

REVIEW OF MERCURY BIOMONITORING ALONG THE BRAZILIAN COAST

Monica F. Costa¹; Helena A. Kehrig².

¹Departamento de Oceanografia da Universidade Federal de Pernambuco. Av. Arquitetura s/n, Cidade Universitária, Recife, PE, CEP: 50740-550, Brazil; e-mail: mfc@npd.ufpe.br;

²Departamento de Química da Pontifícia Universidade Católica do Rio de Janeiro (PUC-Rio). Rua Marques de São Vicente no. 225, Gávea, Rio de Janeiro, RJ, Brazil; e-mail: kehrig@domain.com.br

ABSTRACT

The study of Hg speciation and its global cycling in the marine environment has quickly evolved in the last ten years. The role of Hg contamination assessment on coastal environments through biological indicators remained of paramount importance. The present literature review shows that the use of biological indicators in the assessment of Hg contamination in tropical coastal environments is still somewhat meager. We focus on the biomonitoring work done along the Brazilian coast for the last 20 years. The published studies have been scarce, concentrating mainly on GB and other heavily contaminated estuaries. The two types of tissues from biomonitors preferred are muscle from fish of various feeding habits (i.e. Atlantic croaker and mullets) and homogenates of all the soft parts of bivalves (i.e. common mussels and mangrove oysters). Some other plant (algae and mangrove trees) and animal (crustacea, sharks and mammals) groups have also been tested and reported. The works have been divided into two large groups according to their date of publication and analytical and/or ecological quality.

Keywords: mercury, bioindicators, Brazilian coast

INTRODUCTION

The study of Hg speciation and its global and local cycling in the marine and coastal environment has quickly evolved in the last years. Sea surface interaction with the overlaying atmosphere and other atmospheric features of the metal's cycle have been the highlight of these progresses.

However, Hg contamination assessment on coastal environments through biological indicators remains of paramount importance. The biological compartment is a key link between Hg contamination in coastal environments and man through the consumption of fresh or processed fisheries products. This compartment may also be responsible for part of the Hg contamination transfer from coastal to open waters mainly through animal body burden. In addition to the ecological relevance, biomonitoring of coastal waters still is a better analytical alternative than the direct measurement of trace amounts in the water and particulate phases. Unfortunately the use of biological indicators in the assessment of Hg contamination in Brazilian coastal environments is still somewhat meager. This sort of work faces a number of difficulties in developing countries. Hg analysis requires the establishment of dedicated laboratory setups and specific personnel training, being often rated as a secondary priority in detriment of other metals and organic contaminants in general. However, only the analysis of biological tissues as indicators of Hg contamination will be generate reliable informethylation on the metal's bioavailability in coastal environments and the potential risks for human health.

In Brazil, coastal areas are much less studied concerning Hg contamination than the Amazon and Pantanal regions due to the artizanal goldmining activity being one of the most important sources of Hg to the environment in this country. Hg contamination is limited along the Brazilian coast, being considerable only in the vicinities of chlor-alkali plants which operated, or still operate, Hg cells. There are also cases of areas

contaminated by agricultural run-off, from sugar-cane plantations which used Hg compounds as chemical defensives in the form of phenyl-Hg, or near large industrial estates.

Marine or estuarine plants and animals were used, covering a large range of latitudes (2 to 32° S) and therefore very different climatic and oceanographic conditions, as well as different stages of industrial and economic development, urban occupation of coastal areas, deforestation and sustainability of agricultural land. The two types of biomonitors preferred are fish of various feeding habits. The two most used fish were *Micropogonias furnieri* (Atlantic croaker), carnivore; and the genus *Mugil* (mulletts), limnophagous or herbivores. Second are bivalves. Both, *Perna perna* (common mussels) and *Crassostrea rhizophorae* (mangrove oysters), are widely used. All the species are listed on Table 1.

In 1998, the work done in the Amazon and Pantanal regions resulted in a change in the Brazilian regulation from 1975. The new Brazilian legislation established that only non-predatory species destined to human consumption should presented total Hg < 0.50 µgHg.g⁻¹ ww, while the predatory fish species could have total Hg up to 1.00 µgHg.g⁻¹ w.w, and still be fit for human consumption. Although this change was made thinking of the freshwater fish, it included the top of the marine food chain.

Table 1: Species used for mercury contamination assessment along the Brazilian coast.

MACRO ALGAE: <i>Enteromorpha</i> sp.
TRACHEOPHYTES: <i>Avicenia schaueriana</i> ; <i>Laguncularia racemosa</i> ; <i>Rhizophorae mangle</i>
MOLLUSCA BIVALVIA: <i>Anomalocardia brasiliana</i> ; <i>Crassostrea Rhizophorae</i> ; <i>Mytella falcata</i> ; <i>Mytella guyanensis</i> ; <i>Perna perna</i> ; <i>Pitar fulminata</i> ; <i>Tagelus plebeius</i>
CRUSTACEA: <i>Callinectes danae</i> ; <i>Callinectes</i> sp.; <i>Penaeus brasiliensis</i> ; <i>Farfantepenaeus paulensis</i>
SHARKS & RAYS: <i>Carcharhinus</i> spp.; <i>Galeocerdo cuvieri</i> ; <i>Isurus</i> sp.; <i>Mustelus higmani</i> ; <i>Mustelus</i> spp.; <i>Odontaspis</i> sp.; <i>Prionace glauca</i> ; <i>Rhizoprionodon lalandei</i> ; <i>Rhizoprionodon porosus</i> ; <i>Sphyrna</i> sp.; <i>Squalus</i> spp.; <i>Squatina argentina</i>
FISH: <i>Achirus</i> sp.; <i>Anchoviella lepidentostoli</i> ; <i>Arius spixii</i> ; <i>Bagre bagre</i> ; <i>Bagre</i> spp.; <i>Caranx</i> sp.; <i>Centropomus undecimalis</i> ; <i>Centropomus</i> spp.; <i>Chaetodipterus faber</i> ; <i>Cynoscion virescens</i> ; <i>Eucinostomus gula</i> ; <i>Jenysia lineata</i> ; <i>Katsuwonus pelamis</i> ; <i>Lopholatilus villarii</i> ; <i>Lycengraulis grosidensis</i> ; <i>Macrodon ancylodon</i> ; <i>Menticirrhus americanus</i> ; <i>Micropogonias furnieri</i> ; <i>Mugil</i> spp.; <i>Mugil brasiliensis</i> ; <i>Mugil curema</i> ; <i>Mugil liza</i> ; <i>Mugilidae platanus</i> ; <i>Netuma barba</i> ; <i>Odontesthes bonairensis</i> ; <i>Orthopristis ruber</i> ; <i>Pseudoperis numida</i> ; <i>Sphoeroides testudineus</i> ; <i>Stellifer rastrifer</i> ; <i>Trichiurus lepturus</i>
MARINE MAMMALS: <i>Pontoporia blainvillei</i> ; <i>Sotalia fluviatilis</i>

NORTHEAST

São Marcos Bay, Mearim and Pindaré rivers were found uncontaminated by Hg (Cavalcante *et al.*, 1990. In: Hacon *et al.* (1990)). The specimens of *M. falcata* and *C. rhizophorae* showed concentrations of $<0.02-0.04 \mu\text{gHg.g}^{-1}$ and $<0.02 \mu\text{gHg.g}^{-1}$ respectively, being compatible with previous studies made by Juras (1988).

Meyer (1996) and Meyer *et al.* (1998) used oysters from Santa Cruz Channel *C. rhizophorae* as bioindicator, aiming to know the physiological responses of the oyster to Hg contamination and depuration through a transplantation experiment, revealing the capacity of the species to rapidly adapt to new environmental conditions, reflecting satisfactorily the Hg levels. Concentrations in the oysters remained between $0.27-2.21 \mu\text{gHg.g}^{-1}$ along the whole study. Costa *et al.* (1999) and Sant'Anna *et al.* (2000a and b) assessed the possible risk of Hg contamination to which a coastal human population could be exposed through fish consumption. *Mugil* spp. collected at this area presented total Hg and MHg of $0.027 \pm 0.026 \mu\text{gHg.g}^{-1}$ and $0.020 \pm 0.016 \mu\text{gHg.g}^{-1}$ respectively (N=60).

Moreira and Amaral (1990) investigated the Hg concentration estuarine bivalves from the Mandaú-Manguaba lagoon. *M. falcata*, a valuable local economic resource had average Hg concentration in the soft tissue of $0.015 \mu\text{gHg.g}^{-1}$ ww in the wet season and $0.025 \mu\text{gHg.g}^{-1}$ ww in the dry season. Le Campion (1992) investigated *Tagelus plebeius* from the Mundaú-Manguaba lagoons. The concentrations of Hg, were near the detection limit.

Tavares (1982), used bivalves (*Pitar fulminata* and *Anomalocardia brasiliana*) as biomonitors of Todos os Santos Bay. The Hg concentrations in tissues of these

bivalves were below the USEPA. limit of $1 \mu\text{gHg.g}^{-1}$ ww. A study on the inhibition of the embryonic development of *C. rhizophorae* by Cu, Zn, Pd, Cd and Hg, also in Todos os Santos Bay (Nascimento, 1982) calls attention to the fact that estuarine and coastal larvae live in the bay, directly exposed to metal pollution, compromising the recruitment of the species. Abnormalities were observed at different concentrations of Hg and other metals. Santos (1982) made a comparative study of different methods of preservation and mineralization of soft bivalves tissues for Hg analysis.

SOUTHEAST

Jesus *et al.* (2000) investigated Hg concentrations in Vitória Island estuary using *Perna perna*, *M. guyanensis* and *C. rhizophorae* as bioindicators.

In Guanabara bay (GB) *Perna perna* was used as bioindicators for Hg contamination during 1988 (N=55) (Pinto *et al.*, 1990. In: Hacon *et al.* (1990)). Both sampling stations used are harvested by mussel pickers for their living and for sale to restaurants in Rio de Janeiro. The concentrations were $0.014-0.049 \mu\text{gHg.g}^{-1}$.

Kehrig (1992) compared the total Hg levels in *M. furnieri*. 224 individuals were sampled in three estuaries in Rio de Janeiro (GB (N=61), Sepetiba Bay (N=63), Ilha Grande Bay (N=57)) and in Conceição Lagoon (N=43), from 1990 to 1991. Concentrations ranged from $0.014-0.43 \mu\text{gHg.g}^{-1}$ ww. *Macrodon ancylodon*, a herbivore, also consumed by the local population in Sepetiba Bay ranged from $0.010-0.13 \mu\text{gHg.g}^{-1}$ ww in the autumn 1991. Both species presented direct and significant relationships between total length, weight, and total Hg in their tissues. It was also possible to observe seasonal variations, showing that the physico-chemical characteristics of the water have an influence on the assimilation of the metal (Kehrig *et al.*, 1997; Kehrig *et al.*, 1998).

Mauro *et al.* (1997) determined the Methylation potential in GB, which was rated as low. The total Hg levels were between $0.017-0.27 \mu\text{gHg.g}^{-1}$ ww (MHg = 93%) for *M. furnieri* (N=61). *M. americanus* (N=19) showed levels between $0.010-0.13 \mu\text{gHg.g}^{-1}$ ww and *P. perna* (N=31) $0.014-0.048 \mu\text{gHg.g}^{-1}$ ww, three times less than total Hg in the fish muscle. *P. brasiliensis* (N=10) presented total Hg concentrations in the range of $<0.003-0.005 \mu\text{gHg.g}^{-1}$ ww. These levels were considered inexpressive regarding human consumption, provided future environmental conditions remain the same.

Comparisons of total Hg results in *Perna perna* from GB showed no significant difference between 1988 and 1998, and mussels remained safe for human consumption, in respect to Hg (Costa *et al.*, 2000).

Comparing different trophic levels Kehrig *et al.* (2000) used 291 specimens of *M. furnieri*, *M. liza* and *P. perna* were collected in different periods between 1988 and 1998 at GB. All presented low total Hg and MHg concentrations, below the maximum permitted limit. *M. furnieri* showed higher total and MHg concentrations. There was a significant difference in the M:total Hg ratio between the species: carnivorous fish presented higher MHg percentage (98%) than detritivorous fish (54%) and

bivalves (33%). Biomagnification of the organic form of Hg is probably occurring in the food chain.

In Kehrig *et al.* (2001), a total of 101 specimens *M. furnieri* and *M. liza*, 120 individuals of *P. perna* and water samples were collected in different periods between 1990 and 2000 at GB. The total Hg and MHg contents of mussel varied according to the sampling point and the water quality. MHg concentration in *M. liza* was similar to MHg concentration in *P. perna*. The total Hg concentration in *M. liza* tissue was lower than in *P. perna*. The bioaccumulation factor observed in relation to the total Hg accumulation by the bivalves (N=40) from their surrounding water, was in an order of 10^3 . Dissolved Hg concentration ranged from 0.72-5.23 ngHg.l⁻¹.

Kehrig *et al.* (2002) compared total and MHg in fish and mussels sampled in GB from 1990 to 2000. Longer time series of Hg data in bioindicators are better reflecting the metal's behaviour along the food chain and Methylation risks.

Pinho (1998) investigated total Hg levels in fish from 14 to 22° S. The Hg concentration in the muscle of 55 fish of 4 species (*Katsuwonus pelamis*, *Lopholatilus villarii*, *Pseudopersis numida* and *Trichiurus lepturus*) had an average concentration of $0.27 \pm 0.37 \mu\text{gHg.g}^{-1}$ ww, significantly lower than the average value for sharks and rays ($1.01 \pm 0.94 \mu\text{gHg.g}^{-1}$ ww). She analysed the edible muscle of 160 sharks (Pinho *et al.*, 1999). The species of *Mustelus*, which prey mostly on small invertebrates, had levels from 0.05-1.54 $\mu\text{gHg.g}^{-1}$ ww ($x=0.40 \pm 0.34 \mu\text{gHg.g}^{-1}$; N=92). *Galeocerdo cuvieri* which is a less selective feeder, had values from 0.17-0.25 $\mu\text{gHg.g}^{-1}$ ($x=0.21 \pm 0.04 \mu\text{gHg.g}^{-1}$; N=3). Three species of *Carcharhinus* and two species of *Squalus*, which are mainly fish feeders, had concentrations from 0.32-2.57 $\mu\text{gHg.g}^{-1}$ ($x=1.37 \pm 0.74 \mu\text{gHg.g}^{-1}$; N=9) and 0.34-4.06 $\mu\text{gHg.g}^{-1}$ ($x=2.07 \pm 0.68 \mu\text{gHg.g}^{-1}$; N=55).

Morales-Aizpurúa *et al.* (1999) analysed the total Hg contents in 26 individuals *Squatina argentina*, *Prionace glauca*, *Sphyrna* sp., *Odontaspis* sp. and *Isurus* sp. from São Paulo. The levels ranged from 0.04-4.71 $\mu\text{gHg.g}^{-1}$, 54 % of them above the new Brazilian legal acceptable limit for human consumption of 1.00 $\mu\text{gHg.g}^{-1}$ for predatory fish (Brasil, 1998).

Lacerda *et al.* (2000) show Hg concentrations in three shark species (*Rhizoprionodon lalandei*, *R. porosus* and *Mustelus higmani*) from the South eastern Brazilian coast. The total Hg values ranged from 0.0008-0.28 $\mu\text{gHg.g}^{-1}$ d.w., were considered low compared to values reported for shark species from the Southwestern Atlantic.

Moreira and Pinto (1989) and Moreira and Pinto (1991) used five different fish species from GB for determine totalHg. They choose the fish species based on their feeding habits and consumption by the local population. All the samples presented total Hg concentration below 0.26 $\mu\text{gHg.g}^{-1}$ ww Only two of the fish species studied presented a direct relationship between total length and totalHg content. Moreira and Pinto (1991) used *M. furnieri*, *Orthopristis ruber* and a crab (*Callinectes* sp.) based on the same criteria. *O. ruber* Hg concentration in the muscle tissue (N=22) ranged from 0.083-0.32 $\mu\text{gHg.g}^{-1}$

¹ ww and; *M. furnieri* (N= 23) varied from 0.023-0.20 $\mu\text{gHg.g}^{-1}$ ww. *Callinectes* sp. (N=10) showed concentrations of 0.035-0.066 $\mu\text{gHg.g}^{-1}$ ww *M. furnieri* was the only species to show a relationship between Hg concentration and age.

Eysink (1990) and Boldrini (1990) (In: Hacon *et al.* (1990)) focus on São Paulo littoral, and result from a long term study. Hg was present in the physical compartments within the minimum criteria for the preservation of aquatic life (0.05 $\mu\text{gHg.L}^{-1}$ and 0.2 $\mu\text{gHg.L}^{-1}$), in spite of eventual high concentrations in the biota. Amongst the fish, prawns, oysters and crabs analysed none presented Hg levels systematically above 0.50 $\mu\text{gHg.g}^{-1}$. Only carnivorous fish were unfit for human consumption. Slightly higher values were detected in the muscle and stomach contents of *Sphoeroides testudineus* (0.40 and 0.12 $\mu\text{gHg.g}^{-1}$) and *Centropomus* spp. (0.34 and 0.10 $\mu\text{gHg.g}^{-1}$). The species of economic interest in the estuarine region of Iguape and Cananéia were *Lycengraulis grossidens*, *Anchoviella lepidentostoli*, *Mugil curema* and *Mugil liza*. The highest Hg content amongst these was detected in the muscle of *L. grossidens* (0.26 $\mu\text{gHg.g}^{-1}$), and in the others the 0.50 $\mu\text{gHg.g}^{-1}$ maximum limit was never exceeded.

Boldrini (1990) studied Hg contamination along Santos coast, where the Cubatão industrial estate is. In 1975 the first evaluation, aiming to detect possible risks to human health, used fish, mollusks and crustacea. The maximum value was 0.80 $\mu\text{gHg.g}^{-1}$, and 16.7% of the samples were above the recommended 0.50 $\mu\text{gHg.g}^{-1}$ limit. A monitoring program was undertaken until 1980. High levels of Hg were observed in the stomach contents of *Mugil liza* (0.98 $\mu\text{gHg.g}^{-1}$), *Achirus* sp. (0.56 $\mu\text{gHg.g}^{-1}$) and in the muscle tissue of *Eucinostomus gula* (0.73 $\mu\text{gHg.g}^{-1}$) from Santos and São Vicente estuaries. In Santos Bay high levels were observed in all sampling campaigns for the species *Arius spixii* (up to 1.00 $\mu\text{gHg.g}^{-1}$), *Micropogonias furnieri* (up to 4.80 $\mu\text{gHg.g}^{-1}$), *Stellifer rastrifer* (up to 0.90 $\mu\text{gHg.g}^{-1}$) and *Netuma barba* (up to 1.00 $\mu\text{gHg.g}^{-1}$). These high levels were more frequently observed in the stomach contents of the fish. A significant Hg bioconcentration factors was observed in muscles (444 to 5611 fold) of *Caranx* sp., *Centropomus undecimalis*, *Eucinostomus gula*, *Chaetodipterus faber*, *Bagre bagre*, *Arius spixii*, *Netuma barba*, *Trichiurus lepturus*, *Cynoscion virescens* and *M. furnieri*; and in the stomach contents (167 to 5944 fold) of *Caranx* sp., *Mugil brasiliensis*, *Mugil curema*, *Bagre bagre*, *Arius spixii*, *Netuna barba*, *Achirus* sp., *M. furnieri* and *Stellifer rastrifer*. The crab *Callinectes danae* also had occasional high Hg contents 0.69-0.76 $\mu\text{gHg.g}^{-1}$, reflecting the environmental Hg contamination at their detritivorous feeding habit. Oyster and mussel samples were below the maximum permitted concentration for human consumption. Leafs of the mangrove trees were evaluated for retention from the contaminated sediments. The values were between 0.0002-0.42 $\mu\text{gHg.g}^{-1}$ for *Rhizophorae mangle*; 0.0003-0.16 $\mu\text{gHg.g}^{-1}$ for *Laguncularia racemosa* and; 0.001-0.54 $\mu\text{gHg.g}^{-1}$ for *Avicenia schaueriana*. In comparison with mangrove leaves from non-contaminated areas, this region was

found to accumulate Hg up to 105 fold. CETESB intensified the biomonitoring efforts in 1983, starting to include the Cubatão River. For these samples specifically, none was above the maximum permitted concentration of $0.5 \mu\text{gHg.g}^{-1}$ for human consumption.

Di Benedetto *et al.* (2000) investigated Hg contamination in marine mammals, *Pontoporia blainvillei* and *Sotalia fluviatilis*. *P. blainvillei* (N=7) presented Hg levels of $0.02\text{-}0.27 \mu\text{gHg.g}^{-1}$ ww in their muscle tissue and $0.12\text{-}2.7 \mu\text{gHg.g}^{-1}$ ww in the liver tissue. Female *S. fluviatilis* (N=3) presented Hg levels of $0.32\text{-}1.40 \mu\text{gHg.g}^{-1}$ ww in the muscle tissue and $1.1\text{-}15.8 \mu\text{gHg.g}^{-1}$ ww in the liver tissue. Male (N=5) presented Hg levels of $0.31\text{-}0.91 \mu\text{gHg.g}^{-1}$ ww in the muscle tissue and $1.1\text{-}21.7 \mu\text{gHg.g}^{-1}$ ww in the liver tissue. This species presented a high environmental sensibility to Hg contamination. Both have restricted migrating habits, and occupy the highest trophic level in the food chain around the estuary of Paraíba do Sul River. *Trichiurus lepturus* which is preferably preyed by *S. fluviatilis* was also investigated.

Lailson-Brito *et al.* (2001) analysed total and MHg in liver, kidney and muscles of *Sotalia fluviatilis* from GB. The liver presented the highest concentrations of total Hg, $0.22\text{-}132.62 \mu\text{gHg.g}^{-1}$ ww. The muscle presented the highest ratios of MHg (100 %) and the liver the lowest (3.9%). The cetacean species presented direct and significant relationships between the total length and the total Hg content in their liver tissues.

SOUTH

Seeliger and Knak (1982) approach the combined Hg and copper contamination of the Patos Lagoon using *Enteromorpha* sp.. Their hoped that continuous, frequent and concomitant monitoring of Hg levels in the water and algal tissues could eventually establish a higher confidence on the biomonitor response to the water quality changes, and that future monitoring could be satisfied by analysis of algae tissues alone. The area, industrial district and superport, is also breeding and nursery ground for the coastal fauna and represents a significant percentage of the national fishery resources which contamination by Hg would lead to biological and economic implications. Hg in *Enteromorpha* sp. tissues were significantly higher nearer Rio Grande. Concentrations of bioaccumulated Hg were found to be a linear function of total dissolved metal in the water column, but could be considered low, compared to other estuaries, reflecting low dissolved Hg concentrations in the water. The bioaccumulation and the toxicity of Hg in *Jenynsia lineata*, from Patos Lagoon, was investigated by La Reza (1983).

Ustra (2001), studied total Hg concentrations in fish and prawns from the southernmost part of the Patos Lagoon estuary. *Micropogonias furnieri* ($0.276 \pm 0.087 \text{ mgHg.kg}^{-1}$ ww), *Bagre* sp. ($0.0804 \pm 0.0054 \text{ mgHg.kg}^{-1}$ ww), and *Mugilidae platanus* ($0.0267 \pm 0.0007 \text{ mgHg.kg}^{-1}$ ww) were investigated. *Farfantepenaeus paulensis* showed concentrations of $0.028 \pm 0.013 \text{ mgHg.kg}^{-1}$ ww.

Niencheski *et al.* (2001) studied Hg in *M. furnieri* and *Odontesthes bonairensis* from the Patos and Mirim

Lagoons. Hg levels were below $200 \text{ ngHg.g}^{-1}\text{ww}$. Specimens collected near Rio Grande have concentrations of Hg above the natural levels. MHg contents were always above 50%, and a significant correlation was found between the fish length and its Hg content.

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