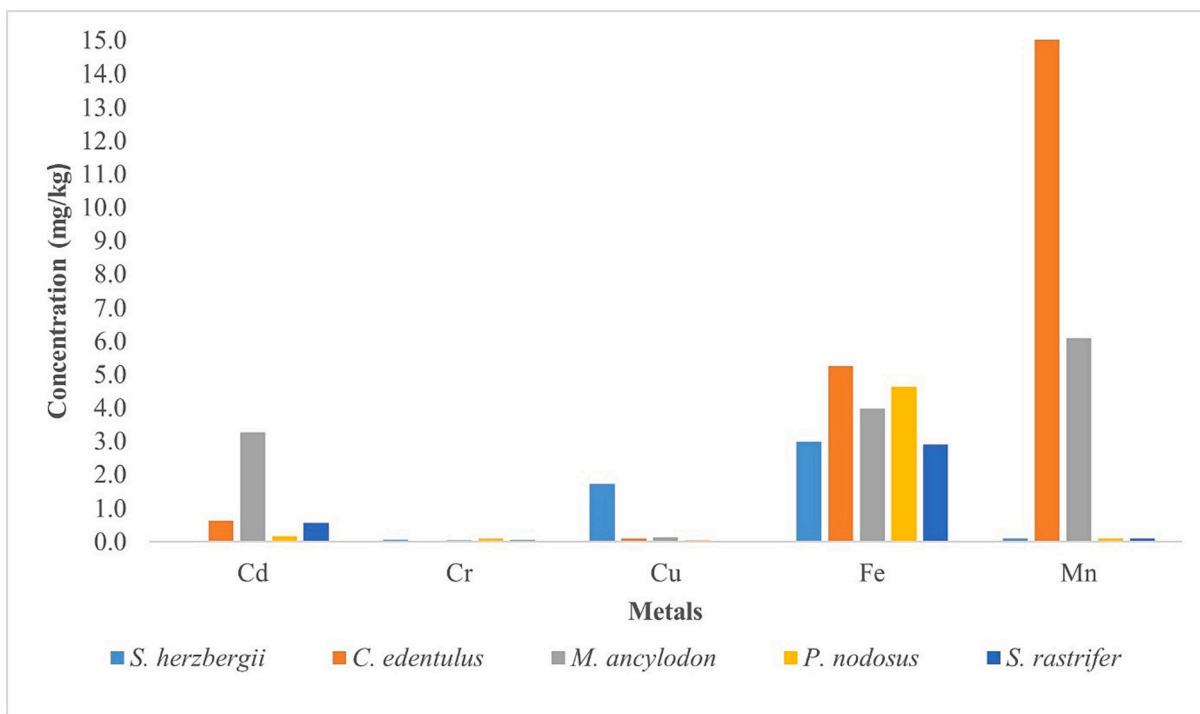


Fig. 2. Concentrations of Cd, Cr, Cu, Fe and Mn in muscle tissue of fish species caught in Gulf of Maranhão.

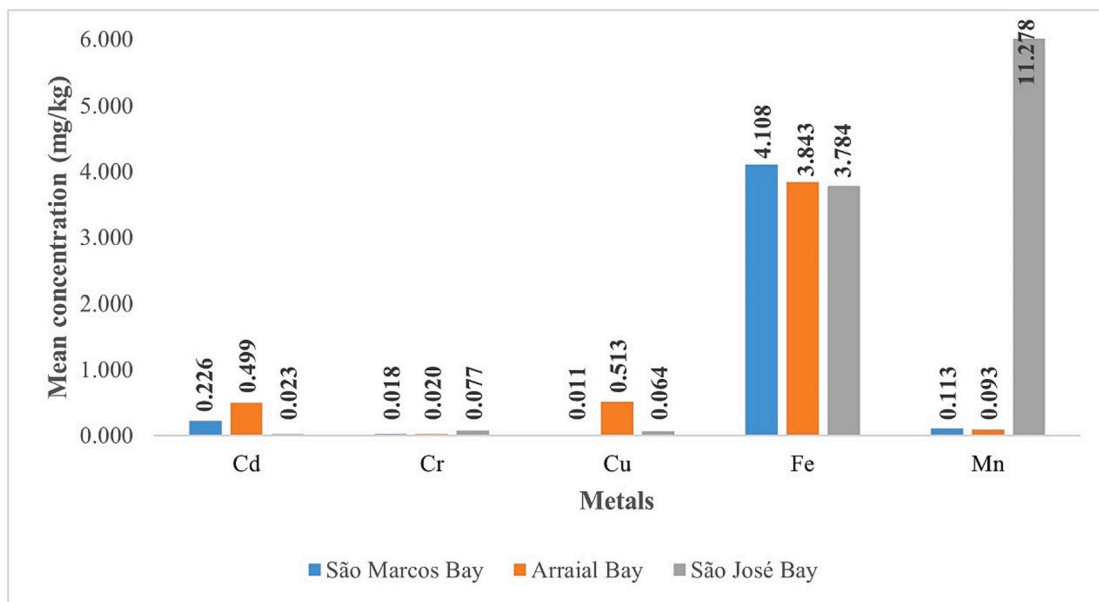


Fig. 3. Concentrations of Cd, Cr, Cu, Fe and Mn in muscle tissue of fish species caught in different bays of Gulf of Maranhão.

substances found in these organisms (Burger and Gochfeld, 2005). The increase in industrialization and urban expansion in the Industrial District of the city of São Luís has led to the introduction of undesirable quantities of pollutants in the aquatic environment (Santos et al., 2019).

Mean Cd and Cr levels found in the muscle tissue of the fish species were above the permissible limits set by different international or national guidelines. In previous studies, Vetsis et al. (2021) found bio-magnification in the food chain and Töre et al. (2021) found high Cd concentrations associated with the use of pesticides, while Cr levels were below the established legal limits in the Tigris River in Turkey. In contrast, Rajeshkumar and Li (2018) found that the edible parts of commonly available fishes were not heavily burdened with these metals

in Meiliang Bay of Taihu Lake in China. Cd is highly toxic to the kidneys and accumulates in higher concentrations in proximal tubular cells. Cr can induce DNA damage, causing chromosome mutations as well as changes in the replication and transcription of DNA (Jaishankar et al., 2014; O'Brien et al., 2001; Matsumoto et al., 2006).

High concentrations of Fe were found in all species analyzed (range: 0.161 to 22.050 mg/kg). Santos et al. (2019) report similar results in studies conducted in the estuary of the Cachorros River, which is located in the transition area between São Marcos Bay and Arraial Bay. River discharge one of the major sources of dissolved Fe in oceans (Bergquist and Boyle, 2006). Thus, the strong freshwater inputs to the Gulf of Maranhão constitute a determinant factor. Moreover, the port receives

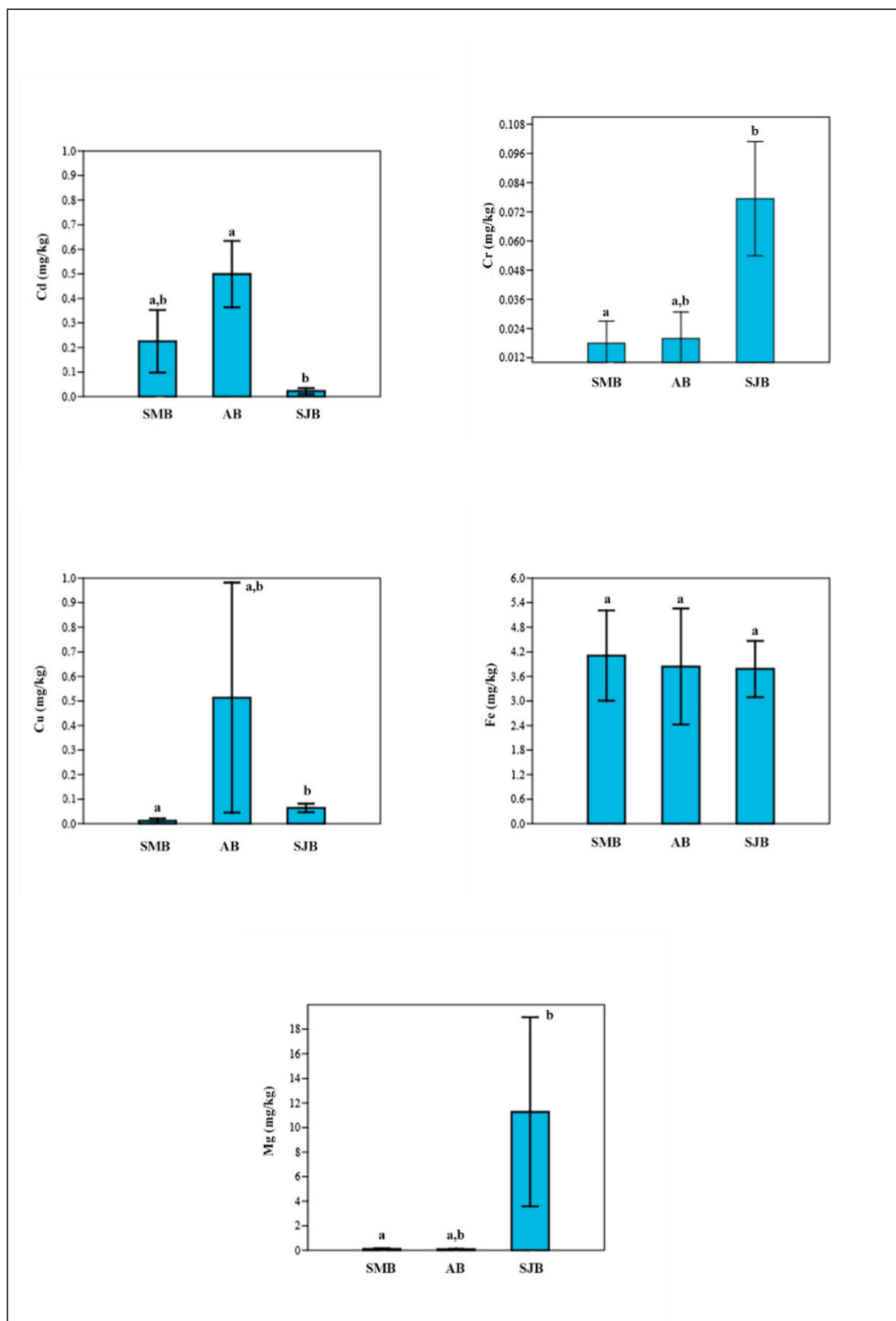


Fig. 4. ANOVA results for concentrations of metals (Cd, Cr, Cu, Fe and Mg) in each sampling location (SMB – São Marcos Bay; AB – Arraijal Bay; SJB – São José Bay).

cargo ships that transport iron, which may explain the high concentration of this element in the samples analyzed.

High Mn concentrations were found in the samples analyzed, especially species caught in São José Bay, suggesting an association with focal sources in the region. The accumulation of this metal does not seem to be influenced by the geochemical patterns of the gulf, as the

concentration was not homogeneous among all sampling locations. The most common source of this element in the estuarine system is the discharge of industrial effluents and domestic sewage (Butler and Timperley, 1996). Although there is no specific legislation regarding the quantity of Mn in foods, the National Sanitary Vigilance Agency (ANVISA) reports that the recommended daily intake of this metal is

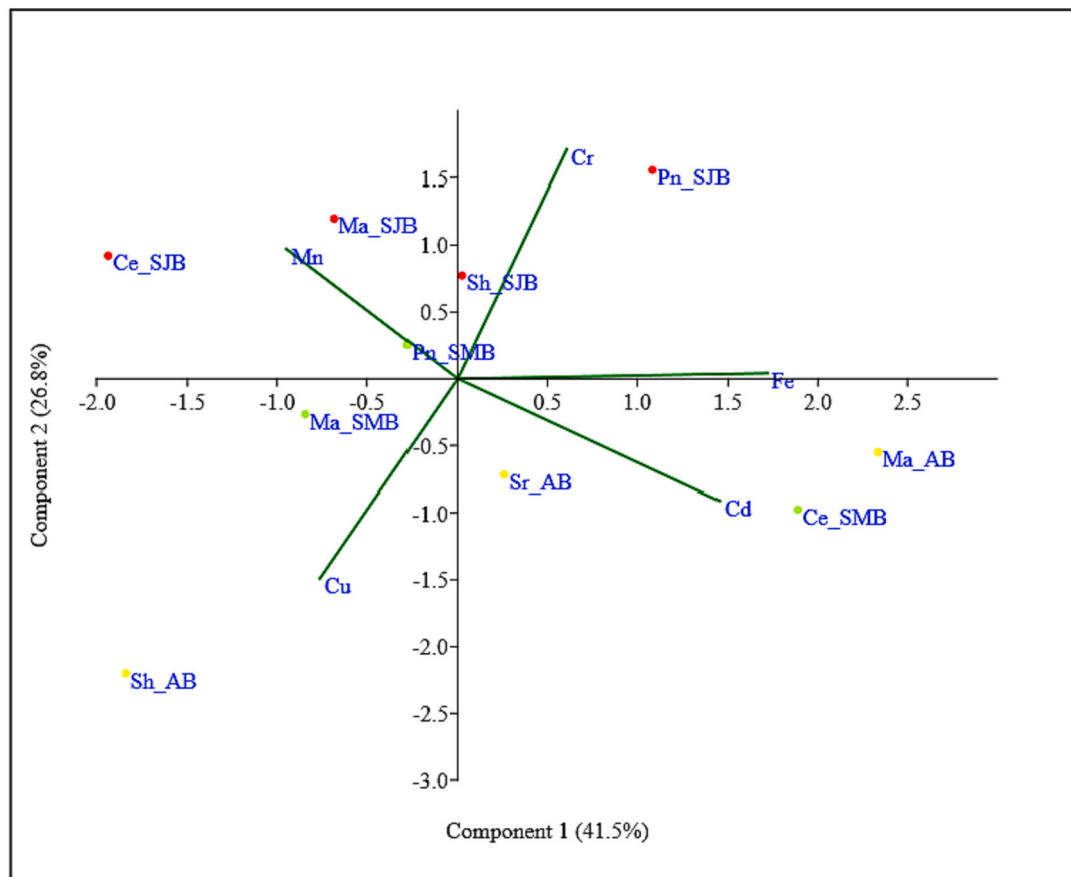


Fig. 5. Principal component analysis showing relations among bioaccumulation of metals (Cd, Cr, Cu, Fe and Mn), species on different trophic levels and capture locations. Sh = *Sciades herzbergii*; Ce = *Cetengraulis edentulus*; Ma = *Macrodon ancylodon*; Pn = *Pseudauchenipterus nodosus*; Sr = *Stellifer rastrifer*; AB = Arraial Bay; SJB = São José Bay; SMB = São Marcos Bay.

360 mg for adults (Brasil, 2005). The results of this study are consistent with data from other studies comparing the concentration of Mn in different tissues of various fish species (Ghosn et al., 2020; Grotto et al., 2012; Varol et al., 2022; Varol and Sünbül, 2020; Vetsis et al., 2021).

Concentrations of Cu found in the muscle tissue of the species do not pose a threat to the health of the fishes or constitute a risk of contamination for the local population, as the levels were well below the maximum acceptable limit. According to Kalay and Canli (2000), Pereira et al. (2010) and Lima et al. (2015), copper is essential to the organism and is therefore easily regulated by the metabolism, making its accumulation unlikely.

The spatial analysis indicated significant differences for Cd, Cr, Cu and Mn. This may be related to local characteristics and human activities, which can exert an influence on the levels of assimilation of species in different locations. In contrast, the concentrations found in the present study were lower than those reported for highly impacted systems, such as Guanabara Bay (Abreu et al., 2016), Sepetiba Bay (Fonseca et al., 2013) and the Paranaguá Estuarine Complex (dos Anjos et al., 2012; Sá et al., 2006), all of which are located in Brazil, as well as the Port of Toulon in France (Bouchoucha et al., 2018).

Copper was correlated more with the São Marcos and Arraial Bays and the species *M. ancylodon* (carnivore) and *S. herzbergii* (omnivore), which may reflect high levels of binding proteins in the muscle tissue of these species (Subotić et al., 2013). Cadmium and iron were strongly associated with the Arraial and São José Bays, whereas Mn was strongly associated with only the São José Bay, with higher concentrations found in carnivorous and herbivorous species. Relatively high concentrations of these metals may be related to feeding habits, as carnivorous fishes move actively through the water in search of prey and are known to

accumulate high levels of heavy metals in the body due to bioaccumulation (Karadede et al., 2004; Varol et al., 2020). Although herbivores tend to accumulate a lower quantity of metals, absorption can occur by processes of ion transport through membranes (Jabeen et al., 2012; Mert et al., 2014; Pereira et al., 2010). Levels of elements in fishes can also be influenced by seasonal and biological differences (species biology and proximate composition, size, dark/white muscle, age, sex and sexual maturity) as well as environmental factors, such as water chemistry, salinity, temperature and contaminants (Alam et al., 2002; Kalantzi et al., 2019, 2016, 2013).

## 5. Conclusions

In this study, levels of Cd, Cr, Cu, Fe and Mn were investigated in the muscle tissue of *Sciades herzbergii*, *Cetengraulis edentulus*, *Macrodon ancylodon*, *Pseudauchenipterus nodosus* and *Stellifer rastrifer* caught in the three bays of the Gulf of Maranhão. Concentrations of metals above the maximum permitted limit were found in all three bays studied. Mean levels of Cd and Cr were above the limits set by different international or national guidelines. High concentrations of Fe were found in all species analyzed. High concentrations of Mn were also found in the samples, especially in species caught in São José Bay. Concentrations of Cu found in the muscle tissue of the species do not pose a threat to the health of the fishes or constitute a risk of contamination for the local population, as the levels were well below the maximum acceptable limit. The spatial analysis indicated significant differences for Cd, Cr, Cu and Mn. Copper was correlated more with the São Marcos and Arraial Bays as well as the species *M. ancylodon* (carnivore) and *S. herzbergii* (omnivore). Cadmium and iron were strongly associated with Arraial and São José Bays, while

Mn was strongly associated with only São José Bay, with higher concentrations found in carnivorous and herbivorous species.

The present analyses indicate that fish species are responding to the direct effects caused by environmental changes of an anthropogenic origin. The region has evidence of contamination by different types of metals. The effects of this contamination are not restricted to São Marcos Bay, which is an apparently more anthropized environment, but also extend to the Arraial and São José Bays, as evidenced by the high concentrations of metals found in fishes caught in these locations. High concentrations of the elements analyzed were found in all trophic categories considered, suggesting risks with regards to the consumption of products originating from artisanal fishing activities in the region. The findings suggest that fishes can be used as biomarkers of contamination by metals in the Gulf of Maranhão. However, there is a need to investigate other matrices to test the bioaccumulation of metals, such as sediments and macroinvertebrates.

### 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.

### Data availability

Data will be made available on request.

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