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Metal bioavailability and distribution in the fish community in a tropical estuary, Sepetiba Bay, Rio de Janeiro, Brazil

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RESUMO

A Baía de Sepetiba possui uma riqueza de espécies de peixes (total de 148), assim como, uma vasta área de manguezais e inúmeras ilhas rochosas, importantes locais de reprodução da vida marinha. Este ambiente peculiar da costa brasileira contém um dos mais importantes centros industriais do sudeste do Brasil. Este local vem sendo impactado há décadas pelo lançamento de emissões industriais (outras) e efluentes com altas cargas metálicas. Os intervalos de concentração de metais no músculo de peixes das espécies *Micropogonias furnieri, Genidens genidens, Cathorops spixii, Notarius grandicassis, Diapterus rhombeus, Selene vomer, Prionotus punctatus, Citharichthys spilopterus, Achirus lineatus, Trinectes Hypanistanus, Symphatus guttus* : Al 0,02-555,9 µg g^{-1} dw, As: 00002-20,1 µg g^{-1} dw, Cd: <0,0002-0,2 µg g^{-1} dw, Cu: 0,2-2,3 µg g^{-1} dw, Fe: <0,02-244,9 µg g^{-1} dw, Zn: 0,5-227,3 µg g^{-1} dw e Pb: <0,001-1,3 µg g^{-1} dw). O teste de Kruskal-Wallis revelou diferenças significativas (p <0,05) nos teores de As, Cu, Fe, Pb e Zn entre as espécies de peixes. Análises químicas do material particulado em suspensão durante a operação de dragagem revelaram as seguintes concentrações de metais Al (6059 ± 6268 µg g^{-1}), Cd (0,2 ± 0,5 µg g^{-1}), Cu (29 ± 29 µg g^{-1}), Zn (332 ± 892 µg g^{-1}) e Pb (52 ± 70 µg g^{-1}). Os fatores de bioacumulação calculados a partir da fração de metais das frações biodisponíveis do sedimento e do metal total no material particulado em suspensão apresentaram valores inferiores aos do músculo dos peixes. O arsênio foi encontrado em níveis acima do limite máximo para consumo humano de acordo com a legislação brasileira. No entanto, a probabilidade estimada do risco de ingestão de metais via consumo de peixes mostrou que o consumo de todas as espécies apresentou baixo risco.

Palvras-chave: biodisponibilidade, fator de acumulação para biosedimento (BASF), estuário, contaminação, quociente de risco

ABSTRACT

Sepetiba Bay has a wealth of fish species (total 148) as well as a vast area of mangroves and numerous rocky islands, which are important sites of reproduction for marine life. This peculiar environment of the Brazilian coast hosts one of the most important industrial centres of south-eastern Brazil. This site has been impacted for decades by the release of industrial emissions and effluents with high metal loads by the steel industry. The ranges of metal concentrations in fish muscle from the species *Micropogonias furnieri*, *Genidens genidens*, *Cathorops spixii*, *Notarius grandicassis, Diapterus rhombeus*, *Selene vomer*, *Prionotus punctatus*, *Citharichthys spilopterus*, *Achirus lineatus*, *Trinectes paulistanus*, *Symphurus tessellatus* and *Hypanus guttatus* were measured (Al: 0.02-555.9 µg g^{-1} d.w., As: 0.0002-20.1 µg g^{-1} d.w., Cd: <0.0002-0.2 µg g^{-1} d.w., Cu: 0.2-2.3 µg g^{-1} d.w., Fe: <0.02-244.9 µg g^{-1} d.w., Zn: 0.5-227.3 µg g^{-1} d.w. and Pb: <0.001-1.3 µg g^{-1} d.w.). The Kruskal-Wallis test revealed significant differences (p<0.05) in the As, Cu, Fe, Pb and Zn contents among fish species. The monitoring of suspended particulate matter during dredging operation revealed the mean metal values for Al (6059±6268 µg g^{-1}), Cd (0.2±0.5 µg g^{-1}), Cu (29±29 µg g^{-1}), Zn (332±892 µg g^{-1}), and Pb (52±70 µg g^{-1}). The results of bioaccumulation in fish calculated from the bioavailable sediment fractions and suspended particulate matter showed lower values than those in fish muscle. Arsenic was found at levels above the maximum limit for human consumption according to Brazilian legislation. However, the estimated probability and risk of metal intake via fish consumption showed that the consumption of all species presented low risk.

Keywords: bioavailability, biosediment accumulation factor (BSAF), estuary, contamination, hazard quotient

1 INTRODUCTION

Environmental degradation is led by the imbalance between the growth of the human population and industrial activity and environmental conservation. Metal pollution in coastal zones has been reported in diverse sites in Brazil (MIRLEAN *et al.*, 2009; FONSECA *et al.,* 2013; KIM *et al.*, 2016), showing that the input of metals to aquatic ecosystems is affected by several urban and industrial sources, such as deforestation, untreated sewage, street run off, and chemical and petrochemical industry activities.

Sepetiba Bay has been impacted since the end of 1940s by the first coal terminal serving the National Steel Company and by ore processing industries since the 1960s (BARCELLOS; LACERDA 1994). Fiszman *et al.* (1984) conducted the first study of metal contamination in the bay in the early 1980s. Furthermore, the increase in domestic wastewater discharged in the bay is an additional source of contamination (COPELAND *et al.*, 2003). According to the 2010 census conducted by the Brazilian Institute of Geography and Statistics (IBGE, 2010), the Sepetiba Bay drainage basin has an estimated population of 403,643 individuals, many of whom live in houses without sewage treatment.

The presence of high metal concentrations, mainly zinc and cadmium, was reported as a consequence of activities from the steel industry (CARVALHO GOMES *et al.*, 2009; RIBEIRO *et al.*, 2013). The company discharged 24 tonnes $year^{-1}$ of Cd and 3,660 tonnes $year^{-1}$ of Zn in the bay until its closure after 30 years of activity, causing a nearly 200-fold increase in the deposition of these metals in the coastal environment (BARCELLOS *et al.*, 1991; BARCELLOS; LACERDA 1994). Additionally, arsenic (As) contamination in bay sediments is related to the arsenic trioxide (As_2O_3) used for coal purification (MAGALHÃES *et al.*, 2001). The discharge of effluent into the bay by Companhia Siderúrgica Mercantil Ingá S.A. ceased in 2008 with the start of the remediation project, which concluded in 2015. During remediation, sediment from some areas with higher metal levels was removed by dredging, and the material was disposed of in an underwater confined disposal facility.

Although many cleaning actions have been performed in the bay, some areas still have high concentrations of metals in sediment (TONHÁ *et al.*, 2020; MONTE *el al.*, 2015). Moreover, dredging can remobilize and resuspend metals from the anoxic sediment layer, facilitating pollutant biodisponibilization (GOOSSENS; ZWOLSMAN 1996; MONTE *el al.*, 2015). The frequent dredging to maintain navigability of the bay could represent a risk of metal contamination to marine life if sediment resuspension control is not undertaken during this activity.

Sediment toxicity evaluation has been employed using fish species as sentinel organisms (HARTL 2002; BERVOETS; BLUST 2003). Indeed, sentinel fish species monitoring is widely used to assess the degree of accumulation of pollutants and the effects on their health state (FITZGERALD *et al.*, 1999; DE LA TORRE *et al.*, 2000; NENDZA 2002; JIMENEZ-TENORIO *et al.,* 2007).

Fish have been used to assess the environmental risk of metal contamination as they can assimilate it through their gills and diet (PHILLIPS 1977; VAN DER OOST *et al.* 2003). In the present study, the evaluation of fish contamination is necessary since this region has intense fishery activity. Considering that the consumption of contaminated fish represents a risk to human health, the aim of the present study was to evaluate metal concentrations in fish and calculate metal transference from the sediment and particulate matter of Sepetiba Bay during dredging.

2 MATERIALS AND METHODS

2.1 STUDY AREA

Sepetiba Bay (Figure 1), which is situated 60 km west of Rio de Janeiro, has an area of 447 km^2 during high tide and 419 km^2 during low tide. The bay has an average depth of 6 m and has brackish water and seawater due to its connection with the Atlantic Ocean. The drainage basin is formed by the following rivers: Itingussu, Piração, Porto, Engenho Novo, Ita, Cação, Piraquê, Guandu, Guarda and São Francisco. The São Francisco River is responsible for 86% of the freshwater input to the bay (BARCELLOS *et al.* 1997; MOLISANI *et al.,* 2006).

Wastewater from the cities of Itaguaí, Mangaratiba, Japeri and Miguel Pereira, which corresponds to a population of 403,643 inhabitants (IBGE 2010), flows into the bay, as does some of the effluent from Rio de Janeiro, Nova Iguaçu, Rio Claro, Piraí, Engenheiro Paulo de Frontin and Vassouras.

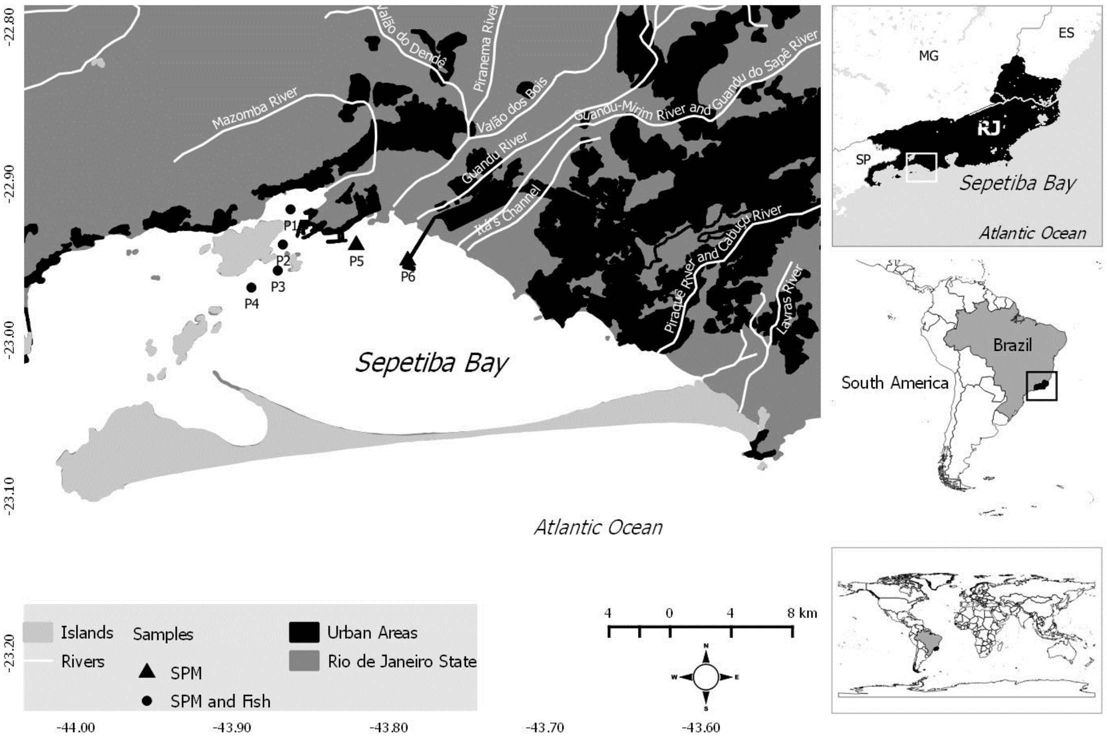

Figure 1
Map of sampling points in Sepetiba Bay

2.2 FISH SAMPLING

Fish sampling was carried out by horizontal trawling using 20 mm mesh in the middle of the net and 10 mm mesh in the funnel (10 m in length with a mouth opening of 3 m in width). Four sites were sampled (Figure 1). Trawling was conducted in circles of 30 m for 20 minutes, with the coordinates corresponding to the centre of the cycle. The collected fish were cleaned with distilled water and kept frozen in an icebox until arrival at the laboratory, where the species were identified, and biometric (weight and total length) data were collected.

Samples of the caught fish species, which included *Micropogonias furnieri* (31), *Genidens genidens* (16), *Cathorops spixii* (4), *Notarius grandicassis* (3), *Prionotus punctatus* (11), *Citharichthys spilopterus* (9), *Achirus lineatus* (3), *Trinectes paulistanus* (3), *Symphurus tessellatus* (7), *Hypanus guttatus* (3), *Diapterus rhombeus* (13) and *Selene vomer* (3), were double bagged in separate clean plastic bags, sealed and labelled accordingly.

Among the fish biological parameters, the morphometric traits and condition factor (K) were assessed. Individual biometry was carried out to determine the total length (Lt, 0.3 cm precision) and total weight (Wt, 0.005 g precision).

Afterwards, the fish were dissected using a scalpel to remove the muscle, and the tissues were lyophilized (72 h) until subsequent homogenization (maceration in agate mortar) and subsequently subjected to chemical treatment to determine the metal concentrations.

Among the caught species, *S. tessellatus* and *P. punctatus* remain within the bay during all life cycle phases (estuarine resident). The other species leave the estuary during the adult phase, returning to it only during periods of foraging and spawning. In relation to food habits, most of the studied species were carnivorous, with the exception of three omnivorous species (*G. genidens*, *C. spixii*, and *N. grandicassis*).

The health status from the length-weight relationship of the fish samples was calculated using the Fulton condition factor (WILLIAMS, 2000; RANNEY *et al.*, 2010), which was calculated by the following equation:

$$K = 100\ W/L^3 \text{ (Equation 1)}$$

where W is the total body weight of the fish (gm) and L is the total length of the fish (cm). Fulton's K was categorized as follows: K = 1: condition is poor, K = 1.2: condition is moderate, and $K \geq 1.40$: condition is relatively good.

2.3 SUSPENDED PARTICULATE MATTER (SPM) SAMPLING

In the field, surface water was collected using a Van Dorn bottle. The samples were collected monthly in 2014 (January to June) at six points (Figure 1). The water was stocked in acid-cleaned bottles (HNO_3, 2%) (SHAFER; OVERDIER, 1995). After this, in the laboratory, the water was filtered in a vacuum system using membranes of acetate cellulose with 0.45 µm pores that were precleaned in acid and Milli-Q water. The membrane with suspended particulate matter was oven dried for posterior metal analysis.

2.4 METAL ANALYSIS

Fish tissue and suspended particulate matter (SPM) were digested in a microwave (BERGHOF-SPEEDWAVE 4) (0.3 g dry muscle + 2 mL H_2O_2 + 5 mL HNO_3) (suspended particulate matter + 6 mL HNO_3). The metal concentrations were analysed with an ICP-MS (XSeries II, Thermo Fisher). The quality control analysis was verified through blank samples and a certified reference material (NIST SRM-1566B, oyster tissue). The metals Al (115.3%), Cd (87.2%), Fe (112.1%), Pb (93.7%), Cu (110.0%), Zn (107.6%) and As (105.8%) showed good to excellent recoveries.

2.5 METAL BIOCONCENTRATION FACTOR

To evaluate metal bioaccumulation by the fish, the biosediment accumulation factor (BSAF) (USERO *et al.,* 2005), which is defined as the ratio between the metal concentration in the organism and that in the sediment or suspended particulate matter (LAFABRIE *et al*., 2007; WANICK *et al.,* 2013; DIAS; NAYAK 2016), was calculated.

The BSAF was only calculated for the fish species with large numbers of samples (*G. genidens*, *M. furnieri*, *D. rhombeus*, and *P. punctatus*). We used the data in the literature on bioavailable metals in the sediment to calculate the BSAF (MONTE *et al.,* 2015; RODRIGUES *et al.,* 2017) (Table 1). For the suspended particulate matter, the metal concentration data found in the present study were applied.

2.6 ASSESSMENT OF HUMAN HEALTH RISK FROM METAL-CONTAMINATED FISH INTAKE

Two methodologies were adopted to assess the risk of contamination through the ingestion of fish contaminated with metals. First, the total concentration of metals in the muscle of the investigated fish species was compared to national (ANVISA) and international (FAO, European Commission) regulatory values. The second methodology was to estimate the probability and risk from metal intake by means of the assessment of the hazard quotient (HQ) according to the following equation (U.S. EPA, 2002):

$$HQ = \frac{ADD}{RfD} \quad \text{(Equation 2)}$$

where ADD is the average daily dose and RfD is the oral reference dose. The ADD is calculated by the equation:

$$ADD = \frac{C * IR * EF * ED}{BW * AT} \quad \text{(Equation 3)}$$

where C is the average metal concentration in fish tissue, IR is the human ingestion rate (the control population has a mean Brazilian fish intake (IBGE 2010) of 10 kg $\mathrm{year^{-1}}$ or 0.027 kg $\mathrm{day^{-1}}$ and the fishing population has an intake of 0.2 kg $\mathrm{day^{-1}}$), EF is the exposure frequency (control population: 48 days per year and fishing population: 365 days per year), ED is the average exposure duration (years, 30 years), BW is the average body weight (mean for Brazilian men and women > 20 years of 67 kg) (IBGE 2010) and AT is the average time (AT = 365 x EDd). RfD is the reference dose (mg/kg/day) (inorganic As: 0.0003 mg/kg/day; Cd: 0.001 mg/kg/day; Cu: 0.04 mg/kg/day; Pb: 0.003 mg/kg/day; and Zn: 0.3 mg/kg/day) (U.S EPA 1991).

In relation to As, we calculated the concentration of inorganic As (the most toxic species) considering that 2 to 30% of the total As in fish muscle is in the inorganic species according to Kirby and Maher 2002.

Table 1- Bioavailable metals in the sediment applied for calculation of the BSAFs (MONTE *et al*. 2015 and RODRIGUES *et al*. 2017). Values given in $\mu g\ g^{-1}$

Metal	Mean (min–max)	
	Sediment	Particulate Matter
Al		6059.2
Cd	5.13 (0.2-17.77)	-
Cu	6.5* (3.2-1.7)	28.4
Fe	6064.4 (5162.5-13600.0)	-
Pb	15.0 (5.1-35.3)	51.8
Zn	904.2 (90.3-3854.0)	332.4

*Mean metal concentration calculated based on MONTE *et al*. (2015) and RODRIGUES *et al.* (2017) (HCl extraction–before resuspension).

3 RESULTS

The SPM concentration distribution collected in 2014 varied from 2.86±1.81 mg L^{-1} in March to 34.36±13.04 mg L^{-1} in February on average. In general, the data presented a tendency towards higher values close to the mouth of the São Francisco channel and Guandu River. Rodrigues et al. (2009) observed values of suspended particulate matter similar to those found by this study, which varied from 11.35 to 32.2 mg L^{-1} in the wet period (January) and 7.32 mg L^{-1} to 6.36 mg L^{-1} in the dry period (June).

During sampling, the 106 specimens from 12 species were collected. Of this total, 70% of individuals were represented by 4 species from 4 different families: *M. furnieri* (Sciaenidae), 29.2%; *G. genidens* (Ariidae), 15.1%; *D. rhombeus* (Gerreidae), 12.3%; and *P. punctatus* (Triglidae), 10.4%.

The condition factor is an estimation of the general well-being of fish (JONES *et al.,* 1999). It is based on the hypothesis that heavier individuals of a given length are in better condition than less weighty individuals (BAGENAL; TESCH, 1978). In general, the fish collected during the winter season showed a mean total length that ranged from 10.4±2.7 cm (*D. rhombeus*) to 24.0±1.8 cm (*H. gutattus*) and a mean total weight that ranged from 17.2±17.0 g (*D. rhombeus*) to 517.3±114. 0 g (*H. gutattus*).

Freire *et al.* (2020) observed 130 specimens of the catfish *G. genidens* that were caught in the spring months of 2013 and 2014 in three bays of the Rio de Janeiro state (including Sepetiba Bay) and recorded a mean total length of 19.1-21.4 cm, which was similar to that found in this study (13-26 cm), and a mean total weight range of 72.2-89.5 g, which was less than that reported in this study (19.3-132.3 g).

Similar to that observed by Freire *et al.* (2020), in general, the poor physiological states of the fish indicated by Fulton's Q possibly reflected the extremely poor environmental quality of Sepetiba Bay; the fish showed an average general condition factor of 1.2, with the exception of *H. guttata*, which had a factor of 3.7 (n = 3 specimens).

This worsening of the quality of the bay's environment likely increased the exposure time of the fish to the effects of suspended particulate material with metals. Moreover, dredging activities can also have adverse effects on ichthyofauna by reducing food abundance. The removal of sediments leads to the death of the benthic fauna and the increased water column turbidity, reducing primary productivity, although temporary, reduces primary productivity. In addition, discharges of domestic effluents with a range of toxic substances can have synergistic effects on aquatic fauna.

3.1 METALS IN FISH

The statistical analysis of the Kruskal-Wallis test did not show significant differences ($p>0.05$) in Al and Cd concentrations among the fish species (Figure 2a-c). In addition, significant differences ($p<0.05$) in the As, Cu, Fe, Pb and Zn distributions among the species were observed (Figure 2b, d-g).

Arsenic had the most variable distribution among the fish species, showing significant differences ($p<0.05$) for *D. rhombeus* x (*C. spixii, G. genidens* and *M. furnieri); G. genidens* x (*C. spilopterus, P. punctatus, S. vomer* and *S. tessellatus*); and *M. furnieri* x (*C. spilopterus* and *S. tessellatus*) (Figure 2b). The Cu concentration presented significant differences ($p<0.05$) among *P. punctatus* x (*D. rhombeus, M. furnieri, N. grandicassis,* and *S. vomer*), *N. grandicassis* x (*S. tessellatus*), and *S. vomer* x (*S. tessellatus*) (Figure 2d).

Four fish species showed differences in Fe concentrations: *A. lineatus* x (*H. guttatus* and *P. punctatus*) and *H. guttatus* x (*N. grandicassis*) (Figure 2e). Only *G. genidens* x (*D. rhombeus*) presented a significant difference in the Pb concentration (Figure 2f). The largest difference in Zn concentrations was found in *D. rhombeus* x (*H. guttatus, G. genidens, M. furnieri, P. punctatus* and *S. tessellatus*), followed by *N. grandicassis* x (*S. tessellatus, P. punctatus* and *H. guttatus*) and *C. spilopterus* x *(H. guttatus* and *P. punctatus*) (Figure 2g).

3.2 METALS IN SUSPENDED PARTICULATE MATTER (SPM)

Collin and Hart (2015) deduced that one of the most commonly observed behaviours by fish in response to elevated suspended sediment is the avoidance of turbid water. On the other hand, it is worth noting that not only the increase in water turbidity but also the exposure time has also produced long-term shifts in local abundance and community composition.

In the studied period, points P1 to P5 were influenced by dredging operations. Although point P6 is not directly influenced by dredging operations, it receives the discharge of the São Francisco and Guandu Rivers, which represents more than 86% of the watershed input. During 2014, 3.7 million m^3 of dredged material was removed from Sepetiba Bay and dumped offshore in a licensed disposal area 6 nautical miles outside of the bay. It is known that dredging operations increase the particulate matter in the area due to sediment resuspension.

The months of January and February showed higher mean concentrations of Al, Cu and Pb in suspended particulate matter than other sampling periods. This concentration can be the result of dredging operations in addition to the rainy season, consequently resulting in the increase of runoff and river discharge from the watershed to the bay.

Except for points P1 to P5 and P2, P3 and P5 in January and April, respectively, in the other months sampled, the Cd concentrations were below the detection limit (< 0.0002 µg g^{-1}). In the month of June, Pb and Cu at points P2 to P6 showed concentrations < 0.001 µg g^{-1} and < 0.002 µg g^{-1}, respectively (Figure 3).

Higher mean Zn concentrations were observed in February and June (Figure 3). In March and April, approximately 50% of the samples presented Zn concentrations < 0.02 µg g^{-1}.

3.3 BIOSEDIMENT ACCUMULATION FACTOR

The fish species with the highest BSAFs for particulate matter for all analysed metals was *D. rhombeus*, while those for the BSAFs in sediment were *G. genides* (for Cu, Fe, Pb) and *P. punctatus* (for Zn) (Table 2).

4 DISCUSSION

4.1 METALS IN FISH

The similarity of Al and Cd distributions among the fish species can be related to the fact that these are non-essential elements, and therefore, organisms have physiological mechanisms for non-assimilation and/or efficient excretion (WOOD *et al.,* 2012a, b). Furthermore, the differences in the metal (Fe, Zn, Cu) distributions among the species can be related to the essential nature of these elements (UTHUS 1992; WOOD *et al* 2012a, b), the specific physiological requirements of the species and food habits.

Some studies that have investigated the food habits of ichthyofauna in Sepetiba Bay have indicated that the index of relative importance for the studied species included the following organisms: Polychaeta (for *M. furnieri*, *A. lineatus*, and *T. paulistanus*), Polychaeta and Crustacea (for *G. genidens*, *P. punctatus*, and *S. tessellatus*), Copepoda, Ostracods and Polychaeta (for *C. spixii* and *D. rhombeus*), Teleostei (for *C. spilopterus*) and Isaeidae and Polychaeta (for *S. tessellatus*) (GUEDES; ARAÚJO 2008; GUEDES *et al*., 2015). Some investigations on the Brazilian coast have indicated that the main food items for species are as follows: *S. vomer* (crustaceans and fish) (HÖFLING *et al.,* 1998), *H. guttatus* (crustaceans and molluscs) (CARQUEIJA *et al*., 1995; SILVA *et al.,* 2001), and *N. grandicassis* (crustaceans) (MENDES; BARTHEM 2010).

In the present study, considering the food items reported in the literature, we observed the importance of diet in metal accumulation in fish muscle (supplementary material S1). For example, *D. rhombeus*, which consumes zooplankton (copepods), showed lower As and Pb concentrations and higher Cu and Zn concentrations than other species whose food included Polychaeta and crustaceans. According to the compilation shown in supplementary material S1, molluscs had the highest metal concentrations and are an important group in the transfer of metals to the fish community.

Furthermore, to assess the potential risk of human consumption of these fish, the metal concentrations were calculated for wet weight considering an average moisture of 80% in fish muscle (MURRAY; BURT 1983).

4.1.1 ALUMINIUM

The high Al concentrations in fish are related to gill inflammation and increased mucus production (PLAYLE *et al.* 1989; WITTERS *et al.* 1991). In addition, this metal reduces the growth rate and reproduction success (WOOD *et al* 2012b). In humans, Al accumulation in the brain has been suggested to be involved in the development of neurodegenerative disorders, amyotrophic lateral sclerosis and Alzheimer's disease (BONDY 2010). The average Al concentration found in the *M. furnieri* species in the present study (1.7 $\mu g\ g^{-1}$ w.w.) was half that found (3.8 $\mu g\ g^{-1}$ w.w.) by Carneiro *et al.* (2011) and an order of magnitude lower than that (76.1 $\mu g\ g^{-1}$ w.w.) observed by Medeiros *et al.* (2012) in fish (*M. furnieri*) purchased at the São Pedro fish market in Niterói in south-eastern Brazil. Unfortunately, in that study, the authors did not determine the origin of the fish purchased in the market. The species *Symphurus tessellatus* showed a higher mean Al concentration (20.3 $\mu g\ g^{-1}$ w.w.) in this study than the one (9.4 $\mu g.\ g^{-1}$ w.w.) conducted on the Macaé coast of south-eastern Brazil (CARVALHO *et al*., 2000).

In Sepetiba Bay, the discharge of rivers and the effluent from the water treatment Guandu station for the human water supply of Rio de Janeiro (second largest water treatment plant in the world, namely, the ETA-Guandu) were the most likely sources of Al to the bay (Professor Silva-Filho personal communication).

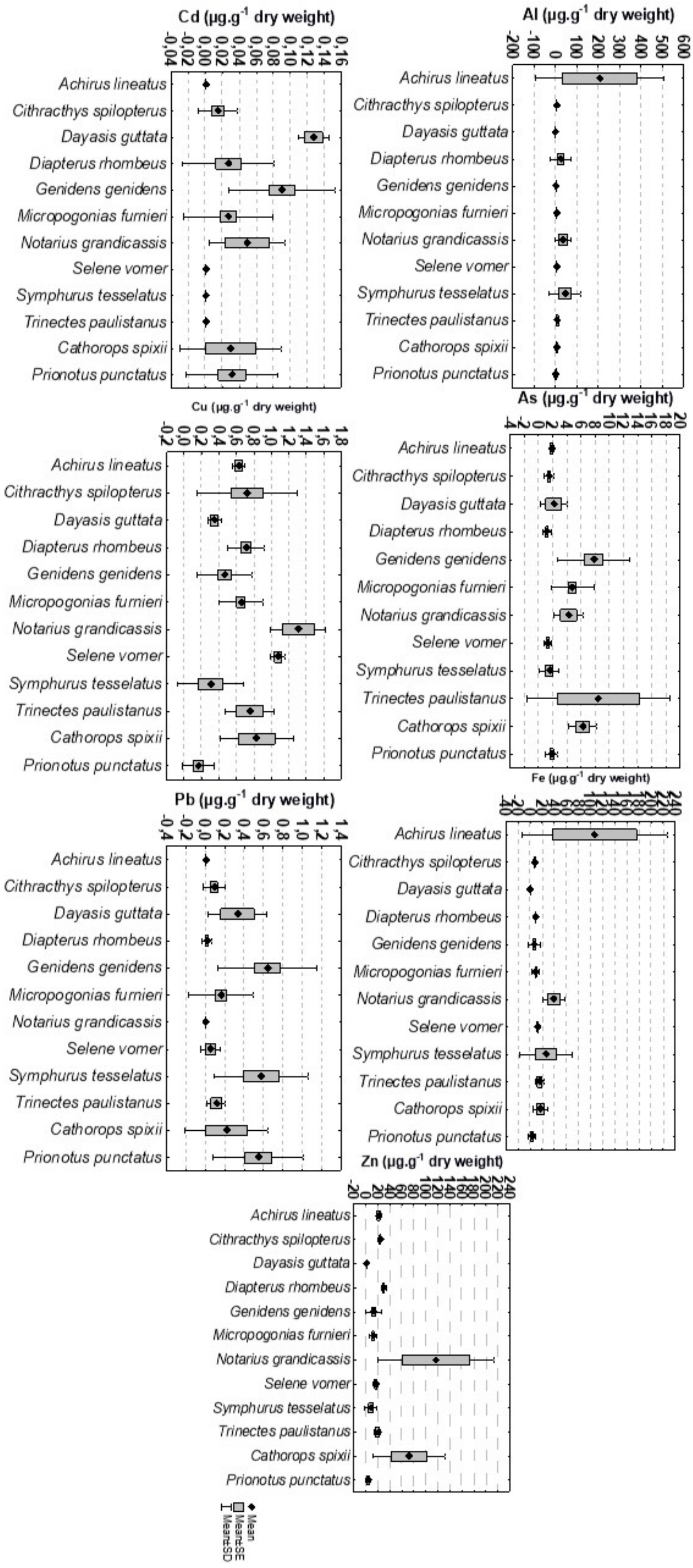

Figure 2
Metal distribution in fish species of Sepetiba Bay

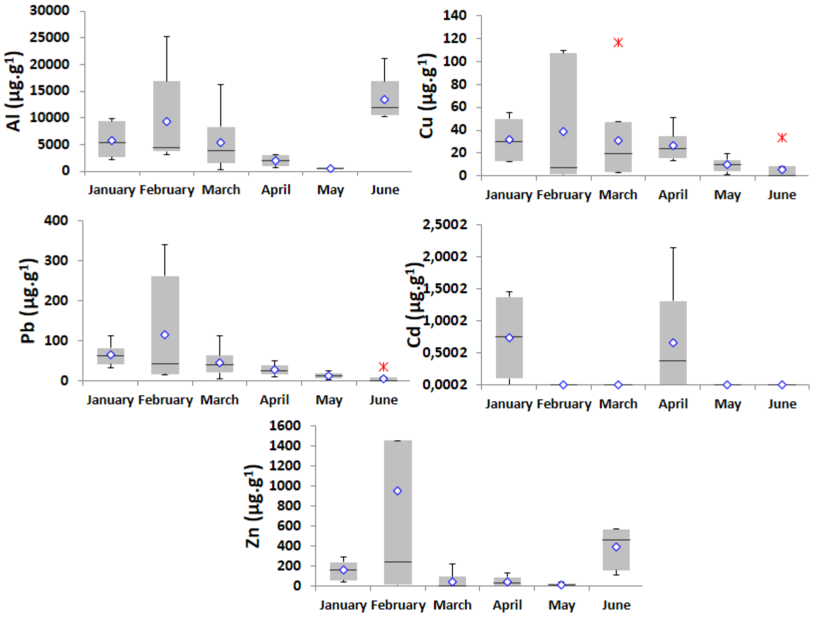

Figure 3
Metals in SPM: (diamond) mean, (-) median, and (*) outlier

Table 2 - Biosediment accumulation factors (BSAFs) in fish from Sepetiba Bay, Brazil

Metal	BSAF (sediment bioavailability)				BSAF (particulate matter)			
	G. genidens	*M. furnieri*	*D. rhombeus*	*P. punctatus*	*G. genidens*	*M. furnieri*	*D. rhombeus*	*P. punctatus*
Al	-	-	-	-	0.001	0.001	0.004	0.003
Cd	0.02	0.02	0.02	0.02	-	-	-	-
Cu	0.07	0.1	0.11	0.05	0.02	0.02	0.03	0.01
Fe	0.003	0.002	0.001	0.002	-	-	-	-
Pb	0.06	0.05	0.01	0.05	0.02	0.01	0.003	0.01
Zn	0.01	0.01	0.03	0.004	0.04	0.04	0.09	0.01

4.1.2 ARSENIC

Arsenic accumulation in fish tissue can cause a reduction in growth and fertility as well as skin lesions and developmental disorders (WOOD *et al.*, 2012b). The immunotoxic effects in fish from chronic exposure to this element have been demonstrated (DATTA *et al.*, 2009).

Higher As concentrations in the sediments were reported in Sepetiba Bay (MAGALHÃES *et al.*, 2001). The mean As concentration in tissue observed in the present study for *M. furnieri* (1.0 µg.g^{-1} w.w) was close to that reported in estuaries from South America (south-eastern Brazil 1.2 µg g^{-1} w.w. (MEDEIROS *et al.,* 2012) and Uruguay (1.2 µg g^{-1} w.w. (CORRALES *et al.,* 2016)). The *C. spixii* species from Sepetiba (1.2 µg g^{-1} w.w.) presented half of the As concentration found in this species from Paranaguá (3.4 µg g^{-1} w.w.) (south-eastern Brazil) (ANGELI *et al.,* 2013). In contrast, the *G. genides* in the present study (1.6 µg.g^{-1} w.w.) were enriched in As in comparison to those from Paranaguá (1.0 µg g^{-1} w.w.) (ANGELI *et al.,* 2013).

4.1.3 CADMIUM

Elevated concentrations of Cd in fish are associated with ion imbalances, the reduction in growth and reproduction, immunosuppression and endocrine disruption (WOOD *et al*., 2012b). Human diseases related to Cd are ionic imbalances in serum and osteoporosis (YOUNESS *et al*., 2012).

Cadmium contamination in sediment was reported for Sepetiba Bay (BARCELLOS; LACERDA 1994). According to the cited authors, Cd accumulation was related to the operation of Mercantil Ingá, which processes iron ore. The results of the present study showed a low concentration of this element. The highest concentration was approximately 0.02 µg.g^{-1} w.w. in *C. spixii*, *D. rhombeus*, *P. punctatus*, *S. tessellatus*, and *H. guttatus*.

On the Brazilian coast, the following values have been reported: 0.0004-0.07 µg g^{-1} w.w. for *M. furnieri* (KEHRIG *et al.,* 2007; MEDEIROS *et al.,* 2012; NIENCHESKI *et al.,* 2014), 0.002-0.7 µg g^{-1} w.w. for *C. spixii* (BARBIERI *et al*., 2010; AZEVEDO *et al*., 2012; ANGELI *et al*., 2013; NIENCHESKI *et al*., 2014), and 0.06 µg g^{-1} w.w. for *H. gutattus* (CARVALHO *et al*., 2000).

4.1.4 COPPER

Copper is an essential trace element for all biological organisms from bacterial cells to humans and is a key constituent of metabolic enzymes (CRAIG *et al.,* 2007, FESTA; THIELE, 2011)

Elevated Cu exposure in fish can cause olfactory inhibition, a reduction in neuron sensitivity in the lateral line, an increase in cortisol levels and catabolism of proteins, a reduction in the swimming capacity and immunosuppression (WOOD *et al*., 2012a). Excess Cu can cause hepatic diseases in humans.

The Cu concentration (0.06 µg g^{-1} w.w.) for *H. gutattus* from Sepetiba Bay was one order of magnitude lower than the coast of Macaé, RJ, Brazil (1.1 µg g^{-1} w.w.) (CARVALHO et al., 2000). The species *M. furnieri* showed Cu concentrations lower than those in other coastal regions of Brazil: Guanabara Bay, 0.6 µg g^{-1} w.w. (KEHRIG *et al*., 2007) and Patos Lagoon, 0.06-1.1 µg g^{-1} w.w. (NIENCHESKI *et al*., 2014). The species *C. spixii* and *G. genidens* from Sepetiba showed the lowest Cu accumulations of 0.2 µg g^{-1} w.w. and 0.08 µg g^{-1} w.w., respectively. In other Brazilian estuarine environments, the concentrations in these species range from 0.07-0.32 µg g^{-1} w.w. (AZEVEDO *et al.,* 2012; ANGELI *et al*., 2013; NIENCHESKI *et al*., 2014).

4.1.5 IRON

Iron concentrations vary by fish species (SHIAU; SU 2003). This characteristic was also observed in the present study. The physiological actuation of Fe in vertebrates is related to its participation in respiratory pigments, cytochrome c-oxidase, DNA synthesis and the immune system. Elevated Fe concentrations can cause alterations in the liver and kidneys as well as reductions in growth and immunosuppression.

Data from the literature have described the Fe concentrations in *M. furnieri* from Guanabara Bay at 2.1 µg g^{-1} w.w. (KEHRIG *et al.,* 2007), which is the same order of magnitude found in the present study (2.4 µg g^{-1} w.w.). Only *S. tessellatus* showed Fe concentrations that were two times higher (12.2 µg g^{-1} w.w.) in Sepetiba Bay than in Macaé, RJ (6.3 µg g^{-1} w.w.) (CARVALHO *et al*., 2000).

4.1.6 LEAD

The main source of Pb in the aquatic environment is the atmospheric deposition of particulate material from the burning of fossil fuels (RENBERG *et al*., 2000). Pb addition in fuel has been banned in Brazil since the 1990s, but this metal is still used in other activities, such as ship painting. Furthermore, local inhabitants burn domestic waste (personal observation), which is carried to tributary rivers running to the bay.

Fish exposed to Pb showed histological alterations in the liver and kidneys, reductions in growth and immunosuppression (MUÑOZ *et al*., 2015). In addition, humans exposed to Pb develop kidney disease, haematological disorders and neuronal disturbances (loss of memory and cognitive impairment) (SILBERGELD *et al.*, 2000).

Higher Pb concentrations were observed in two catfish species, *G. genidens* and *C. spixii* (Figure 2). These concentrations can be associated with the feeding behaviour of the species, which are bottom feeders consuming contaminated sediments from the bay. Higher values (11.2 µg g^{-1} w.w.) of this metal in *C. spixii* are reported in the north-eastern region of the Brazilian coast (BARBIERI *et al.,* 2010).

4.1.7 ZINC

Zinc is an essential element in fish. However, exposure to higher concentrations can produce hyperplasia and higher mucus secretion in gills. Zn participated in the metabolism of proteins, nucleic acids, carbohydrates and lipids. Zn also acts in the immunologic system and neurotransmission (WOOD *et al*., 2012, a). In humans, Zn acts as an enzyme cofactor. Furthermore, higher concentrations can alter Cu and Fe metabolism, reduce high-density protein in serum and depress the immune system.

High concentrations of Zn (9.7 µg g^{-1}) were reported in *M. furnieri* from Sepetiba Bay (CARNEIRO *et al.,* 2011) that were approximately 3 times higher than the present study (2.4 µg g^{-1} w.w.). This species presents a large variation in Zn along the Brazilian coast (0.4-8.1 µg g^{-1} w.w. in Patos Lagoon (NIENSCHESKI *et al.,* 2014) and 3.2 µg g^{-1} w.w. in Guanabara Bay (KEHRIG *et al.,* 2007)). The same variation was found in *C. spixii* (4.3-15.6 µg g^{-1} w.w. (BARBIERI *et al.,* 2010; AZEVEDO *et al.,* 2012; ANGELI *et al.,* 2013; NIENSCHESKI *et al.,* 2014). In Cananéia in south-eastern Brazil, there are higher Zn concentrations in the muscle of *C. spixii* from the pristine area (1.5 times higher than the polluted area) (AZEVEDO *et al*., 2012).

4.2 HUMAN HEALTH RISK ASSESSMENT

This study indicated that Cd and Pb, which are non-essential metals, were below the permissible limit suggested by the European Commission and ANVISA (Table 3).

For As only, ANVISA established (1 µg g^{-1} w.w.) the maximum values for human consumption. The concentrations in *G. genidens* (1.6±1.0 µg g^{-1} w.w), *C. spixii* (1.2±0.4 µg g^{-1} w.w) and *T. paulistanus* (1.7±2.0 µg g^{-1} w.w) found in Sepetiba Bay exceeded this limit. This metal presents a variety of chemical forms, and arsenobetaine is the most abundant in fish tissue (KIRBY; MAHER 2002; VILLA-LOJO *et al.,* 2002). Arsenobetaine has been identified as a major water-soluble As compound in the tissues of marine organisms (EDMONDS; FRANCESCONI, 1993).

Other studies have reported As concentrations in the tissues of *C. spixii*, *G. genidens* and *M. furnieri* from the Brazilian coast that were above the human consumption limit established by the ANVISA (MEDEIROS *et al*., 2012; ANGELI *et al* 2013). According to Mirlean *et al.* (2011, 2012), sediments from the south-eastern Brazilian coast are naturally enriched in As due to detritus from geniculate calcareous algae and iron oxyhydroxides, which are rich in As. Moreover, Sepetiba Bay possesses historical anthropogenic As contamination (MAGALHÃES *et al.,* 2001).

Table 4 shows the HQ for all species investigated. As reported by Horta et al. (2011), the fishing population, due to the consumption of a greater amount of fish, has a higher risk of metal intake. Compared to the control population, the risk of metal intake in the fishing population increased by 56 times for inorganic As, Cu and Zn, 62 times for Cd and 54 times for Pb.

Although the fishing population presented a significant difference in the metal exposure risk compared to the control, the results found in the present study indicated a low risk of metal contamination from fish intake from Sepetiba Bay in both populations investigated. The hazard index was in the range of $3x10^{-4}$ to $4x10^{-5}$ for the control population and $1x10^{-2}$ to $2x10^{-3}$ for the fishing population. *T. paulistanus* was the species with the highest hazard index, while *S. vomer* showed the lowest hazard index (Table 4).

Table 3 - Maximum metal concentrations for human consumption.

Legislation	$\mu g\ g^{-1}$ wet weight		
	As	Cd	Pb
ANVISA (2013)	1.0	0.05 0.1 (Bonito, Carapeta, enguia, tainha, jurel, imperador, cavala, sardinha, atum, linguado) 0.2 (Melva) 0.3 (Anchova, Espada)	0.3
FAO (2011)	-	2.0	0.3
European Commission (2006, 2008)	-	0.05 0.1 (*Sarda sarda*, *Diplodus vulgaris*, *Anguilla anguilla*, *Mugil labrosus labrosus*, Trachurus species, *Luvarus imperialis*, Scomber species; *Sardina pilchardus*, Sardinops species, Thunnus species, Euthynnus species, *Katsuwonus pelamis*, *Dicologoglossa cuneata*) 0.2 (Auxis species) 0.3 (Engraulis species; *Xiphias gladius*)	0.3

4.3 METALS IN SUSPENDED PARTICULATE MATTER

In the present study, the concentrations of Cd, Cu, Pb and Zn in suspended particulate matter (SPM) from Sepetiba Bay were lower than those found in previous reports (LACERDA *et al.,* 1987; FRANZ 2004) (Table 5). This reduction in metals in SPM is related to diverse initiatives that have occurred since 2008 to clean up the bay once almost all the superficial contaminated sediment from the northern region of the bay was dredged and kept in subaquatic confined disposal facilities in the bottom of the bay.

4.4 BIOSEDIMENT ACCUMULATION FACTOR

The BSAF of the bioavailable fraction of metals in the sediment followed the decreasing sequence Cu>Pb> Cd>Zn>Fe, while the decreasing sequence of the BSAF for the particulate matter was Zn>Cu>Pb>Al. Meanwhile, in fish muscle, the order of accumulation was Zn>Fe>Al>As>Cu>Cd.

The metal concentrations in sediments (bioavailable fraction) and SPM were higher than that in fish muscle, indicating a low transference between the environmental compartments to muscle tissue in fish. However, the As in fish muscle showed concentrations superior to that recommended for human consumption, indicating that chronic contamination was misrepresented by the sediment and SPM concentrations. For this reason, food items would have a greater influence on the concentration of metals (which varies between species) in the studied organisms.

By applying the data of the present study (Table 1) and the metal concentrations in shrimp (*Litopenaeus schmitti*) sampled from 2011-2012 (NASCIMENTO *et al.,* 2016), we calculated the BSAFs. The BSAF results for the sediment bioavailable fraction were 0.04 for Zn and Cd, 3.1 for Cu and 0.003 for Pb. Meanwhile, the BSAFs in the particulate fraction were 0.1 for Zn, 0.7 for Cu and 0.001 for Pb. These results showed that the BSAFs for shrimp were one order of magnitude higher than those in the fish in the present study.

Wanick *et al.* (2013) also found BSAF values (58.2 for Zn; 1.5 for Cd and 5.7 for Cu) that were hundreds of times higher in the digestive gland of the oyster *Crassostrea rhizophorae* from Sepetiba Bay when compared to the values found in fish and shrimp.

The lower BSAF values found in the present study illustrated the capacity for metal homeostasis in fish, although studies have indicated that > 50% of metal is weakly bound to sediments (RODRIGUES *et al.*, 2017). Moreover, the target organs in the metal detoxification process in fish are the liver, gills and kidney. In the present study, muscle was analysed, which can indicate chronic exposure.

Table 4- Hazard quotients for metal intake via fish consumption. As(2) – 2% inorganic; As(30) – 30% inorganic; ∑(HQ) – Hazard Index

Species	Brazilian Population (intake 27 g fish/day)							Fishers (intake 200 g fish/day)						
	As(2)	As(30)	Cd	Cu	Pb	Zn	**∑(HQ)**	As(2)	As(30)	Cd	Cu	Pb	Zn	**∑(HQ)**
T. paulistanus	$2.E^{-05}$	$2.E^{-04}$		$5.E^{-07}$	$2.E^{-06}$	$2.E^{-06}$	**$3.E^{-04}$**	$9.E^{-04}$	$1.E^{-02}$		$3.E^{-05}$	$1.E^{-04}$	$1.E^{-04}$	**$1.E^{-02}$**
G. genidens	$2.E^{-05}$	$2.E^{-04}$	$3.E^{-06}$	$3.E^{-07}$	$9.E^{-06}$	$1.E^{-06}$	**$3.E^{-04}$**	$9.E^{-04}$	$1.E^{-02}$	$2.E^{-04}$	$2.E^{-05}$	$5.E^{-04}$	$7.E^{-05}$	**$1.E^{-02}$**
C. spixii	$1.E^{-05}$	$2.E^{-04}$	$3.E^{-06}$	$6.E^{-07}$	$9.E^{-06}$	$7.E^{-06}$	**$2.E^{-04}$**	$7.E^{-04}$	$1.E^{-02}$	$2.E^{-04}$	$3.E^{-05}$	$5.E^{-04}$	$4.E^{-04}$	**$1.E^{-02}$**
M. furnieri	$9.E^{-06}$	$1.E^{-04}$	$3.E^{-07}$	$4.E^{-07}$	$7.E^{-06}$	$1.E^{-06}$	**$2.E^{-04}$**	$5.E^{-04}$	$8.E^{-03}$	$2.E^{-05}$	$2.E^{-05}$	$4.E^{-04}$	$6.E^{-05}$	**$9.E^{-03}$**
N. grandicassis	$8.E^{-06}$	$1.E^{-04}$	$1.E^{-06}$	$9.E^{-07}$		$1.E^{-05}$	**$1.E^{-04}$**	$4.E^{-04}$	$7.E^{-03}$	$7.E^{-05}$	$5.E^{-05}$		$6.E^{-04}$	**$8.E^{-03}$**
H. gutattus	$6.E^{-06}$	$1.E^{-04}$	$3.E^{-06}$	$2.E^{-07}$	$5.E^{-06}$	$8.E^{-08}$	**$1.E^{-04}$**	$4.E^{-04}$	$5.E^{-03}$	$2.E^{-04}$	$1.E^{-05}$	$3.E^{-04}$	$4.E^{-06}$	**$6.E^{-03}$**
S. tessellatus	$5.E^{-06}$	$7.E^{-05}$	$3.E^{-06}$	$3.E^{-07}$	$7.E^{-06}$	$8.E^{-07}$	**$9.E^{-05}$**	$3.E^{-04}$	$4.E^{-03}$	$2.E^{-04}$	$2.E^{-05}$	$4.E^{-04}$	$4.E^{-05}$	**$5.E^{-03}$**
P. puntactus	$5.E^{-06}$	$7.E^{-05}$	$3.E^{-06}$	$2.E^{-07}$	$7.E^{-06}$	$3.E^{-07}$	**$8.E^{-05}$**	$3.E^{-04}$	$4.E^{-03}$	$2.E^{-04}$	$1.E^{-05}$	$4.E^{-04}$	$2.E^{-05}$	**$5.E^{-03}$**
A. lineatus	$4.E^{-06}$	$6.E^{-05}$		$4.E^{-07}$		$2.E^{-06}$	**$6.E^{-05}$**	$2.E^{-04}$	$3.E^{-03}$		$2.E^{-05}$		$1.E^{-04}$	**$3.E^{-03}$**
C. spilopterus	$3.E^{-06}$	$5.E^{-05}$	$9.E^{-07}$	$6.E^{-07}$	$2.E^{-06}$	$2.E^{-06}$	**$6.E^{-05}$**	$2.E^{-04}$	$3.E^{-03}$	$5.E^{-05}$	$3.E^{-05}$	$1.E^{-04}$	$1.E^{-04}$	**$3.E^{-03}$**
D. rhombeus	$2.E^{-06}$	$3.E^{-05}$	$3.E^{-06}$	$5.E^{-07}$	$2.E^{-06}$	$3.E^{-06}$	**$5.E^{-05}$**	$1.E^{-04}$	$2.E^{-03}$	$1.E^{-04}$	$3.E^{-05}$	$1.E^{-04}$	$2.E^{-04}$	**$3.E^{-03}$**
S. vomer	$2.E^{-06}$	$3.E^{-05}$		$7.E^{-07}$	$2.E^{-06}$	$2.E^{-06}$	**$4.E^{-05}$**	$1.E^{-04}$	$2.E^{-03}$		$4.E^{-05}$	$1.E^{-04}$	$9.E^{-05}$	**$2.E^{-03}$**

Table 5 - Historical metal (µg g^{-1}) concentrations in the SPM of Sepetiba Bay.

	Mean (min–max) dry weight			
Author	**Cd**	**Cu**	**Pb**	**Zn**
LACERDA *et al.* 1987	3.2	61.6	139.0	390.0
LACERDA *et al.* 1988	3.2	85.0	68.2	478.0
FRANZ 2004^{s}	4.1	-	52.7	752.7
FRANZ 2004^{w}	3.2	-	39.7	749.9
Present work	<0.002-1.3	28.4 (<0.002-116.1)	51.8 (<0.001-339.5)	332.4 (<0.02-4887.4)

^{s} Summer; ^{w} Winter

5 CONCLUSIONS

The BSAFs from the bioavailable sediment fractions and suspended particulate matter showed lower metal transference to fish muscle. Considering this result, we can hypothesize that the most important pathway for metal contamination in fish in the bay is via the food web. The concentrations of As observed in the species *C. spixii*, *G. genidens* and *T. paulistanus* were above those allowed for human consumption by Brazilian legislation. However, the estimated probability and risk of metal intake via fish consumption showed that the consumption of 0.2 Kg day^{-1} of all species presented low risk. Due to the high toxicity of As, future studies are necessary to investigate the chemical speciation of this element in the environmental compartments and biota of Sepetiba Bay to determine the source (whether natural or anthropogenic) of this metal. Moreover, other studies are needed to investigate the metal contents in larval and juvenile fish in different tissues to understand the transfer of metals in the ichthyofauna of Sepetiba Bay.

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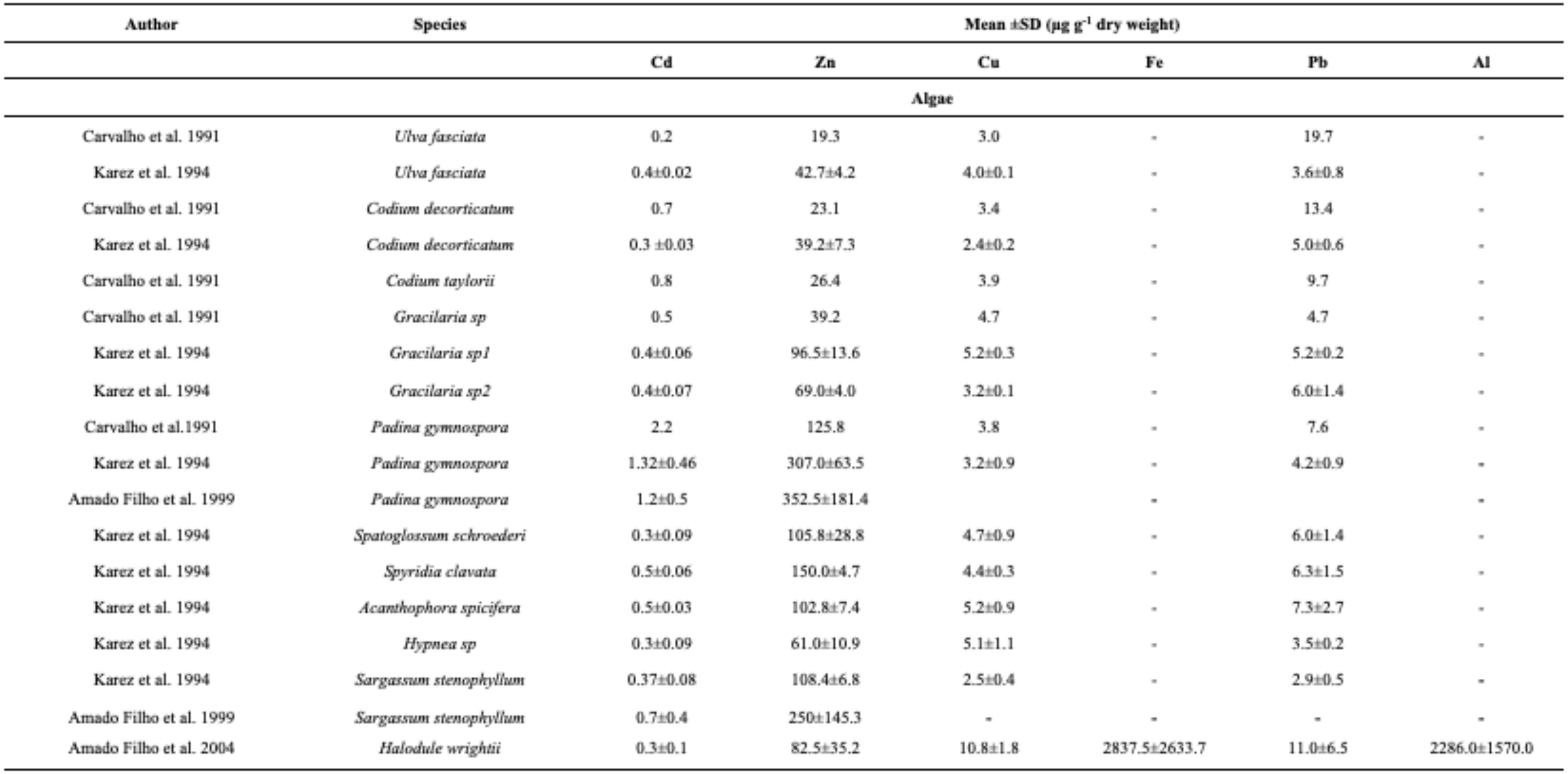
Table S1 - Compilation of metals in food items of fish from Sepetiba Bay



Author	Species	Mean ±SD (µg g^{-1} dry weight)					
		Cd	Zn	Cu	Fe	Pb	Al
				Algae			
Carvalho et al. 1991	*Ulva fasciata*	0.2	19.3	3.0	-	19.7	-
Karez et al. 1994	*Ulva fasciata*	0.4±0.02	42.7±4.2	4.0±0.1	-	3.6±0.8	-
Carvalho et al. 1991	*Codium decorticatum*	0.7	23.1	3.4	-	13.4	-
Karez et al. 1994	*Codium decorticatum*	0.3 ±0.03	39.2±7.3	2.4±0.2	-	5.0±0.6	-
Carvalho et al. 1991	*Codium taylorii*	0.8	26.4	3.9	-	9.7	-
Carvalho et al. 1991	*Gracilaria sp*	0.5	39.2	4.7	-	4.7	-
Karez et al. 1994	*Gracilaria sp1*	0.4±0.06	96.5±13.6	5.2±0.3	-	5.2±0.2	-
Karez et al. 1994	*Gracilaria sp2*	0.4±0.07	69.0±4.0	3.2±0.1	-	6.0±1.4	-
Carvalho et al.1991	*Padina gymnospora*	2.2	125.8	3.8	-	7.6	-
Karez et al. 1994	*Padina gymnospora*	1.32±0.46	307.0±63.5	3.2±0.9	-	4.2±0.9	-
Amado Filho et al. 1999	*Padina gymnospora*	1.2±0.5	352.5±181.4		-		-
Karez et al. 1994	*Spatoglossum schroederi*	0.3±0.09	105.8±28.8	4.7±0.9	-	6.0±1.4	-
Karez et al. 1994	*Spyridia clavata*	0.5±0.06	150.0±4.7	4.4±0.3	-	6.3±1.5	-
Karez et al. 1994	*Acanthophora spicifera*	0.5±0.03	102.8±7.4	5.2±0.9	-	7.3±2.7	-
Karez et al. 1994	*Hypnea sp*	0.3±0.09	61.0±10.9	5.1±1.1	-	3.5±0.2	-
Karez et al. 1994	*Sargassum stenophyllum*	0.37±0.08	108.4±6.8	2.5±0.4	-	2.9±0.5	-
Amado Filho et al. 1999	*Sargassum stenophyllum*	0.7±0.4	250±145.3	-	-	-	-
Amado Filho et al. 2004	*Halodule wrightii*	0.3±0.1	82.5±35.2	10.8±1.8	2837.5±2633.7	11.0±6.5	2286.0±1570.0

Table S1 (continuation) - Compilation of metals in food items of fish from Sepetiba Bay

Author	Species	Mean ±SD (µg g^{-1} dry weight)					
		Cd	Zn	Cu	Fe	Pb	Al
				Crustacea			
Carvalho et al. 1991	*Balanus sp.*	6.4	5151.7	5.8	-	9.8	-
Carvalho et al. 1991	*Callinectes danae*	<DL	94.2	59.1	-	<DL	-
Corrêa Junior et al. 2000	*Ucides cordatus (hepatopancreas)*	-	181.0±16.0	-	-	-	-
Carvalho et al. 1991	*Megabalanus sp.*	35.9	15515.0	16.3	-	<DL	-
Carvalho et al. 1991	*Litopenaeus schimitti*	0.3	79.2	72.2	-	11.8	-
Nascimento et al. 2016	*Litopenaeus schimitti*	-	34.5±3.0	23.5±9.6	-	-	-
				Mollusc			
Carvalho et al. 1991	*Thais haemasiana*	11.4	2508.0	48.8		7.0	
Carvalho et al. 1991	*Perna perna*	1.0	205.3	6.5		<DL	
Francioni et al 2004	*Perna Perna*	60.0*	40100.0*	1200.0*	-	-	-
Carvalho et al. 1991	*Litorina sp.*	11.5	4373.6	83.6		6.5	
Carvalho et al. 1991	*Tegula viridula*	1.4	372.3	54.7		6.8	
Carvalho et al. 1991	*Anomalocardia brasiliana*	2.6	91.2	4.5		1.4	
Carvalho et al. 1991	*Crassostrea brasiliana*	8.5	95000.0	24.5		13.4	
Carneiro et al. 2011	*Crassostrea brasiliana*	-	3199.0	17.5	219.0	-	-
Lima et al. 1986	*Crassostrea rhizophorae*	8.6	8073.0	-	-	-	-
Carvalho et al. 1991	*Crassostrea rhizophorae*	6.9	2244.0	-	-	-	-
Rebelo et al. 2003b	*Crassostrea rhizophorae*	1.7	11984.0	-	-	-	-
Rebelo et al. 2003a	*Crassostrea rhizophorae*	2.9	12205.7	-	-	-	-
Amaral et al. 2005	*Crassotrea rhizophorae*	1.1	9770.0	-	-	-	-
				Echinoderm			
Carvalho et al. 1991	*Equinaster brasiliensis*	3.0	132.8	26.2	-	6.5	-

*Wet weight

Table S2 - Metal concentrations (µg.g^{-1} dry weight) in fish from Sepetiba Bay

Species (n) Length (cm)	Al		As		Cd		Cu		Fe		Pb		Zn	
	Mean ± SD	Min–Max	Mean ± SD	Min - Max	Mean ± SD	Min–Max	Mean ± SD	Min - Max	Mean ± SD	Min - Max	Mean± SD	Min - Max	Mean ± SD	Min - Max
Micropogonias furnieri (31) *6.5 to 26.5*	8.4 ±5.4	0.02-17.4	4.8 ± 3.0	1.8-14.3	0.01 ± 0.03	0.0002-0.2	0.6 ± 0.3	0.20-1.1	12.2 ± 2.8	0.02-18.7	0.7 ± 0.3	0.001-1.2	11.9 ± 6.7	0.7-19.4
Genidens genidens (16) *12.5 to 26.0*	6.2 ± 3.8	0.02-11.6	7.8 ± 5.1	3.3-20.1	0.1 ± 0.04	0.0002-0.02	0.4± 0.3	0.17-1.0	19.2 ± 4.1	0.02-25.3	0.9 ± 0.4	0.001-1.3	12.6 ± 13.4	2.8-35.7
Cathorops spixii (4) *19*	3.6 ± 4.1	0.02-8.2	6.2 ± 2.0	3.9-8.0	0.1	0.0002-0.1	0.8 ± 0.4	0.2-1.2	21.0 ± 3.4	0.02-23.4	0.9	0.001-0.9	95.2 ± 46.0	0.9-140.7
Notarius grandicassis (3) *12.5 to 18.0*	35.8 ± 34.6	3.0-69.0	4.1 ± 2.1	2.1-6.2	0.04 ± 0.01	0.0002-0.05	1.2 ± 0.3	1.0-1.6	36.4 ± 17.6	20.7-55.4	<0.001	<0.001	116.3 ± 97.0	54.9-227.3
Diapterus rhombeus (13) *7.5 to 16.0*	23.9 ± 50.6	3.0-188.9	1.2 ± 0.6	0.0002-2.5	0.09 ± 0.07	0.0002-0.2	0.7 ± 0.2	0.5-1.3	9.1 ± 1.6	6.6-12.7	0.2	0.001-0.2	29.9 ± 4.1	24.5-38.5
Selene vomer (3) *17.0 to 18.0*	3.5 ± 1.9	2.1-5.6	1.2 ± 0.5	0.8-1.7	<0.0002	<0.0002	1.0 ± 0.05	1.0-1.1	11.7 ± 1.4	10.5-13.3	0.2	0.001-0.2	15.6 ± 2.6	13.1-18.3
Prionotus punctatus (11) *6.5 to 23.5*	15.6 ± 11.2	0.02-23.6	2.4 ± 1.0	0.5-3.8	0.1 ± 0.02	0.0002-0.2	0.3 ± 0.1	0.2-0.5	14.4 ± 5.1	0.02-18.0	0.7 ± 0.2	0.001-1.0	3.2 ± 4.4	0.8-12.8
Cithracthys spilopterus (8) *12.0 to 18.0*	4.6 ± 3.1	0.4-9.5	1.7 ± 0.5	0.002-2.3	0.03 ± 0.003	0.0002-0.03	0.8 ± 0.6	0.5-2.3	7.6 ± 2.2	4.7-12.2	0.2 ± 0.1	0.001-0.3	23.9 ± 3.4	18.7-28.9
Achirus lineatus (3) *10.0 to 12.0*	208.0 ± 301.5	23.0-555.9	1.9 ± 0.3	1.6-2.1	<0.0002	<0.0002	0.6 ± 0.07	0.6-0.7	106.5 ± 120.6	23.5-244.9	<0.001	<0.001	21.3 ± 4.2	16.6-24.9
Trinectes paulistanus (3) *10.0 to 15.0*	9.1 ± 7.1	4.4-17.2	8.4 ± 10.1	1.8-20.0	<0.0002	<0.0002	0.7 ± 0.3	0.5-1.0	15.7 ± 7.3	10.2-24.0	0.2 ± 0.02	0.001-0.2	19.4 ± 5.1	14.1-24. 4
Symphurus tessellatus (7) *9.5 to 18.5*	101.5 ± 91.8	0.02-187.2	2.50 ± 1.50	0.002-5.4	0.1 ± 0.03	0.0002-0.1	0.4 ± 0.3	0.2-0.9	60.9 ± 50.7	0.02-106.20	0.7 ± 0.3	0.001-0.8	8.2 ± 10.3	0.5-26.3
Dayasis guttata (3) *22.5 to 26.0*	<0.02	<0.02	3.3 ± 0.7	0.002-3.8	0.1 ± 0.02	0.11-0.15	0.3 ± 0.08	0.3-0.4	<DL	<DL	0.5 ± 0.1	0.001-0.6	0.8 ± 0.06	0.8-0.9