Research Articles

Total and Methylmercury Levels of a Coastal Human Population and of Fish from the Brazilian Northeast *

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Abstract. A descriptive exposure survey was conduced at Santa Cruz Channel, Pernambuco, Brazil, to assess the possible risk of mercury contamination to which a coastal human population could be exposed through fish consumption. The area was formerly known to be contaminated due to a chlor-alkali plant installed along one of the tributaries of the channel. The values for total and methylmercury were measured in human hair from inhabitants of Itapissuma $(1.9 \pm 1.6 \text{ ng THg/mg and } 1.2 \pm 1.0 \text{ ng MHg/mg} - mean values), a coastal fishing village, and fish ($ *Mugil sp.* $) catch in the area (<math>26.9 \pm 26.1 \text{ ng THg/g}$ and $19.6 \pm 16.0 \text{ ng MHg/g} -$ mean values). Both human hair and fish muscle presented relatively low values for total and methylmercury concentrations. These were found to be well within the average when compared to other Brazilian coastal environments.

Keywords: Brazilian Northeast; fish consumption; human hair; mercury contamination; methylmercury; *Mugil sp.*; Santa Cruz Channel; total mercury

Introduction

Santa Cruz Channel is a 'U' shaped coastal feature, sheltered and shallow, separated from the Atlantic Ocean by Itamaracá Island, 40 km north of Recife, in Northeastern Brazil. The channel has about 22 km of extension and an average depth in the order of a few meters. It extends from 07°40'S to 07°40'S and 34°45'W to 35°00'W. The whole area is bordered by highly productive mangrove swamps and fisheries of both fish and shellfish is abundant in both catch or man-made ponds. The channel has a number of tributaries among small creeks and larger rivers. The largest of them is Botafogo River, which arrives at the northern part of the channel (Fig. 1).

During the last decade, the native communities along Santa Cruz Channel have suffered sudden and severe changes in their ways of relating to nature, caused by unplanned development, irrational land occupation and a resulting loss of environmental quality. Traditionally, the area was occupied by sugar cane and coconut plantations, associated to other subsistence cultures and artisanal fisheries. Presently, the



Fig. 1: Schematic view of Botafogo River, Itapessuma City (human hair samples) and Canal de Santa Cruz (fish samples) in Pernambuco, Northeast Brazil

occupation of the land has been altered mainly through seasonal tourism, in response to the natural features of this region. Heavy industrialisation of the mainland municipalities upstream to the channel's tributaries has also contributed significantly to the decaying scenario. On top of that, unusually elevated population growth rates started to be registered in the areas along the channel. The rates ranged from 3.2 up to 4.2% p.a., being the highest of Recife's metropolitan area in the 90's (Lima & Quinamo 1998).

A chlor-alkali plant using the traditional electrodes method has operated a few kilometres up Botafogo River since 1963. Since

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1986, operations have been combined with a precipitation basin, which only started operating at its technically designated efficiency in 1991. The total mercury input to the channel up to 1987 may have been on the order of 22 to 35 tons (Meyer 1996). The present input is estimated at 0.03 ton/p.a. This plant, together with its associated industries, is also responsible for the dumping of large amounts of chlorine in the river.

The presence of the plant has been the reason for the spreading of unfounded rumours about the contamination of Santa Cruz Channel with mercury. Two surveys were then made by the São Paulo State Environmental Agency (CETESB 1981 and 1984 in Meyer 1996), contracted by Pernambuco State's Government, and one assessment was made by the Pernambuco State Environmental Agency (CPRH 1982 in Meyer 1996) and resulted inconclusive. None of those works were made available to the public or to the local authorities. Little or no attention was paid to the combination of other discharges, and the resulting, uncommon physicochemical conditions in which the inorganic mercury had been lost by the plant (Meyer 1996).

An extensive work by Meyer was performed in 1994/95 on the contamination status of Santa Cruz Channel concerning the particulate matter, the oysters and the sediment. At the time, she undoubtedly characterised Botafogo River and its vicinities as the areas most significantly polluted by mercury. On a tentative mass balance for determining the fate of mercury in Santa Cruz Channel, she hypothesises that most of the mercury released by the plant has not settled in the mangrove system, but has been exported to the sea. The present amount in the sediments of the channel is estimated at 1.2 to 2.5 tons of mercury.

The aim of the present work is to assess if some of the mercury remaining in the sediments and particulate matter which passes through the channel, could be assimilated by the biota and consequently put to risk the human populations who depend on the local fisheries for their nutrition and survival.

1 Material and Methods

Sampling was conducted during two months: August and September 1999. Hair samples from 8 men and 84 women was sampled at the Health Centre in Itapissuma. This Health Centre has been traditionally dedicated to the fishermen and their families. Fishermen themselves only look for medical care when in an emergency situation. Most of the patients who attend the Health Centre are women, children and elderly people. Also, a questionnaire about the subject's eating habits in respect to seafood and social issues, as well as their former and present occupations, was filled in.

The fish chosen for this survey was *Mugil curema* (Menezez and Figueiredo 1985), a genus (*Mugil*) which is known on the whole Brazilian coast under the generic name of 'tainha'. The sample (60 adult individuals) from Santa Cruz Channel was collected with the help of local fishermen. This fish is widely consumed in the region due to its low price, \$ 2.00 US or less per kilogram. The better quality and higher priced catch is usually reserved for sale to local restaurants and in Recife. This gender has a herbivorous feeding habit. Measurements of the total length were made and the left lateral muscle was taken for analysis accordingly to the recommendations of FAO (FAO 1983) and the NS&T Program (NOAA 1987). Sub-samples were individually frozen at -18°C. Transportation of the fish samples to Japan was made in dry ice.

The total and MHg analyses were performed at the National Institute for Minamata Disease (NIMD), in Minamata, Japan, following the procedures developed at this institute which are fully described in Akagi (1997). Human hair and fish samples were finely chopped before analysis. The analyses were made alongside with determinations of THg and MHg in CRMs. An IAEA 142 was used (total and methylmercury; mussel homogenate). CRMs for human hair were not available for this work, but the NIMD laboratory has taken part in a number of international calibration exercises in which its method and performance for total and methylmercury analysis in human hair has been tested and approved.

2 Results

The IAEA 142 CRM (mussel homogenate) analysis made during this work had an average of 113.3 ± 3.08 ng THg/g and 37.0 ± 0.7 ng MHg/g. This is 10% and 21% below the certified values (126 ± 7 ng THg/g and 47 ± 4 ng MHg/g, respectively), but still acceptable as a quality control reference value.

The results of human hair analysis appear in Fig. 2 a and b. The maximum, minimum and average values found for this



Table 1: Summary of the results for the human hair analysis (N = 93)

	ng THg/mg	ng MHg/mg	% MHg
range	0.1-12.5	0.1-6.0	13.0-100
average	1.9	1.2	70.2
st dev	1.6	1.0	29.9

analysis are listed in Table 1. In spite of the maximum value of the range for THg being 12.5 ng Hg/mg, only two of the values (12.5 and 7.3 ng Hg/mg), corresponding to approximately 2% of the sampled population, were found to be above 6.0 ng Hg/mg. Even then, these two individuals presented low MHg concentrations in their hair (1.6 and 6.0 ng Hg/mg, respectively).

The results for the fish sample analysis are shown in Fig. 3 a and b. The maximum, minimum and average values obtained during this analysis are shown in Table 2. All of the analysed individuals had both THg and MHg levels well below the maximum concentration permitted by the World Health Organization (WHO 1994) and by the Brazilian regulation regarding human consumption (500 ng Hg/g). Only two individuals presented an MHg% below 50%.

cury could also have been transferred from the water and sediments to other compartments of the environment, like the atmosphere and biota. The biotic compartment, including man, is the one of biggest concerns. Mercury analysis in edible parts of fish and human hair samples is widely recognised in the literature as important tools in the diagnosis of environmental mercury contamination (WHO 1994). The present work has then assessed the levels of mercury in two sub-compartments which are closely related. The fish chosen is related to the sediments through its detritivorous feeding habits. The local population is its direct consumer.

The values found in human hair from residents of Itapissuma (0.1-12.5 ng THg/mg) were within the national average of reported control populations (Fig. 4). It may also have been the case for MHg results in hair samples (0.1-6.0 ng Mhg/mg), although this result is more difficult to confirm due to the lack of data on MHg for control populations. The high average of the MHg% in relation to THg for this population (70.2 \pm 29.9%) suggests that they are exposed to mercury contamination mainly via fish/shellfish consumption.

Correlations between the mercury hair content and the eating habits declared in the questionnaires resulted inconclu-



Fig. 3 a and b: Total and MHg concentrations, and % of MHg in fish (Mugil curema) from Santa Cruz Channel, Pernambuco, Brazil, in August 1999

Table 2: Summary of the results for fish (Mugil curema) muscle analysis (N = 60)

	Length (cm)	ng THg/g	ng MHg/g	% MHg
range	27.2-36.6	4.6 - 167.0	1.8 - 91.6	5.7-100
avg.	30.6	26.9	19.6	80.0
st dev.	1.9	26.1	16.0	23.8

3 Discussion

The work from Meyer (1996), was a survey of the level of contamination of Santa Cruz Channel by inorganic mercury based on sediments and oysters total mercury contents. She has also produced a mass balance of the mercury in the channel, considering the total amount lost by the chlor-alkali plant in the last 35 years of its operation and the amount found in her survey. She concluded that less than 10% of the total mercury amount remains in the channel sediments today.

The rest of the mercury would have been exported to the sea in association with suspended solids. Part of the mer-



Fig. 4: Comparison between the THg values found in human hair during this work and control populations of other works. 1=the present work; 2=Vasconcellos, personal communication; 3=Vasconcellos et al., 1994; 4=Abe et al., 1995; 5=Nogueira et al., 1997; 6=Camara et al., 1998; 7=Camara et al., 1986; 8=Carvalho et al., 1985; 9=Mazzilli & Munita, 1986



Fig. 5: Comparison between the THg values found in fish muscle during this work and other Brazilian estuaries. 1=the present work; 2=Kehrig et al., 1998. A=Guanabara Bay (RJ); B=Sepetiba Bay (RJ); C=IIha Grande Bay (RJ); D=Conceição Lagoon (SC)

sive. There were no correlations between the different groups (A=eats fish/shellfish weekly; B=eats fish/shellfish monthly; C=eats fish/shellfish occasionally) and their respective total or methylmercury in hair. Due to the very humble social condition of the people interviewed, we suspect that the questionnaire might have induced the subjects to demonstrate an excessive preference for seafood. Also, it was clear that this population does not depend exclusively on this source of animal protein as was common in the past. Many have reported a quite diverse diet. Such could be the result of both a shift in their traditional occupations and the consumer market induction towards beef and poultry consumption from supermarkets.

The mercury concentrations in fish muscle from the channel (4.6-167.0 ng THg/g w.w.) were below the average concentration for other highly to moderately polluted Brazilian estuaries (Fig. 5). The four other estuaries pictured in Fig. 5 (Guanabara Bay, Sepetiba Bay, Ilha Grande Bay and Conceição Lagoon) have been investigated based on the total mercury concentrations in muscle of *Micropogonias furnieri*, a carnivorous fish known locally as 'corvina' (Kehrig et al. 1998). In spite of the differences between the fish and the environmental conditions, it is possible to consider that the levels in fish from Santa Cruz Channel are low. However, due to the recent past history of the place, it is unlikely that these concentrations are still at a background level.

The only existing report of THg in fish made in Santa Cruz Channel/Botafogo River are from whole fish samples:

- 0.08-1.30 ppm (53% samples >0.05ppm) 'various species' (CETESB 1981, in Meyer 1996);
- ~0.09 ppm w.w. (*Lile piquitinga*) and ~0.08 ppm w.w. (*Anchovia clupeidis*) (Meyer 1996).

The lower values found by these authors might be explained by the fact that they used different species of fish, with smaller sizes and different feeding habits. MHg reports on values for fish and shellfish from tropical/ sub-tropical coastal areas are rare in the literature (Kannan et al. 1998). The MHg% contents of this sample ($80.0 \pm 23.8\%$) seems to behave similarly in fish from tropical freshwater environments (Kehrig and Malm 1999), being typically higher than 50%. This behaviour is also observed for coastal fish species in which the relationship between THg and MHg stays around 80 to 100% (Kannan et al. 1998). The MHg% levels are also in agreement with the herbivorous feeding habits of *Mugil sp*.

The importance of these findings rests not only on the information as such, but also in the fact that it is providing a clarification for the long-standing social and economic issue. For many years, the channel was believed to be contaminated with mercury at such a level that the consumption of its fish and shellfish would put residents and other consumer markets at risk. Such might have been true in the past, but there was no survey of the fish or shellfish contamination status at the time, in spite of the public concern. This work is the first attempt to assess this risk. Although quite preliminary, this assessment already made it possible to believe that there is no immediate risk to the population through the consumption of fish from Santa Cruz Channel. The diminished, present risk can be attributed to a number of causes, and not necessarily to the measures taken by the plant to clean its discharges. Among them, the rapid decline of the local artisanal fisheries in the last ten years or due to the impoverishment of environmental conditions and the male occupational shift towards tourism and industry; the improvement of fishermen's and their families' eating habits, with the introduction of other sources of animal protein; and also the efficient transport of the mercury to the sea by particulate matter. A deeper survey of the social and economic issues may confirm the two first hypothesis (Lima and Quinamo 1998).

Under the light of these results and some more which are still being collected, the issue of mercury contamination will be discussed with the community by the Federal University of Pernambuco/Oceanography Department researchers to inform and clarify their doubts, as well as to thank them for their collaboration in submitting to the hair sampling. Their complaints about physical discomfort will have to be further investigated by specialized medical doctors, but these are presently being attributed to occupational stress, due to the rough nature of the work in the mangroves (especially for women and children), and to other sorts of aquatic pollution.

4 Conclusions

- The present work was the first attempt to assess the level of total and methylmercury contamination to the human populations living around Santa Cruz Channel.
- The human subjects have shown normal levels of total and organic mercury content in their hair when compared to an average group of Brazilians.
- In the case of this population, it was not possible, using the available tools, to relate eating habits and mercury contents in hair.
- It was also the first work on speciation of mercury contents of the local fish and shellfish consumed by the local population and sold in the markets.
- The fish muscle mercury contents for both mercury species analysed was found to be within normal limits and safe for consumption in respect to mercury.
- This work will be followed by a feedback work with the local community, especially those fishermen and their families from whom the hair has been sampled.
- The plant from which mercury has found its way into the channel must not disregard the importance of monitoring the surrounding populations only because the levels presented by humans and one species of animal are relatively low. It is their moral responsibility with this community to ensure that the mercury contamination they caused is properly studied and understood.
- The tracing of mercury methylation and mobilization processes in this environment should still be investigated further.

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