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COMBINED EFFECTS OF ENVIRONMENTAL STRESSORS IN THE AQUATIC ENVIRONMENT

Mercury (Hg) in fish consumed by the local population of the Jaguaribe River lower basin, Northeast Brazil

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Abstract The knowledge of Hg concentrations in fish is of considerable interest since these organisms are a major source of protein to coastal human populations and fishing communities. The main source of human exposure to Hg contamination occurs through the consumption of fish. In this paper, we compare Hg concentration in 13 fish species from Jaguaribe River lower basin and an adjacent coastal region in the northeastern coast of Brazil. We sampled fish from three stretches of the river: fluvial, estuarine, and marine regions. We tested the hypothesis that Hg concentration in muscle tissue vary according to species, location, and trophic level. Significant differences were observed among species and trophic level, but these could not be observed among the regions studied. As expected, the highest concentrations were observed in carnivorous fish (5.6–107.5; 26.9±18.8 ng g^{-1}). Hg concentrations observed in this study are similar to those observed in regions of low environmental contamination. We estimated Hg intake to vary between 0.02 and 0.22 ng Hg kg body $weight^{-1}$ $week^{-1}$, for the average body weight of 56.7 kg, which was considered as low exposure and therefore, a low risk to consumers of fish from the regions studied.

Keywords Fish · Jaguaribe River · Mercury · Human exposure

Introduction

Fish is the main source of animal protein for many human populations, especially for fishing villages and/or riverside communities, which, due to their traditions and food availability, present high levels of fish consumption. For its organoleptic properties, fish becomes a functional food of high nutritional value, promoting the reduction of cholesterol and heart problems and diabetes, and assists in the formation of bone structure. However, when prevenient from contaminated areas, fish ceases to be a beneficial food, and may become harmful to health (Health Canada 2007). Heavy metals, especially Hg, are among the possible sources of fish contamination which, due to its high toxicity and its kinetics in living organisms, can accumulate in organisms as well as along the trophic chain (Burguer 2008).

Hg emission to water bodies can originate from diffuse sources such as atmospheric deposition and leaching from soils, as well as from point sources, mostly from industrial, urban, and mining effluents, as well as from inadequate disposal of hospital and electronics wastes (UNEP 2013). The Jaguaribe River lower basin has witnessed a rapid increase in urbanization, agriculture, and intensive shrimp aquaculture; all these activities are potential sources of Hg to the watershed. Studies in the region have pointed out the presence of Hg in different environmental compartments including, bottom and suspended sediments, water, and biota (Vaisman et al. 2005; Costa et al. 2013; Lacerda et al. 2013). Estimates revealed annual anthropogenic emission to the lower basin of about 325 kg of Hg; being 75 kg from waste waters, 150 kg from solid waste disposal, and 0.35 kg from intensive shrimp aquaculture (Lacerda et al. 2011). Although the contribution from aquaculture is relatively small, it should be noted that this is the only activity that releases its effluents directly into the river. Costa et al. (2013) showed that 78 % of the Hg present in effluents from shrimp farming occurs as dissolved species, which are more easily assimilated by the biota. Once incorporated, Hg concentrates in organisms at the top of the food chain, which makes these organisms a potential risk to human consumers (Bastos et al. 2006).

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Because it is a major source of human exposure to Hg (Harris et al. 2003), fish has been the target of repeated investigations, being more frequent in regions where the incidences of mining and industrial activities occur. However, because of its high residence time in the environment and its ubiquitous presence in many effluents, it is necessary to monitor Hg concentrations even in regions not impacted by industrial or mining activities, such as the Jaguaribe Basin in northeastern Brazil, since the damage caused by Hg is mainly related to chronic exposure.

This study aims to compare the concentration of Hg in 13 fish species in the Jaguaribe River lower basin and adjacent coastal region, and estimate Hg exposure of local fishing/riverside communities, characterized by relative high fish frequency in their diet.

Materials and methods

Along the basin, we visited six localities (Fig, 1) where 297 local villagers from 115 households were sampled to whom we have applied structured questionnaires on their food habits and anthropomorphic data (age, sex, and weight) and acquired fish samples used in their diet. Field campaigns were carried on in 24–28 December, 2010; 25–29 March, 2011; and 22–26 October, 2011. The specimens used in this study were purchased from local fishermen on the same day of their capture. Sampling localities were grouped in marine sites, defined where salinity is always higher than 30; estuarine, where salinity rages form near 0 to 30; and fluvial, where fresh water is always present. The hydrological and hydrochemistry of these sectors can be viewed in detail in (Dias et al. 2009).

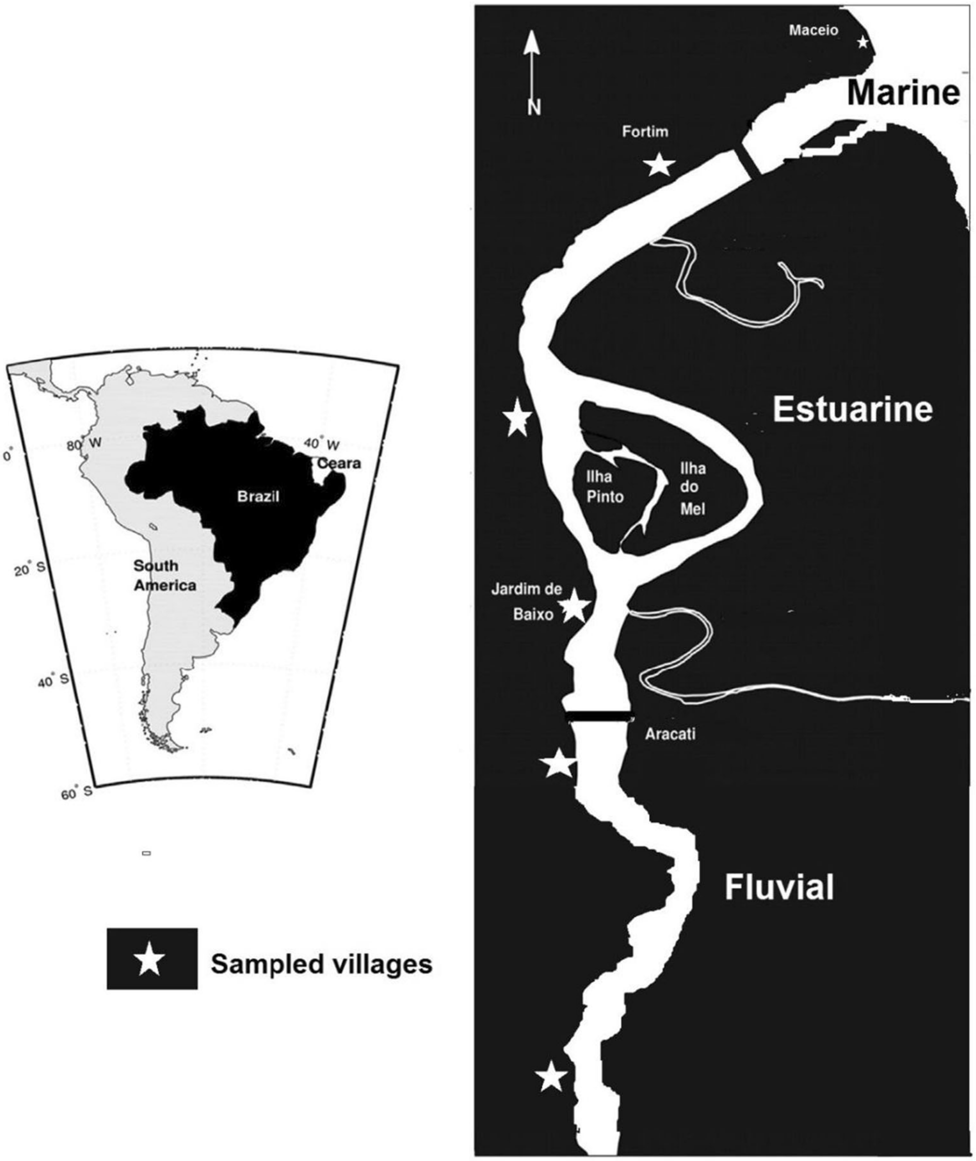

Fig. 1 Location of the sampled villages along the lower basin of the Jaguaribe River in NE Brazil

Table 1 Location, feeding habit, fish species, and number of individuals analyzed (*n*) and Hg concentrations (minimum, maximum and mean± standard deviation). Brazilian legal limits for human consumption: non-carnivorous fish=500 ng g^{-1}; carnivorous fish=1.000 ng g^{-1} (ANVISA, Agência Nacional de Vigilância Sanitária. Portaria no 685, de 27 de agosto de 1998)

Location	Feeding habit	Species	Number	Length (cm)	Weight (g)	[Hg] ng g^{-1}
River	Detritivore	*Prochilodus argenteus*	9	19.0–31.0 23.2±4.5	128.0–578.4 264.6±160.2	1.0–8.4 4.0±2.4
River	Omnivorous	*Leporinus friderici*	4	18.5–23.0 20.9±1.9	140.0–180.0 160.0±22.0	12.5–49.5 26.1±16.1
River	Carnivorous	*Serrasalmus rhombeus*	10	13.3–21.0 17.4±2.5	40.0–204.0 130.5±57.4	7.6–68.5 40.9±24.8
River	Omnivorous	*Oreochromis niloticus*	8	15–23.5 20.2±2.5	94.0–300.0 223.4±69.2	0.1–8.6 5.0±2.7
River	Carnivorous	*Cichla* sp.	10	21.5–29.0 25.3±2.6	130–362 242.6±88.1	6.7–59.5 24.8±15.5
Estuary	Carnivorous	*Cathorops spixii*	11	10.8–50.0 27.1±12.5	12.0–1774.0 383.3±531.6	10.10–80.9 32.2±24.9
Estuary	Carnivorous	*Eugerres brasilianus*	27	11.4–27.5 19.6±5.3	22.0–322.0 125.9±88.7	5.7–107.5 21.3±19.2
Estuary	Carnivorous	*Rhomboplites aurorubens*	6	14.0–19.6 16.5±2.2	40.0–130.0 78.67±34.4	21.0–37.9 26.6±6.3
Estuary	Carnivorous	*Plagioscion squamosissimus*	4	22.5–29.0 25.2±2.9	88.0–204.0 163.5±52.9	10.77–49.5 29.6±18.0
Estuary	Carnivorous	*Centropomus paralelus*	11	23.0–47.0 31.1±7.0	140.0–724.0 285.3±159.2	9.8–43.7 24.8±8.9
Estuary	Omnivorous	*Archosargus rhomboidalis*	4	24.0–32.0 27.2±3.4	260.0–550.0 352.5±133.0	20.85–86.7 49.2±27.4
Marine	Carnivorous	*Haemulon plumierii*	6	19.0–23.0 20.2±1.5	100.0–175.0 133.3±25.8	14.12–50.5 24.9±13.7
Marine	Detritivore	*Holocentrus adscensionis*	6	24.0–26.5 24.8±1.4	100.0–175.0 133.3±38.2	28.5–37.1 31.9±4.6

After acquiring the specimens, they were kept frozen under ice and stored in coolers to transport to the Coastal Biogeochemistry Laboratory of the Federal University of Ceará, where the specimens were identified and their morphometric data were obtained. Subsequently, necropsy was performed to obtain samples of muscles. Two muscle samples from each individual were cut from the central part of the body and stored in sealed plastic bags and frozen (−20 °C) till analysis. Average values from these duplicate samples were used for statistical analysis.

Prior to analysis, muscle tissues were lyophilized, and subsamples of 0.5 g of lyophilized tissue were digested in 10 ml of concentrated nitric acid (HNO_3) using heating and temperature ramp determined by Pluss Mars CEM microwave equipment used (Bezerra et al. 2013). After digestion, 1 ml of hydrogen peroxide (H_2O_2) was added to the sample in order to avoid re-complexion of Hg with particulate material (Adair and Cobb 1999). All samples were taken to 100-ml flasks with distilled water for subsequent quantification. Quantification of Hg was performed by cold vapor atomic absorption spectrometry with Nippon Instruments Corporation (NIC) model RA3210A. For the reduction of Hg, we used a stannous chloride solution ($SnCl_2$) prepared in 10 % sulfuric acid (20 % H_2SO_4). Simultaneously, certified standards to the biological material (NIST 2,976 Mussel Tissue) were also analyzed. Hg recovery from these standards averaged 94.9± 6.7 %.

Human exposure to Hg was estimated for the locations for which data on their weekly consumption of fish and frequency of intake were obtained through the structured questionnaires. Together with the concentration of Hg in fish, these constituted the necessary data for exposure level estimation (WHO – World Health Organization 1989). We investigated the species of fish consumed by the population, weekly consumption of fish, and corporal weight of respondents.

Mercury concentrations were expressed in wet weight to directly compare with limit values from the Brazilian legislation (ANVISA, Agência Nacional de Vigilância Sanitária.

Fig. 2 Mean, standard deviation, and error of the Hg concentration in fish from the lower basin of the Jaguaribe River, grouped by location

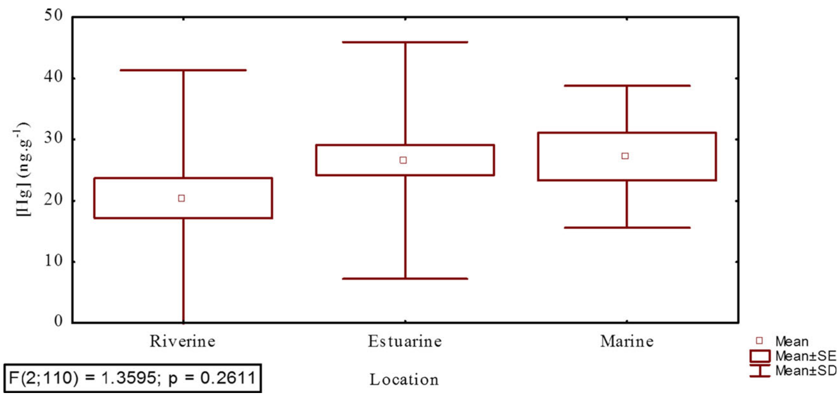

Table 2 Pearson correlation coefficients calculated between the Hg concentration in muscles and biometrics variables (weight and length) of the fish species from the Jaguaribe River low basin ($\alpha-5$ %)

Species	Number	Weight	Length
Prochilodus sp.	9	0.2048	0.2639
Leporinus friderici	4	−0.3093	−0.9551*
Serrasalmus rhombeus	10	0.8057*	0.8653*
Oreochromis niloticus	8	0.6649	0.5185
Cichla sp.	10	−0.2180	−0.1503
Cathorops spixii	11	0.1244	0.4090
Eugerres brasilianus	27	0.4786*	0.5416*
Rhomboplites aurorubens	6	−0.0984	−0.0897
Plagioscion squamosissimus	4	0.1517	−0.6564
Centropomus paralelus	11	0.0374	0.1627
Archosargus rhomboidalis	4	0.9615*	0.8138
Haemulon plumierii	6	0.0358	−0.0222
Holocentrus adscensionis	6	0.8805	0.9871*

*Significant correlation at the 5 % level

Portaria no. 685, de 27 de agosto de 1998). For statistical analysis, we used STATISTICA 7 software with a significance level of 95 %.

Results and discussion

Thirteen fish species were collected from the Jaguaribe river lower basin; five were collected in the fluvial region, six in the estuary, and two from the adjacent marine area. Most species captured along the entire sampled area were carnivorous (eight), followed by omnivorous (three), and detritivorous (two). No exclusive herbivorous species were capture in this study (Table 1). Mercury concentrations observed in fish from all sites were comparable to those reported for uncontaminated regions (Azevedo et al. 2012; Costa et al. 2009, 2012) and showed similar concentrations to those reported for the same area in previous studies (Costa et al. 2013). In relation to the legal limits regulated for human consumption (ANVISA, Agência Nacional de Vigilância Sanitária. Portaria no 685, de 27 de agosto de 1998) all concentrations were lower than the 500 ng g^{-1} for non-carnivorous fish and 1.000 ng g^{-1} for carnivorous species.

Comparison of Hg concentrations in fish showed no significant difference ($p<0.05$) between the different regions of the fluvial–marine continuous sampled (Fig. 2). There is a paucity of data on Hg concentration in this region or most of these species, but previous studies have already observed low Hg concentration in fish from the estuarine and the adjacent coastal area of the Jaguaribe lower basin (Costa et al. 2012, 2013). Vaisman et al. (2005) based on the Hg concentrations observed in local bottom estuarine sediments (average 10± 2 ng g^{-1}) and oysters (52±24 ng g^{-1}), classified the region as not impacted by Hg. On the other hand, Costa et al. (2013) quantified Hg concentrations in wastewater from aquaculture farms, highlighting the continuous input of Hg to the estuary from this activity; totaling to annual emissions of 0.35 kg of Hg (Lacerda et al. 2011).

In addition to external environmental factors, such as season (Barletta et al. 2012; Costa et al. 2009) and the concentration and availability of the metal in the environment (Harris et al. 2003); individual characteristics also have an important influence on the incorporation of Hg by organisms. The cumulative effects of Hg has been reported by several authors, including bioaccumulation (Costa et al. 2013) and food web biomagnification (Gentès et al. 2013) as well as detoxification mechanisms (Soares et al. 2011). The correlation between the concentration of Hg and morphometric data (weight and length) was only identified for some species (*Leporinus friderici*, *Serrasalmus rhombeus*, *Eugerres brasilianus*, *Archosargus rhomboidalis*, and *Holocentrus adscensionis*), as shown in Table 2, although the relative small number of samples hampers a better discussion of these relationships. Notwithstanding, although environmental and organismal variables could affect Hg content in fish, the generally very low concentrations found in the Jaguaribe basin fish will not

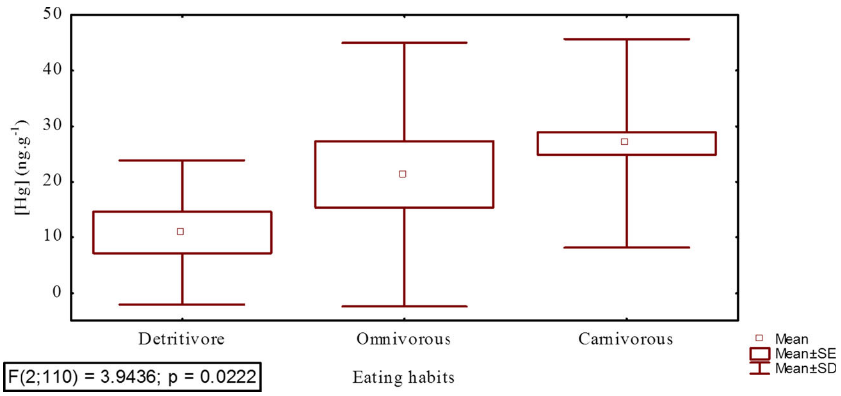

Fig. 3 Mean, standard deviation, and error of the Hg concentration in fish from the lower basin of the Jaguaribe River, grouped by eating habits

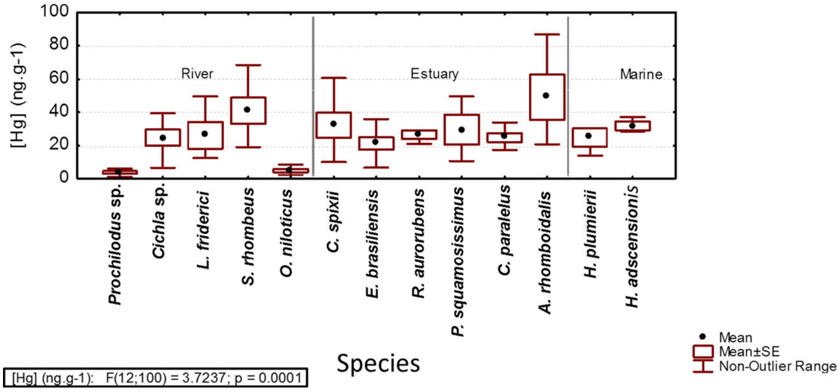

Fig. 4 Mean values and range of Hg concentrations measured in individual fish species of the lower basin of the Jaguaribe River, in the different areas of capture (α=5 %)

reach values close to the legal limits for human consumption, at least under the present magnitude of local Hg sources.

The highest average concentrations of Hg were observed in the muscles of carnivorous species, demonstrating the process of biomagnification; however, the concentration differences were not significant between carnivorous and omnivorous species (ANOVA, $p<0.05$) (Fig. 3). Figure 4 shows the mean and range of Hg concentrations measured in all fish species from the three different locations. It can be seen that the carnivorous species *S. rhombeus* shows the highest concentration among the fluvial species, whereas the lowest was found in *Prochilodus* sp. and *Oreochromis niloticus*. In turn, these are also higher when compared to the species *Cathorops spixii* ($p<0.05$; 0.023/0.048, respectively) and *A. rhomboidalis* ($p<0.05$; 0.004/0.002, respectively), both caught in the estuary. Overall, however, concentrations found in fish from the Jaguaribe River are much lower than those reported for these species in contaminated areas in Brazil (Bastos et al. 2006; Küttner et al. 2009; Passos et al. 2008).

The average consumption of fish in fishing/riverine communities was 0.253 kg week^{-1}, which are higher than the values set by WHO – World Health Organization (2007) (230 g/week) for a proper diet, and higher than those estimated for the northeastern region of Brazil (Costa et al. 2009). The average individual weight was 56.7 kg. A 150-g serving of fish provides about 50–60 % of protein requirement for adults (FAO and Food and Agriculture Organization of the United Nations 2010); however, the risks and benefits of fish consumption have been discussed by some authors (Domingo et al. 2007; Burguer and Gochfeld 2007). To compare the estimated exposure of the local population with that recommended by WHO – World Health Organization (2007) and the USEPA and Guidance for Assessing Chemical Contaminants data for use in fish Advisories (2013) we extrapolated our exposure values based on the local average body weight to an average weight of 70 kg, thus enabling comparison with other regions.

The level of exposure of the regional population to Hg resulting from the consumption of fish is shown in Table 3. Considering the maximum levels of Hg allowed for fish consumption (ANVISA, Agência Nacional de Vigilância Sanitária. Portaria no. 685, de 27 de agosto de 1998), and based on the fish consumption rate recommended by WHO (230 g week^{-1}) and body weight of 70 kg, the dose limit for the consumption of

Table 3 Weekly Hg exposure of the population in the lower basin of the Jaguaribe River based on each species of fish consumed (minimum, maximum, mean± standard deviation)

Species	Intake Hg (ng Hg kg body weight^{-1} week^{-1}) 56.7 kg	Intake Hg (ng Hg kg body weight^{-1} week^{-1}) 70.0 kg
Prochilodus sp.	0.004–0.038 (0.018±0.011)	0.004–0.030 (0.014±0.009)
Leporinus friderici	0.056–0.221 (0.117±0.072)	0.045–0.179 (0.094±0.058)
Serrasalmus rhombeus	0.0340–0.306 (0.183±0.110)	0.028–0.248 (0.148±0.089)
Oreochromis niloticus	0.001–0.038 (0.022±0.012)	0.001–0.031 (0.018±0.010)
Cichla sp.	0.030–0.265 (0.111±0.069)	0.024–0.215 (0.090±0.056)
Cathorops spixii	0.045–0.361 (0.144–0.111)	0.037–0.292 (0.116±0.090)
Eugerres brasilianus	0.025–0.480 (0.095–0.086)	0.020–0.389 (0.077±0.070)
Rhomboplites aurorubens	0.094–0.169 (0.119±0.028)	0.076–0.137 (0.096±0.023)
Plagioscion squamosissimus	0.048–0.221 (0.132±0.080)	0.039–0.179 (0.107±0.065)
Centropomus paralelus	0.044–0.195 (0.111–0.040)	0.035–0.158 (0.090±0.032)
Archosargus rhomboidalis	0.093–0.387 (0.219±0.122)	0.075–0.313 (0.178±0.099)
Haemulon plumierii	0.063–0.225 (0.111±0.061)	0.051–0.182 (0.090±0.050)
Holocentrus adscensionis	0.127–0.166 (0.142±0.021)	0.103–0.134 (0.115±0.017)

fish is of 1.6 and 3.2 μg Hg kg body weight^{-1} week^{-1}, for non-carnivorous and carnivore fish, respectively. Which are 5–6 orders of magnitude higher than those observed in the present study. Our results are estimates considering the whole intake being of a single species, which may overestimate the exposure in the case of carnivorous species, but underestimate when considering the species with lower Hg content. But since the results from the applied questionnaires showed a mixed consumption of at least six species, the range of exposure presented are probably very close to reality. The values presented here are much lower than those observed in the Amazonian region for example (Passos et al. 2008) and southeastern Brazil (Küttner et al. 2009), where much higher Hg concentrations in fish are observed. These results suggest that current levels of Hg measured in fish from the Jaguaribe lower basin are insufficient to represent any risk to the local human consumers.

Conclusion

Among the analyzed fish in the Lower Basin of the Jaguaribe River, carnivorous species presented the highest Hg concentrations, but much lower than the limits established by the National Health Security Agency of Brazil and consequently resulting in low levels of exposure doses. However, the increased emission of Hg witnessed in this basin, the local fish consumption rates being higher than most observed in other Brazilian populations and the fact that the northeastern coast of Brazil is undergoing important changes in the environment, which can affect the dynamics of Hg incorporation by aquatic organisms and Hg exposure levels for the human populations, strongly suggest the continuation of the monitoring of Hg concentrations in the local fish and the resulting exposure of the local human consumers.

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