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Mercury in freshwater, estuarine, and marine fishes from Southern Brazil and its ecological implication

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Abstract In this study, we measured the mercury concentration in 27 different fish species with high commercial value. Samples were taken from a region characterized by the diversity of aquatic environments. Mercury concentration in marine fish species varied from 30.4 to 216 ng g^{-1}, while in estuarine species, it varied from 12.4 to 60.3 ng g^{-1}. Compared to mercury concentration in marine species, none of the specimens from estuarine environment has reached a mercury concentration of 100 ng g^{-1}.However, mercury concentrations in species from the freshwater Patos lagoon are remarkably higher (15.3 to 462 ng g^{-1}) than those from the estuarine or marine region. Even though mercury concentrations in these fish species did not exceed the maximum level (500 ng g^{-1}) allowed by WHO for human consumption, they represent the main food source for sea birds and mammals coming from South Pole during their migration period.

Keywords Mercury · Ecological impact · Aquatic environment · Patos lagoon · Southern Brazil

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Introduction

Contamination of fish products by mercury is of great concern throughout the world. Of particular threat to human health are those mercury-contaminated regions where fish is a basic component in the diet of the local population (Harada 1995).

It is estimated that in Brazil, an average person consumes on 6.8 kg fish $year^{-1}$. Furthermore, Southeast and South regions represent 88.5% of the total marine/estuarine fish catch of the country.

Studies regarding contamination in aquatic environments pollution by mercury were conduced in the Northern and Central regions of Brazil, where high mercury levels are mostly caused by gold mining, industry effluents, and pesticide use (Moraes et al. 1997; Lacerda et al. 2000; Bastos et al. 2006). In the Southeast and South regions, the major sources of mercury contamination come from industries and domestic effluents as well as atmospheric emissions (Paraquetti et al. 2004; Silva-Filho et al. 2006; Mirlean et al. 2003, 2005).

The state of Rio Grande do Sul state, located in the South region of Brazil, has a population of more than 10 million people. There are 40 industries of fish processing plants situated in three cities: Porto Alegre, Rio Grande, and Pelotas. Moreover, the Patos lagoon contains the third most important harbor of Brazil, which facilitates the transport of chemical products, grains, and agricultural machines, among other pollutant sources.

Due to the diversity of aquatic environments, this region also has great ecological significance because it represents an important feeding and breeding area for aquatic birds and mammals coming from the South Pole (Antarctica and Patagonia) during migration.

In this region, previous studies have found that a higher concentration of mercury (up to 17 mg kg^{-1}) was present in sediments of the shallow zone of the estuary. Industrial and urban effluents were determined to be the most probable sources of mercury pollution in the estuary. An increased concentration of mercury was also recorded in atmospheric precipitation in zone affected by industrial emissions. This area covers the estuary, the freshwater part of the lagoon, and the coastal area with freshwater lakes (Mirlean et al. 2003, 2005).

The first data on mercury sediment contamination from Patos lagoon were obtained in 1999 during the inspection of a tanker accident in the Rio Grande harbor (Mirlean et al. 2001). The concentration of total mercury in the canal sediments during the accident had reached 5 mg kg^{-1}.

Information reported recently on the aquatic mercury environmental contamination in the southernmost Brazilian state of Rio Grande do Sul (Mirlean et al. 2001) has prompted the study of mercury concentrations in fish from the Patos Lagoon estuary (Niencheski et al. 2001; Mirlean et al. 2005). To date, however, only a few fish species from this region have been tested for mercury contamination.

The fishing community near Patos lagoon is one of the most populated in Brazil. Fish are caught in both marine and freshwater environments along more than 800 km of the Brazilian coastline. Besides their use as a principal dietary component for the local population, fish are also exported to other Brazilian regions and abroad.

The aim of this study was to determine mercury concentration distribution in fish species from different aquatic systems in Southern Brazil and to assess its ecological environmental risk as a contribution to the knowledge and rational management of these regions in future. The results would serve as a baseline against which future anthropogenic effects can be assessed.

Materials and methods

The study area (29° 18′ S to 33° 45′ S) includes Patos lagoon with its estuary, freshwater coastal lakes, and waters from the Atlantic Ocean with range of 200 miles (Fig. 1).

Fish samples were obtained between October 2003 and June 2005 from public markets in Rio Grande, Pelotas, and Porto Alegre cities, and the fishing catchments were specified by the fisherman. The samples were sorted by species, wrapped in clean plastic bags, and transported to the laboratory in ice boxes. In total, 99 fish specimens totaling 27 different species were collected (Table 1). These species were identified according to Fisher et al. (2004). Information about their migratory habits was taken from Garcia et al. (2003). The trophic level of the fish species was established from data of Froese and Pauly (2006).

Fish specimens were measured and weighed prior to dissection. Edible muscle samples of 3–5 g wet weight were taken from just behind the gills and below the dorsal fin; skin was removed, and the samples were frozen. Clean tissue processing was done following the methods of Niencheski et al. (2001).

Digestion methods for fish tissue were followed according to Zhou and Wong (2000). Fish tissue samples were predigested in 8 mL of concentrated HNO_3 and H_2SO_4 (2:1 v/v) at 25°C for 3 h, then at 60°C for 5 h. Five milliliters of 30% H_2O_2 was added to the samples in 0.5-mL increments, with time allowed for foaming to subside between additions. The temperature was then raised to 65°C, and digestion proceeded until the samples

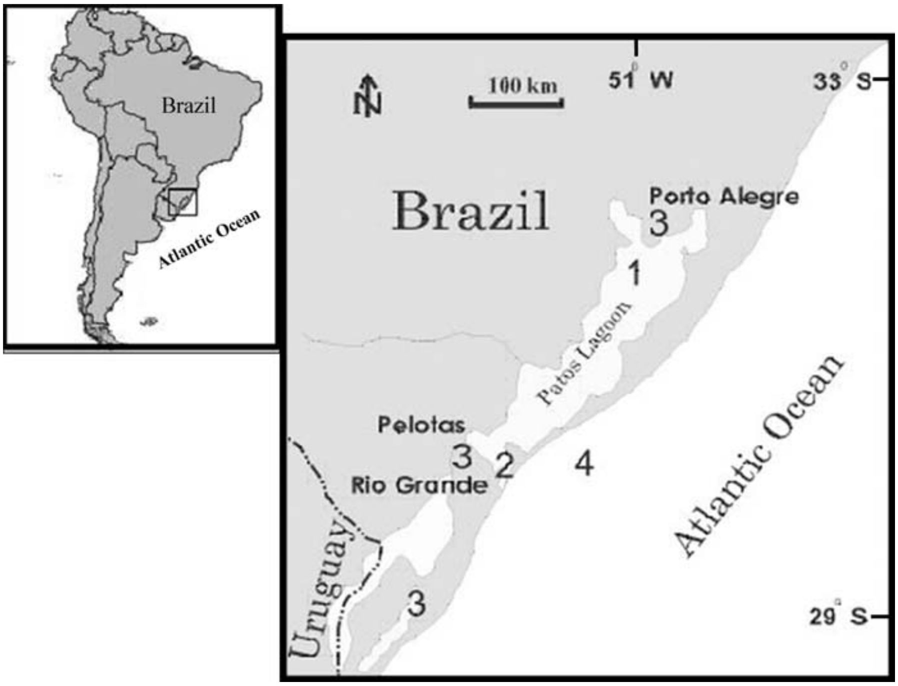

Fig. 1 The study area: 1 - Patos lagoon freshwater; 2 - Patos lagoon estuary; 3 - Inner freshwater lakes; 4 - Brazilian marine exclusive zone

Table 1 Different species collected

Specie name	Vulgar name	Primary trophic class
Hoplias malabaricus	Traira	Piscivore
Oligosarcus jenynsii	Tambica-amarela	Piscivore
Oligosarcus robustus	Tambica-vermelha	Piscivore
Astyanax spp	Lambari	Detrivore
Rhamdia quelen	Jundiá	Omnivore
Pimelodus maculatus	Pintado	Omnivore
Geophagus brasiliensis	Cara	Zooplanktivore
Odontesthes bonariensis	Peixe-rei	Zooplanktivore
Leporinus obtusidens	Piava	Herbivore
Genidens genidens	Bagre	Omnivore
Netuma barba	Bagre	Omnivore
Odontesthes argentinensis	Peixe-rei	Zooplanktivore
Mugil platanus	Tainha	Zooplanktivore
Micropogonias furnieri	Corvina	Carnivore
Paralichtys orbignyanus	Linguado	Carnivore
Conger orbignyanus	Congro-rosa	Zooplanktivore
Urophycis brasiliensis	Abrotea	Carnivore
Cynoscion guatucupa	Pescada	Carnivore
Pomatomus saltatrix	Anchova	Carnivore
Menticirrus littoralis	Papa-terra	Carnivore
Pagrus pagrus	Pagro	Carnivore
Epinephelus marginatus	Garoupa	Carnivore
Merluccius hubbsi	Merlusa	Carnivore
Polyprion americanus	Cherne	Carnivore
Squatina argentina	Cação	Carnivore
Macrodon ancylodon	Pescadinha	Piscivore
Lophius gastrphysus	Peixe-sapo	Piscivore

turned colorless or light yellow. A cold vapor system, coupled with a gold trap and GBC 932 atomic absorption spectrophotometer, was used for determining total mercury concentrations.

All samples were analyzed in triplicate. The coefficients of variation for all triplicates were less than 6%. The accuracy and precision was assessed by analysis of a reference material IAEA 350 tuna homogenate (4.68 ± 0.28 μg g^{-1}). The results obtained (4.53 ± 0.13 μg g^{-1}, $n = 6$) showed that excellent mercury recovery was (97%) and a coefficient of variation of less than 5%, thus validating the employed methodology.

Results and discussion

Mercury concentration in marine fish species varied from 30.4 to 215.8 ng g^{-1} (Table 2). The most prevalent species (Pescada *Cynoscion guatucupa*, Pescadinha *Macrodon ancylodon*, Merlusa *Merluccius hubbsi*, Anchova *Pomatomus saltatrix*)

Table 2 Mercury concentration in fish species

Specie name	Fork length, range (cm)	Sample size	Hg concentration, mean (range)
Freshwater, coastal lakes			
Astyanax spp.	10–12	6	268.7 (171.4–382.9)
Geophagus brasiliensis	14–16	4	32.5 (23.0–46.2)
Hoplias malabaricus	39–42	3	74.3 (63.1–88.5)
Oligosarcus jenynsii	20–22	4	344.3 (269.2–451.2)
Patos Lagoon			
Astyanax spp.	29–29	3	203.6 (76.3–346.6)
Rhamdia quelen	33–36	4	153.9 (89.2–233.0)
Pimelodus maculatus	29–30	3	130.0 (86.5–171.6)
Geophagus brasiliensis	20–22	3	20.9 (15.3–32.0)
Odontesthes bonariensis	37–40	4	75.9 (52.2–113.3)
Leporinus obtusidens	50–50	3	84.6 (82.6–86.6)
Hoplias malabaricus	50–54	3	173.5 (156.3–197.1)
Oligosarcus jenynsii	23–25	4	349.0 (292.7–462.0)
Oligosarcus robustus	28–32	4	187.5 (149.8–237.1)
Patos Lagoon, estuary			
Genidens genidens	36	1	57.8
Netuma barba	49	1	60.3
Odontesthes argentinensis	20–22	3	41.6 (35.4–52.9)
Mugil platanus	31–50	4	12.4 (9.9–19.5)
Micropogonias furnieri	48–50	2	53.4 (32.7–74.1)
Paralichtys orbignyanus	24–50	4	47.1 (14.5–79.0)
Atlantic Ocean, near-shore waters			
Conger orbignyanus	65–78	3	95.9 (57.4–138.3)
Urophycis brasiliensis	46–56	4	35.0 (16.7–54.3)
Cynoscion guatucupa	35–50	4	88.8 (65.3–126.9)
Pomatomus saltatrix	40–54	4	52.8 (32.0–75.1)
Menticirrus littoralis	40–48	4	124.3 (86.8–210.5)
Pagrus pagrus	29–36	3	68.0 (60.7–75.7)
Epinephelus marginatus	90	1	215.8
Merluccius hubbsi	50–78	2	84.2 (66.4–101.9)
Polyprion americanus	46–48	3	48.5 (45.3–51.0)
Squatina argentina	58–64	2	30.4 (24.5–36.3)
Macrodon ancylodon	35–40	4	38.5 (28.9–48.5)
Lophius gastrphysus	43–50	2	95.8 (69.3–122.3)

contained, on average, a mercury concentration lower than 100 ng g^{-1}. The *C. guatucupa* and *M. ancylodon* species from the Southern Brazilian coast showed mercury concentrations of about two to three times lower than the corresponding data from equatorial regions.

The increase in mercury concentrations in fish species from the Atlantic waters off the coast of Surinam is explained by the influence of gold mining in local rivers (Mol et al. 2001). Only two species of sampled fish (Papa-terra *Menticirrus littoralis* and Garoupa *Epinephelus marginatus*) exhibited an average mercury concentration above 100 ng g^{-1}. The Papa-terra mainly feeds on benthonic organisms that can be an additional mercury source, especially in the region where freshwater empties into the ocean. The Garoupa was the largest species of fish examined in the present study, so the relatively elevated concentration of mercury was probably linked to the higher age of this specimen. The low mercury concentration obtained from the majority of analyzed marine fish species could be attributable to the minor influence of local sources on coastal waters contamination, taking into account the effect of contaminant dilution into great water volumes.

Mercury concentration in estuarine fishes varied within small limits, and compared to marine fish, none of the analyzed specimens has achieved a mercury concentration of 100 ng g^{-1} (Table 2). Tainha *Mugil platanus*, which is a very common catch of local fishermen, exhibited the lowest mercury concentration (<20 ng g^{-1}) in this study. Our data were well correlated with the data for the same genus from Guanabara Bay (Kehrig et al. 2002) and showed a slightly lower mercury concentration compared to the specimens isolated from the Mediterranean Sea (Storelli et al. 2006). However, according to Marcovecchio (2004), Tainha (*Mugil liza*) from Samborombon Bay (on the Argentinean Atlantic coast) contains, on average, 20 times more mercury (400 ng g^{-1}) than the ones obtained in this study. This probably reflects the high metal concentrations in surface sediments and suspended particulate matter present in that environment. The Corvina *Micropogonias furnieri*, which usually contributes to a large portion of the local catch, contained significantly ($p < 0.05$) more mercury than did the Tainha species (Table 2). Nevertheless, the Corvina from the Patos lagoon estuary contains almost half as much mercury as the same species from other Brazilian estuaries (Kehrig et al. 1998). Thus, the mercury contamination of Patos lagoon estuarine deposits did not exhibit significant effect on mercury accumulation in estuarine fish species.

The fish samples from the freshwater part of the Patos lagoon contained noticeably more mercury than those taken from the estuary or ocean (Table 2). The distribution of mercury in freshwater fish species as a whole corresponded to their trophic habit. Zooplanktivore species (Cara *Geophagus brasiliensis*, Peixe-rei *Odontesthes bonariensis*) and herbivore ones (Piava *Leporinus obtusidens*) contained, on average, two to five times less mercury than did the piscivorous species (Traira *Hoplias malabaricus* and Tambica *Oligosarcus*). An exception is Lambari Astyanax spp., which is a detrivore. Samples of this species contained more mercury than did some piscivore and omnivore species (Traira, Jundia, Pintado). The highest concentrations measured, 451 and 462 ng g^{-1}, were from samples of Tambica from the suburban coastal lake near the city of Rio Grande and the freshwater region of the Patos lagoon near the city of Porto Alegre. These concentrations are close to the limit (500 ng g^{-1}) for human consumption recommended by the World Health Organization (WHO 1972).

The high content of mercury in Tambica was probably due to biomagnification since this fish species is the principal predator of lambari. We reported earlier that mercury concentration in Lambari from coastal lakes positively correlates with mercury concentration in local atmospheric precipitation (Mirlean et al. 2005), which increases with proximity to urban areas and industrial sources. We suggested that this linkage is closely related to lambari feeding behavior in the upper layer of the water column.

This study shows that the fish species from South Brazilian coastal waters, the estuary of the Patos Lagoon, and freshwater bodies contain mercury concentrations that are below the

WHO-recommended limits. The low level of mercury in fish from contaminated estuarine environment suggests a low bioavailability of mercury in estuarine sediments.

Sulphate-reducing bacteria are recognized as the main mercury methylation mediators in anoxic, marine, estuarine, and freshwater sediments (Gilmour et al. 1992; King et al. 2000), as well as anoxic locustae waters (Matilainen 1995).

The presence of sulphates stimulates sulphate-reducing bacteria activity, increasing methyl mercury production (Gilmour et al. 1992). However, high sulphite levels (induced by sulphate reduction) are susceptible to limited Hg availability via complexation reactions and therefore significantly decrease methylation rates (Gilmour et al. 1998).

Moreover, it has been shown (Mason and Sullivan 1999) that dimethyl mercury, a volatile, organic mercury compound, is predominant in marine environments.

The consumption of two freshwater fish species (Lambari and Tambica) caught close to industrial zones and cities represent a relative danger for human health.

King et al. (2000) showed that bacteria-utilizing acetate as a carbon source seems to methylate Hg more efficiently due to the methyl transferase enzyme. Moreover, in freshwaters environment, Pak and Bartha (1998) observed an increase in mercury methylation due to a specific synergic effect between sulphate-reducing and methanogenics bacteria.

The mercury concentrations found in this study represents no threat to humans. However, data from literature on the subject (Gonzales-Solis et al. 2002; Kunito et al. 2004; Santos et al. 2006) have pointed out that high concentration of mercury and other metals in sea birds and mammals from the South Pole may be due to their feeding habits in this area during their migration periods.

The exposure to low and sub-lethal mercury concentrations, in an environment moderately contaminated environment during a long period of time, could cause damage to the growth, survival, and reproduction of organisms in a ecological community, seriously compromising biodiversity and the sustainability of these communities (Castilhos et al. 2004; Porto et al. 2005).

In particular, piscivorous birds show the highest Hg levels in muscle tissues compared to the species that consumes marine invertebrates ones (Bryan 1979). Low egg production and high embryonic mortality rates were observed in birds that feed on a diet that is highly contaminated with mercury (Ochoa-acuña et al. 2002; Cifuentes et al. 2003; Zolfaghari et al. 2007).

Overall, the levels of mercury that we found in fish species in Southern Brazil suggest that this contaminant may be a concern for wildlife. Even though we saw no deformities indicative of heavy metal poisoning in fledglings, contaminant levels need to continue to be monitored carefully. The disturbance of soils and sediments due to industrial activity, urban growth, and dredging increases metal bioavailability that will in turn increase the exposure of birds and mammals to toxic levels of mercury.

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