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Mercury and selenium concentrations in fish samples from Cachoeira do PiriáMunicipality, ParáState, Brazil

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Abstract

Mercury is a major public health concern because of its widespread occurrence in the environment and its toxic effects on humans, mainly through fish ingestion. On the other hand, selenium is known by its antioxidant effect. For this reason, the knowledge of the correlation between mercury and selenium concentrations in fish samples can bring important information allowing a better understanding of mercury equilibrium in the inhabitants of the Amazon region. In this paper, total mercury and selenium levels were measured in carnivorous (47), omnivorous (44), and herbivorous (4) fish species caught in rivers and in an artificial lake in Cachoeira do PiriáMunicipality, situated in ParáState, Amazon region. The mercury concentration ranged from 1.15 to 13.44 nmol g^{-1} and selenium from 2.44 to 14.56 nmol g^{-1} for carnivorous species. For noncarnivorous species, mercury concentration ranged from 0.08 to 2.03 nmol g^{-1} and selenium from 1.27 to 15.32 nmol g^{-1}. The molar ratios between mercury and selenium contents obtained for carnivorous and noncarnivorous species were 0.65 and 0.14, respectively. Mercury and selenium levels were positively correlated with fish body mass (weight) only for *Hoplias malabaricus* ($n = 35$, $R^2 = 0.565$, $P < 0.005$ and $R^2 = 0.608$, $P < 0.005$, respectively). Selenium and mercury concentrations were statistically positively correlated only for *H. malabaricus* ($n = 35$, $R^2 = 0.787$, $P = 0.005$) and *Leporinus* sp. ($n = 38$, $R^2 = 0.485$, $P < 0.005$) known locally as Traíra (carnivorous) and Aracu (omnivorous), respectively.

Keywords: Mercury; Selenium; Fish; ParáState; Correlation

1. Introduction

Mercury is considered an environmental pollutant, mainly after the environmental disaster at Minamata and several poisoning accidents due to the use of mercury pesticides in agriculture (Koos and Longo, 1998). In the Amazon region, mercury pollution is often related to gold mining activities which use mercury to agglutinate fine gold particles through an amalgamation process. The gold rush took place in the early 1980s and the quantity of mercury released in the Amazon region estimated based on official gold production (700 tons) was approximately 1400 tons of Hg (considering that for each kilogram of Au recuperated usually 2 kg of Hg are needed). This value may be increased to 2000–4000 tons of Hg when the unofficial results (1000 to 2000 tons) (Figueiredo, 2001) are considered. During the 1990s, the gold mining activities decreased because of falling gold prices and only circa 300 tons of gold were produced officially; thus approximately 600 tons of Hg could have been released in the environment. Nowadays, an increase in the production of gold by mining companies has been observed due to the perspective of recuperation on the metal price (DNPM, 2002, 2003).

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Mercury enters the food chain directly as metallic precipitation in crops or after methylation in animals consumed by indigenous and riverside communities. It is also bioconcentrated, mainly as methylmercury, in the trophic fish chain, which constitutes the main protein source for the Amazon population. It is especially dangerous because this methylated form represents 73–98% of the total mercury (Barbosa et al., 1998; Maurice-Bourgoin et al., 2000).

It is widely known that the population of these regions has a fish-based diet with an intake of about 200 g of fish per day (Bidone et al., 1995; Malm, 1998). For this reason, these findings are significant, considering the risk to human health related to the daily consumption of fish by the Amazonian population (Hacon et al., 1997). Among the studied fish species in the Amazon region, carnivorous species are the most commonly found in the diet of the population. According to several studies accomplished in the region, these species present higher mercury contents than noncarnivorous species since they occupy the highest trophic level of the food chain (Palheta and Taylor, 1995; Brabo et al., 1999; Maurice-Bourgoin et al., 2000).

Early studies demonstrated that fish from various ecosystems may often attain mercury levels of toxicological concern even in areas considered not impacted by anthropogenic sources. For contaminated areas that suffer direct influence of the gold mining activities, Lodernius Martin (1992) reported an average Hg value of 1.10 $\mu g\, g^{-1}$ (5.49 $nmol\, g^{-1}$), Maurice-Bourgoin et al. (2000) found a range of 0.33–2.30 $\mu g\, g^{-1}$ (1.64–11.46 $nmol\, g^{-1}$) in carnivorous fish caught in the Beni River, Bolivian Amazonian Basin, and Santos et al. (2000) reported 0.63 $\mu g\, g^{-1}$ (3.14 $nmol\, g^{-1}$) in fish samples from Itaituba Municipality, ParáState. For comparison, in areas without anthropogenic sources, Brabo et al. (2003) found a mean Hg content of 1.287 $\mu g\, g^{-1}$ (6.41 $nmol\, g^{-1}$) in fish samples from Acre State and Barbosa et al. (2003) reported a mean Hg level of 0.69 $\mu g\, g^{-1}$ (3.45 $nmol\, g^{-1}$) in the carnivorous species caught from Negro River Basin.

Thus, the concentrations of mercury in several freshwater fish tissues exceed national and international guidelines (2.49 $nmol\, g^{-1}$ or 0.5 $\mu g\, g^{-1}$), for human consumption, established by the World Health Organization (WHO, 1972; Brasil, 1975).

Despite the widespread presence of mercury in the Amazon environment, there are no official reports of mercurialism in the region so far. For this reason, several studies aiming to evaluate the serious risks to the exposed population produced by potential mercury contamination have been undertaken. One of the most discussed possibilities is related to the effect of selenium compounds acting as a counteracting agent to the toxic effects of mercury (Goyer, 1998; Egeland and Middaugh, 1997). Selenium compounds have a large number of biological functions in humans and several other animal species. It is believed that a high selenium intake can reduce the risk of some forms of cancer and cardiovascular diseases (Egeland and Middaugh, 1997; Ónning, 2000). The most important and known action of selenium compounds, such as selenoprotein, is related to the reduction of the dangerous effects of mercury (Osman et al., 1998).

According to Yoneda and Suzuki (1997), Hg reacts first with a reduced form of selenite in an equimolar ratio, forming the $(Hg–Se)_n$ complex, followed by its binding to the specific plasma protein (selenoprotein P). In addition, the selenium-protein P may be effective primarily in detoxifying heavy metals such as Hg by sequestering these metals as stable complexes.

In the case of fish, the exact mechanisms of interaction between mercury and selenium are not fully understood (Watanabe et al., 1997; Navarro-Alarcón and López-Martínez, 2000). However, data obtained from fish studies in different ecosystems and in marine mammals indicate that Se, like Hg in aquatic organisms, is mostly found in concentrations that increase proportionally with the species trophic level. In the Amazon region, there are not many studies showing the relations of both Se and Hg in fish. Dorea et al. (1998), studying the correlation between mercury and selenium concentrations in fish from the Madeira River, Amazon Basin, reported a mercury range from 0.41 to 6.66 $nmol\, g^{-1}$ and a selenium range from 0.49 to 3.11 $nmol\, g^{-1}$, depending on the fish species.

The objectives of the present work were to assess the mercury levels in some fish species from regions highly exploited by gold mining activities, in Cachoeira do PiriáMunicipality, ParáState, verifying the direct influence of these activities on the fish contamination, and to evaluate the correlation between mercury and selenium concentrations in the fish species.

2. Material and methods

2.1. Description of the area

Cachoeira do PiriáMunicipality (Fig. 1) is situated in ParáState, about 250 km from Belém city, with a population of approximately 15,000 inhabitants (IBGE, 2000). In the 1980s, there was a gold rush in this area. Nowadays, however, there is only a Canadian mining company exploiting the area (Figueiredo, 2001). This region has rivers and lakes that directly and indirectly receive inputs of mercury from gold mining activities. The Lagoa Grande is an artificial lake of approximately $400 \times 150 \times 4$ m that supplies water for the hydraulic disassembly work performed during the exploitation of gold and receives effluents from this process, contaminated with mercury and other elements. It is also

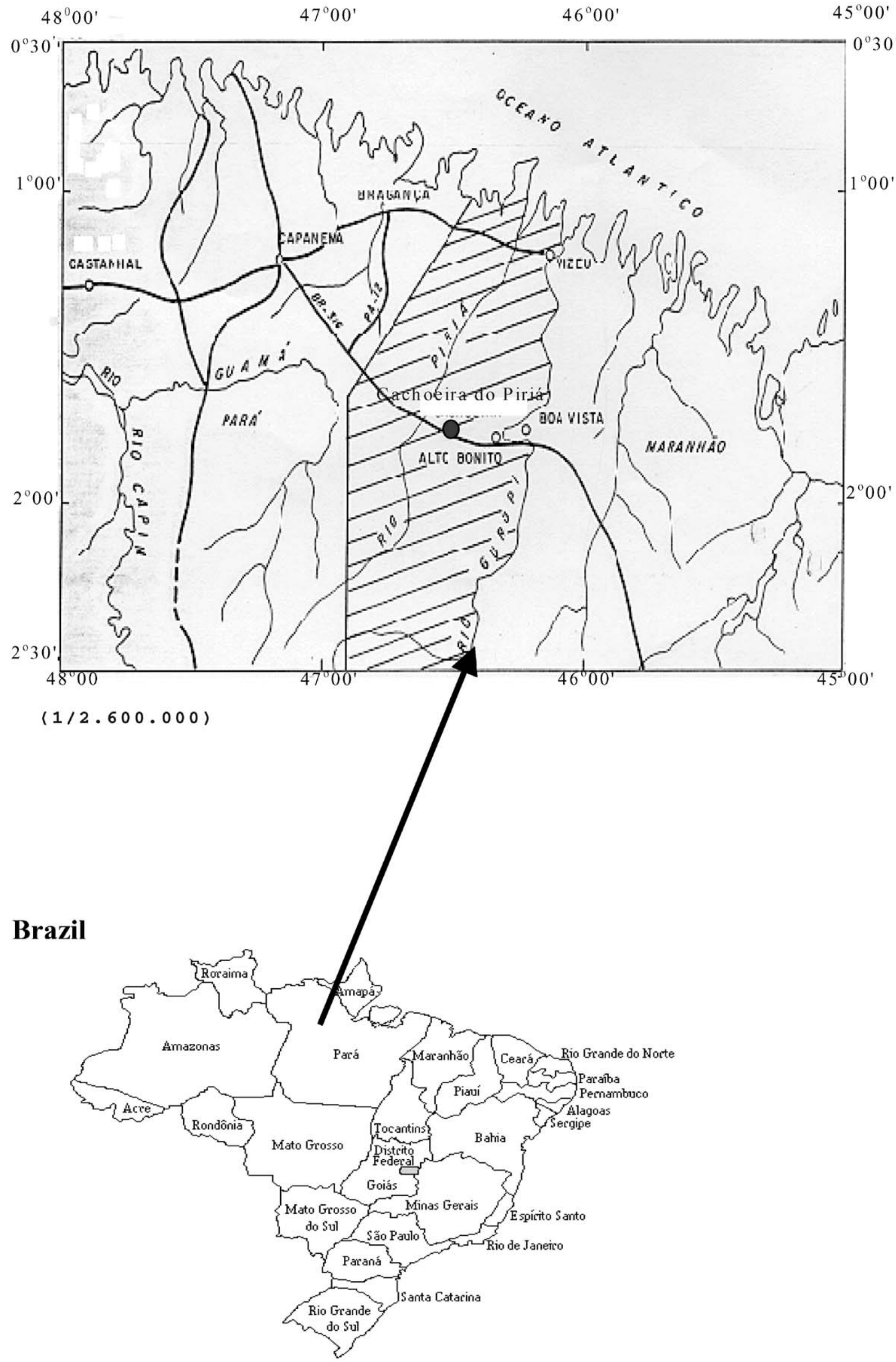


Fig. 1. Map showing the Cachoeira do PiriáMunicipality (DNPM, 1990).

relevant to mention that this Municipality is experiencing continual urban growth, where residences are built on the old gold mining sites and activities such as agriculture and subsistence fishing occur.

2.2. Sampling

The fish samples were collected in three periods, 1999, 2000, and 2001, by the Departamento Nacional de Produção Mineral (DNPM), ParáState. These samples were caught from PiriáRiver and an artificial lake called Lagoa Grande, which are considered important areas of fishing in the Municipality. The selected species were those considered the most important in the population's diet: *Leporinus* sp. (Aracu), *Mylossoma* sp. (Pacu), *Retroculus* sp. (Cará), *Glyptoperichthys* sp. (Acarí-bodó), *Rhamdia quelen* (Jandiá), *Cleithracara* sp. (Acará), *Leporinus* sp. (Piau), *Hoplias malabaricus* (Traíra), *Pseudoplatystoma* sp. (Surubim), and MãeRosa. All samples were weighed, stored in plastic bags, and frozen for transport. A total of 95 samples of carnivorous, omnivorous, and herbivorous species were analyzed.

2.3. Analysis of Hg and Se

Mercury determination was performed using cold vapor atomic absorption spectrometry and the

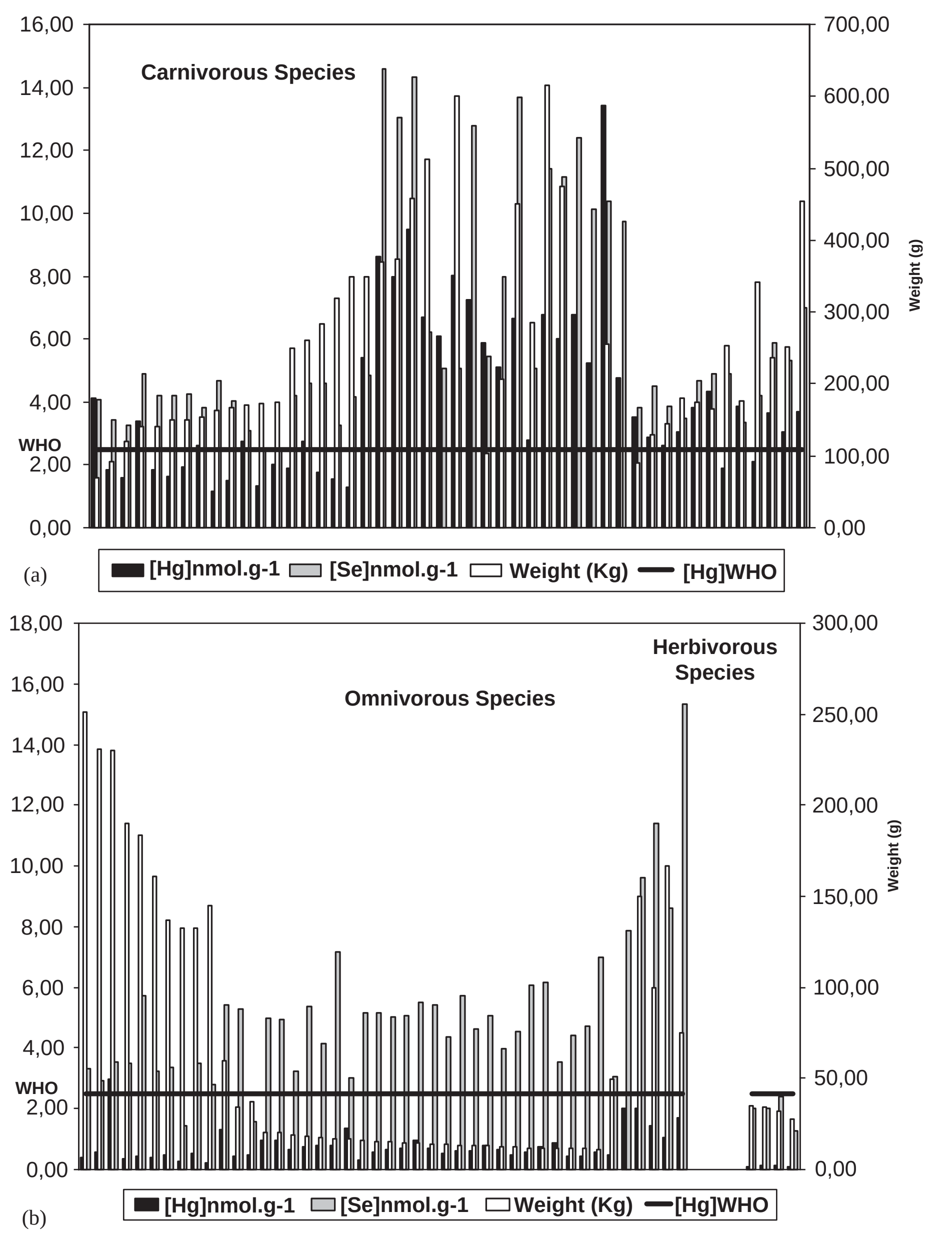


Fig. 2. Fish weight, Hg and Se concentrations, and maximum permissible concentration for human consumption by the World Health Organization for carnivorous and noncarnivorous species.

analytical procedure used (wet digestion) was that of Akagi et al. (1995) with some modifications. For total selenium analysis it was necessary to reduce the Se^{6+} species present in the solution to Se^{4+}, to form a volatile hydride. This reduction was carried out using a sulfamic acid solution (15% (w/v)) and hydrochloric acid (6 M), and heating the final solution for 40 min in a water bath (Ferrer et al., 1999). Selenium determination was done using hydride-generator atomic absorption spectrometry. To compare the concentrations of mercury and selenium with fish weight for each fish species, one-way ANOVA followed by parametric correlation (Pearson coefficient) was used. A P value less than 0.05 was considered statistically significant (SAS, 1995).

3. Results

The validation of the analytical methodology was performed with the certificate reference material from the National Research Council Canada (Dorm2, dogfish muscle; 4.64 ± 0.26 mg/kg for Hg and 1.40 ± 0.09 mg/kg for Se), giving mean concentration values of 4.67 ± 0.09 and 1.580 ± 0.009 mg/kg for Hg and Se, respectively ($n = 3$).

The mercury and selenium concentrations varied depending on the trophic position of the fish species (Fig. 2). For carnivorous species, the average mercury and selenium concentrations were 4.09 ± 2.63 and 6.25 ± 3.53 nmol g^{-1}, respectively, while for noncarnivorous species they were 0.68 ± 0.44 and 4.86 ± 2.55 nmol g^{-1}, respectively (Table 1).

The mean molar ratios between mercury and selenium contents for carnivorous and noncarnivorous species were 0.65 and 0.14, respectively. The molar ratios of Hg/Se increased according to the fish trophic level (Table 1).

There were statistically significant correlations between Hg and Se concentrations only for the carnivorous species *H. malabaricus* ($n = 35$, $R^2 = 0.787$, $P = 0.005$) and for the omnivorous species *Leporinus* sp. ($n = 38$, $R^2 = 0.485$, $P < 0.005$), which are illustrated in Fig. 3.

The mercury and selenium concentrations were significantly correlated with fish weight only for *H. malabaricus* ($R^2 = 0.565$, $P < 0.005$ and $R^2 = 0.608$, $P < 0.005$, respectively) (Fig. 4).

4. Discussion

In this work, it was verified that the distribution of mercury and selenium in fish depends mainly on their dietary habits. Only the carnivorous species (about 68%) presented mercury levels above the maximum permissible level for human consumption (0.50 μg g^{-1} or 2.49 nmol g^{-1}, WHO), which are in the same order of magnitude as that of the other fish from rivers impacted by gold mining activity in the Amazon region (Dorea, 2003) and from sites without anthropogenic sources, i.e., the Negro River (Barbosa et al., 2003) and Acre State (Brabo et al., 2003). In these sites, the high Hg content found in the Negro River, according to Jardim and Fadini (1999), is due to the black waters of this river inhibiting the volatilization of Hg extracted from the soil

Table 1
Mercury and selenium concentrations for carnivorous and noncarnivorous species from Cachoeira do PiriáMunicipality, ParáState, Brazil

Species (popular name)	Food habits (n)	Total weight (g)	Hg		Se		Hg/Se
		Mean ± SD	Mean ± SD (nmol g^{-1})	Range (nmol g^{-1})	Mean ± SD (nmol g^{-1})	Range (nmol g^{-1})	
Leporinus sp. (Aracu)	Omnivorous (38)	60.32 ± 75.99	0.62 ± 0.26	0.23–1.35	4.47 ± 1.31	1.45–7.15	0.14
Mylossoma sp. (Pacu)	Herbivorous (4)	32.42 ± 3.32	0.11 ± 0.02	0.08–0.12	1.93 ± 0.48	1.27–2.41	0.06
Retroculus sp. (Cará)	Omnivorous (1)	—	1.99	—	7.85	—	0.25
Glyptoperichthys sp. (Acarí–bodó)	Omnivorous (2)	125.00 ± 35.36	1.73 ± 0.42	1.44–2.03	10.51 ± 1.25	9.63–11.40	0.16
Rhamdia quelen (Jandiá)	Omnivorous (1)	75.00[b]	1.69	—	15.32	—	0.11
Cleithracara sp. (Acará)	Omnivorous (1)	49.58[b]	0.48	—	3.04	—	0.16
Leporinus sp. (Piau)	Omnivorous (1)	167.00[b]	1.05	—	8.61	—	0.12
Hoplias malabaricus (Traíra)	Carnivorous (35)	270.37 ± 151.20	4.39 ± 2.97	1.15–13.44	6.80 ± 3.91	2.44–14.56	0.65
(MãeRosa)[a]	Carnivorous (5)	144.00 ± 36.64	3.18 ± 0.48	2.63–3.81	4.07 ± 0.49	3.50–4.67	0.78
Pseudoplatystoma sp. (Surubim)	Carnivorous (7)	268.78 ± 100.17	3.22 ± 0.93	1.87–4.33	5.08 ± 1.17	3.36–7.01	0.63
Noncarnivorous	(48)	63.05 ± 71.89	0.68 ± 0.44	0.08–2.03	4.86 ± 2.55	1.27–15.32	0.14
Carnivorous	(47)	255.42 ± 139.87	4.09 ± 2.63	1.15–13.44	6.25 ± 3.53	2.44–14.56	0.65

n, number of individuals for each species; SD, standard deviation
[a]Species not classified.
[b]Only one individual.

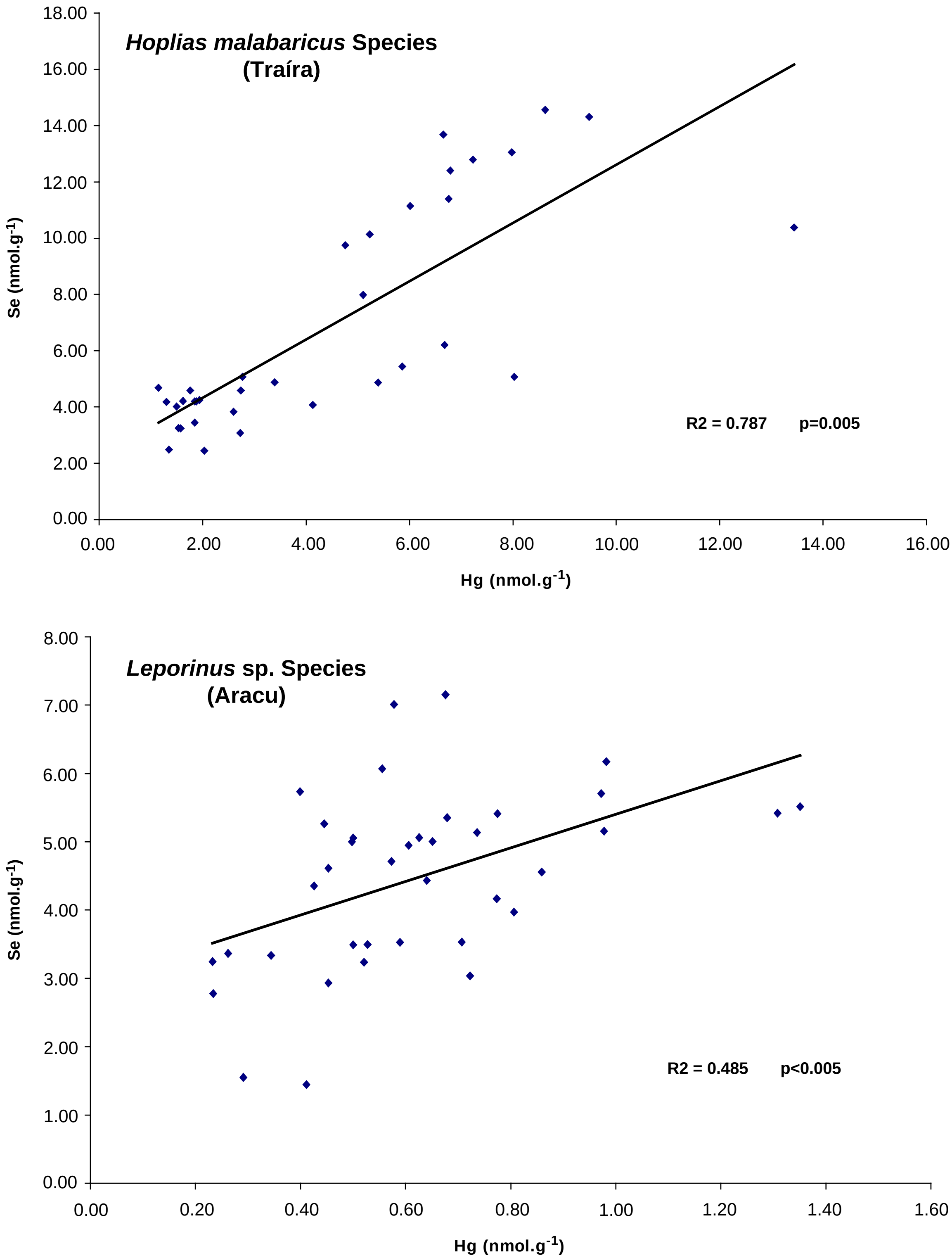
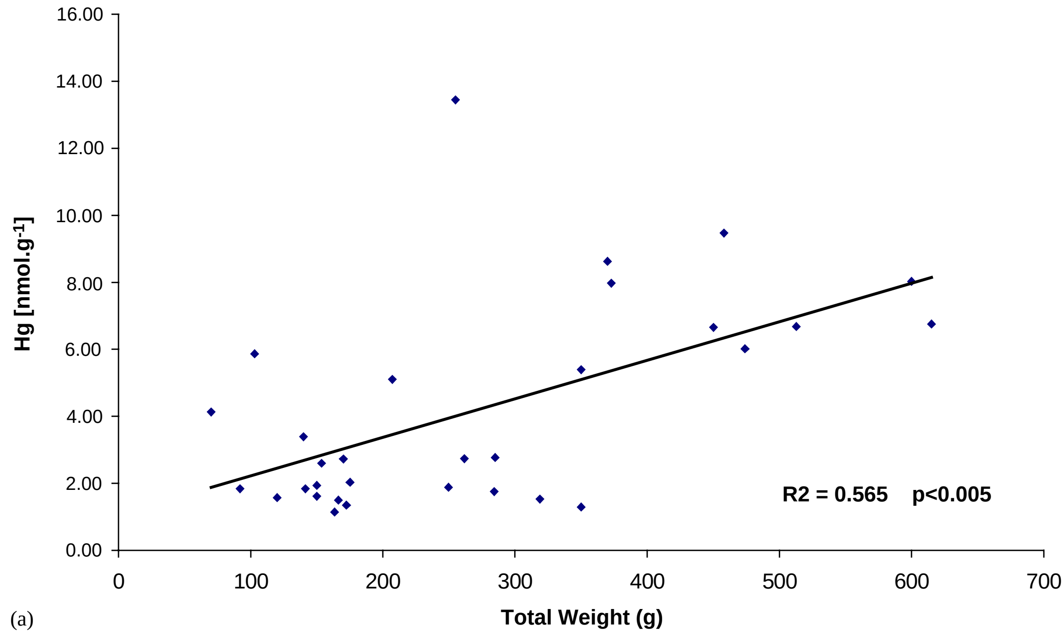


Fig. 3. Correlations between Hg and Se concentrations for *H. malabaricus* (Traíra) (a) and (b) *Leporinus* sp. (Aracu) (b) from Cachoeira do Piriá, ParáState, Brazil.

and assisting the interaction with the organic matter. Brabo et al. (2003) assumed that the high Hg level in fish from the Acre and Purus rivers and some of their tributaries was originally from the ferruginous nodules and fossilized materials. Approximately 66% and 85% of carnivorous and noncarnivorous species, respectively, presented selenium levels within the range requirement for fish, 1.9–6.33 nmol g^{-1}



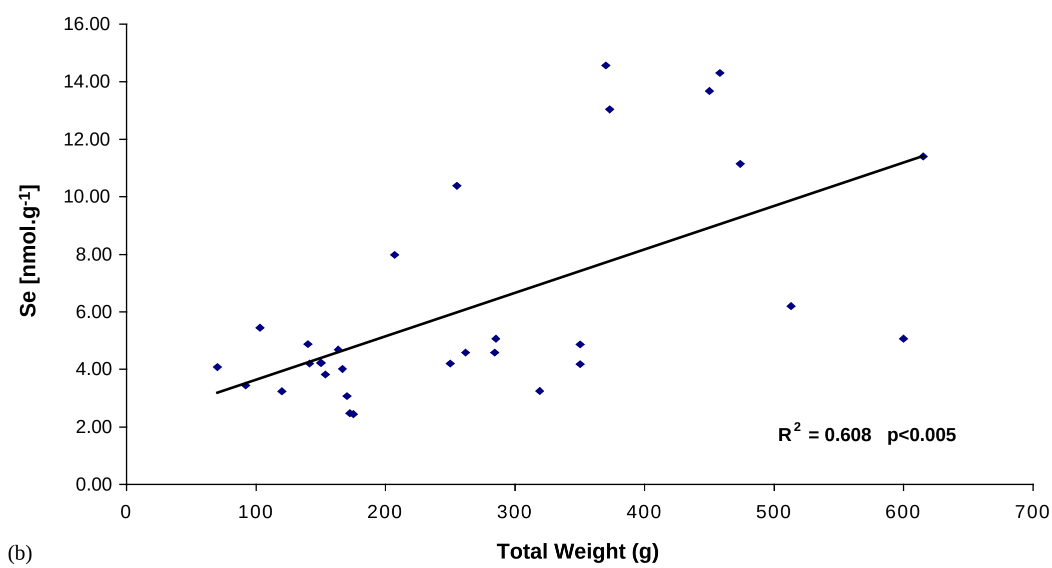


Fig. 4. Correlations between total fish weight and Hg (a) and Se (b) concentrations for *H. malabaricus* (Traíra) from Cachoeira do Piriá, ParáState, Brazil.

(0.15–0.50 $\mu g\,g^{-1}$), established by Watanabe et al. (1997).

The higher levels of Hg (5.10–13.44 $nmol\,g^{-1}$) found in some individuals were expected considering the sampling locale, an artificial lake known as Lagoa Grande. This lake was created for gold exploitation and nowadays is a subsistence fishing area that can bring serious health risks for the local human population. On the other hand, these individuals presented also the highest selenium contents and mean molar ratio obtained between these elements (Table 1), indicating that both are equivalently present in these fish samples. The correlation between these metals was also studied in some fish samples from rivers of the Amazon region (Dorea et al., 1998) and from other freshwater and marine rivers of the world (Cappon and Smith, 1982; Burger et al., 2001).

The mechanism of interaction between mercury and selenium in the bioassimilation processes in fish and their transference to the human population has not been fully explained, but Hagmar et al. (1998) reported a correlation between fish intake by Latvian subjects and plasma selenium and selenoproteins, indicating that fish had a considerable impact on selenium status.

It is important to notice that Vasconcellos et al. (2000) and de Campos et al. (2002) reported similar significant correlations between these elements in hair samples from different sites of the Amazon region. According to these authors, this correlation may originate from the high fish consumption, since it constitutes the most important food source of Hg and Se in the diet of the Amazon population.

In addition, in this study, mercury and selenium levels and fish weight showed significant correlations only for *H. malabaricus* (Fig. 4). Therefore, these correlations are not always observed and this has been reported in the literature. Several factors influence this relationships the species biodiversity found in the Amazon region, the variation among species in metabolism, the food habits, and capacity of migration, and other variable parameters of the aquatic environment (Kadrabova et al., 1997; Boening, 2000; Hylander et al., 2000; Lima et al., 2000; Maurice-Bourgoin et al., 2000; Ónning, 2000; Santos et al., 2000).

5. Conclusion

Our study demonstrated that the presence of Hg in fish is positively correlated to the concentrations of Se. As previously mentioned, the correlation between these metals was also reported in riverside communities, hair samples from the Amazon, which confirms the exposure via consumption of fish to be a significant source of human exposures. Thus, the high contents of Hg found in this work and the absence of official reports on mercury poisoning in the inhabitants of Amazon region suggest that Se can be acting as a detoxification agent. However, a more comprehensive study concerning the interaction of specific mercury and selenium chemical forms is necessary and mercury toxicity should be quantitatively determined in the foods and local population.

Moreover, we also suggest a preventive education campaign in Cachoeira do PiriáMunicipality, aiming to clearly inform this population about the health risks connected to the consumption of fish caught in the lake Lagoa Grande.

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