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Temporal Evolution of Exposure to Mercury in Riverside Communities in the Tapajós Basin, from 1994 to 2010

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Abstract Our objective was to evaluate the temporal evolution of mercury exposure in two riverside communities, Barreiras and São Luiz do Tapajós, downstream of gold mining areas in the Tapajós basin, Brazilian Amazon. The quantification of mercury in hair sample was made by atomic absorption spectrophotometry in the period between 1994 and 2010. In São Luiz do Tapajós the mercury exposure varied, in log units, from the peak of 1.21 ± 0.03 μg/g in 1996 to 1.16 ± 0.07 μg/g in 2007. Mercury exposure in Barreiras varied, in log units, from 1.25 ± 0.04 μg/g in 1994 to 1 ± 0.03 μg/g in 2010, peaking in 1995 at 1.25 ± 0.06 μg/g. Total mercury concentration found in both communities had no statistical differences across the years ($p > 0.05$) and they were higher than non-mercury exposed communities in Brazil and in South America. We concluded that the mercury exposure in the Tapajós basin is more than regulatory levels or higher than the general population.

Keywords Mercury exposure · Tapajós basin · Amazon riverside communities · Environmental toxicology

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Mercury is widely distributed in the environment and the organic form, called methyl mercury, constitutes the most important contaminant agent for population dependent of daily fish consumption. A series of complex chemical transformations permit the existence of mercury in three oxidation states (Hg^0, Hg^{+1}, Hg^{+2}) in the environment. Methylation process is the more important step for mercury to enter in the food chain and to promote human contamination afterwards. In humans, methyl mercury is obtained mainly from sea foods and its absorption occurs in the gastrointestinal system (ATSDR 1999). Methyl mercury (MeHg) is a well known human neurotoxin. Several tragedies occurred related to high mercury exposure of human populations causing irreversible damage to the human nervous system.

In the Amazonian ecosystem, mercury contamination is associated to anthropic activities and natural sources. In the Tapajós river region, deforestation, gold-mining, erosion, and leaching processes have contributed to the contamination of aquatic environment and mercury bioaccumulation. Recent studies performed in this region have demonstrated high mercury concentration in muscle tissue and viscera from some consumed fish species (Vieira et al. 2011). Although the occupational exposure to mercury vapor has been reported in the Amazon, the main concern is still what happens through feeding (Lebel et al. 1997).

The analysis of mercury concentration in hair samples is widely used to provide a biological marker for mercury exposure by feeding and to monitor the dangers of mercury exposure. Based in the accidents in Minamata, Japan, and Iraqi, it is presently known that mercury levels in hair above 10 μg/g correspond to a 5 % risk for the appearance of MeHg effects (WHO 1990).

Previous studies in the Tapajós basin showed mercury concentration above the safety threshold for maternal and

child healthy (Pinheiro et al. 2008). Although it is well known the mercury exposure, it is unknown the consequences of mercury levels across the years. The purpose of the present paper was to evaluate the temporal evolution of mercury contamination in the Tapajós basin, Amazon, Brazil.

Materials and Methods

The samples for the present study comprised residents of two riverside communities, São Luiz do Tapajós and Barreiras, which are located in the Tapajós river basin, near Itaituba city, in the Southwest region of the state of Pará, Brazil. We included in the present study subjects aged above 16 years from which total mercury in hair samples was quantified between 1994 and 2010. São Luiz da Tapajós was studied in 1996 (7 males, 36 females), 1998 (4 males, 25 females), 2001 (9 males, 29 females), 2002 (19 males, 31 females), 2003 (8 males, 56 females), 2005 (13 males, 29 females), 2007 (27 males, 23 females). In São Luiz do Tapajós the male mean age was 38.5 ± 5.4 years-old and female mean age was 32 ± 5.1 years-old. Barreiras was studied in 1994 (16 males, 10 females), 1995 (27 males, 25 females), 1996 (25 males, 37 females), 1998 (23 males, 36 females), 2000 (15 males, 10 females), 2001 (16 males, 32 females), 2003 (6 males, 62 females), 2005 (13 males, 40 females), 2006 (13 males, 49 females), 2007 (13 males, 46 females), 2008 (19 males, 44 females), 2010 (23 males, 47 females). In Barreiras, the male mean age was 39.3 ± 9.3 years-old and female mean age was 33.5 ± 4.4 years-old.

Mercury analysis was performed using atomic absorption spectrophotometry. The head hair samples ranging from 0.1 to 1 g, obtained from 1994 to 1996, were analyzed in the Chemical Engineering Laboratory, Institute of Environmental Science and Technology, Yokohama National University (Yokohama, Japan). From 1997 to 2010, the analysis of head hair sample occurred in the Laboratory of Environmental and Human Toxicology, Tropical Medicine Nucleus, Federal University of Pará (Belém, Pará, Brazil). Both laboratories performed the analysis using the Mercury Analyzer SP-3D (Nippon Corporation, Tokyo, Japan) to quantify the total mercury head hair concentration. Mercury Analyzer HG-201 (Sanso Seisakusho, Tokyo, Japan) was used to do intercalibration test between laboratories equipment of analysis in the samples collected in 2008. Mercury concentration measurements from both laboratories were found to be compatible to the level of 95 % of correlation factor, $p < 0.01$. In these tests, twenty samples of head hair were analyzed in both equipments. For mercury analysis in the Mercury Analyzer HG-201, we used the Akagi's method as described in the manual of the Japanese Ministry of Environment (Suzuki et al. 2004). The precision and accuracy of these measurements were verified using the IAEA 085 International Reference Standard (>95 %).We excluded subjects with hair sample that were analyzed in other laboratories or those with insufficient information for the present study.

Information about age, time of local living, year of the investigation, and total mercury concentration in head hair sample were obtained from the medical record of each subject attended by researchers of the Tropical Medicine Nucleus during the annual visit to the region studied. These data were collected following the tenets of the Declaration of Helsinki, the resolution 196/96 of the Brazilian Healthy National Council and the approval of the Committee of Ethics in Research of the Tropical Medicine Nucleus.

Total mercury head hair concentration was log10-transformed and then the median and standard error of the data were calculated. For comparisons of total mercury head hair concentration across the years, we used one way ANOVA ($\alpha = 0.05$ %, Tukey's post hoc test).

Results and Discussion

In São Luiz do Tapajós, the median for total mercury head hair concentration varied from 1.21 ± 0.03 μg/g in 1996 to 1.16 ± 0.07 μg/g in 2007. The lowest median for mercury concentration was found in 2002 (0.86 ± 0.03 μg/g) while the largest occurred in 1996. The largest individual mercury concentration was found in 2007 (1.78 μg/g). Table 1 shows the median temporal evolution and also the first and third quartiles for total mercury head hair concentration in São Luiz do Tapajós from 1996 to 2007.

In Barreiras, the median for total mercury head hair concentration varied from 1.25 ± 0.04 μg/g in 1994 to 1 ± 0.03 μg/g in 2010. The lowest median for mercury concentration was found in 2008 (0.92 ± 0.04 μg/g) while

Table 1 Total mercury head hair concentration in the population of São Luiz do Tapajós between 1996 and 2007

Year	n	Median ± SE (μg/g)	Q1 (μg/g)	Q3 (μg/g)
1996	43	1.21 ± 0.03	1.08	1.44
1998	29	1.2 ± 0.04	1.07	1.38
2001	38	1.13 ± 0.05	0.82	1.27
2002	50	0.86 ± 0.03	0.76	0.94
2003	64	1.16 ± 0.03	0.99	1.27
2005	42	1.12 ± 0.04	0.95	1.28
2007	50	1.16 ± 0.07	0.73	1.43

n number of samples, *SE* standard error, *Q1* first quartile, *Q3* third quartile

the largest occurred in 1995 (1.25 μg/g ± 0.06 μg/g). The largest individual mercury concentration was found in 1996 and amounted to 1.85 μg/g. Table 2 shows the median temporal evolution and also the first and third quartiles for total mercury head hair concentration in Barreiras from 1994 to 2010.

Figure 1 shows the total mercury concentration across the years measured in both communities. The values for both communities oscillated around the safety threshold for mother and child healthy (1 μg/g in log units, dotted line). Figure 1A-C shows the distribution of mercury concentration in the population of each community for each year studied. There were no differences of mercury concentration across the years inside the same community and there was no differences between both communities considering the overall mean values ($p > 0.05$). In Barreiras and São Luz do Tapajós, along the years of investigation, respectively 58.12 % ± 17.8 and 64.3 % ± 21.8 of the hair samples had mercury concentration above the level of risk for pregnant and children (1 μg/g, in log units).

Figure 1d shows the temporal evolution of values obtained from two subjects living in each community. For these two subjects the mercury concentration remained above the critical values for safety and was stable during the time of study.

Table 3 shows the values for head hair mercury concentration obtained from some populations that were not exposed to mercury for comparison with the levels observed in the two communities of the Tapajós basin along the years covered by the current study.

Total mercury head hair concentration was used as an important marker of the exposure level in different environmental accidents involving the ingestion of MeHg contaminated fish (Soong et al. 1994. In the Amazon, mercury exposure occurs mainly in riverside communities near or in the zone of influence of gold mining activity (Pinheiro et al. 2007). Despite several studies that evaluated mercury exposure levels in Amazon communities, an approach to quantify how the exposure to mercury occurred across the years was missing. The present study evaluated the temporal evolution of the mercury exposure in two communities located in the Tapajós river basin, where most of the studies about mercury exposure in the Amazon were performed.

Table 2 Total mercury head hair concentration in the population of Barreiras between 1994 to 2010

Year	n	Median ± SE (μg/g)	Q1 (μg/g)	Q3 (μg/g)
1994	26	1.25 ± 0.04	1.11	1.36
1995	52	1.25 ± 0.06	0.9	1.4
1996	62	1.23 ± 0.03	1.02	1.39
1998	59	1.11 ± 0.04	1	1.29
2000	25	1.3 ± 0.06	1.12	1.43
2001	48	0.94 ± 0.04	0.76	1.11
2003	65	1.03 ± 0.04	0.81	1.26
2005	53	0.96 ± 0.05	0.76	1.28
2006	62	1 ± 0.04	0.8	1.18
2007	59	1 ± 0.05	0.8	1.17
2008	63	0.92 ± 0.04	0.72	1.04
2010	70	1 ± 0.03	0.85	1.2

n number of samples, *SE* standard error, *Q1* first quartile, *Q3* third quartile

In the beginning of the current study, 1994 in São Luiz do Tapajós and 1996 in Barreiras, the population of both communities had the mercury levels above 1.3 μg/g (log units) or 20 μg/g (absolute values). A slight reduction of median values was observed in the following years of the study period, but this decrement was not enough to be statistically significant, suggesting that both communities were highly exposed to mercury for at least 16 years.

Both communities had usually median values above 1 μg/g (log units). Based in the accidents occurred in Minamata, Japan, and in Iraq, it is known that a mercury concentration of 1 μg/g (log units), constitutes a risk to pregnant women (WHO 1990). Recently, it was observed neuropsychological impairment in adults exposed to low MeHg levels (around 0.62 μg/g in log units) associated to the habit of feeding fish in the Pantanal region of Brazil (Yokoo et al. 2003). Similar mercury levels were found by other investigations performed in riverside communities around the Tapajós and Madeira rivers (Passos and Mergler 2008). In other countries such as Canada and China, it was observed the same magnitude of mercury exposure, where the effects of long-term mercury exposure due feeding of MeHg contaminated fish determined the appearance of clinical symptoms of mercury intoxication (Soong et al. 1994).

It was reported that the general population is not at healthy risk evens when mercury concentration in hair reaches value as high as 1.69 μg/g (log units) or 50 μg/g (absolute values), but that there is a particular risk for fetuses to develop neurological disorders when maternal hair reaches such mercury levels (WHO 1990). After the interpretation of data originated from Iraqi and Minamata disasters, the World Health Organization considered that mercury levels of 1 μg/g (log units) of hair implied in 5 % of health risk (WHO 1990).

The World Health Organization reported that an even low level of mercury ingestion, for a long time, is able to cause neurobehavioral abnormalities such as lack of psychomotor coordination, previously to the appearance of generalized clinical symptoms (WHO 1990). The present study shows that prolonged times of mercury exposure, as well as the high total mercury concentration found in the

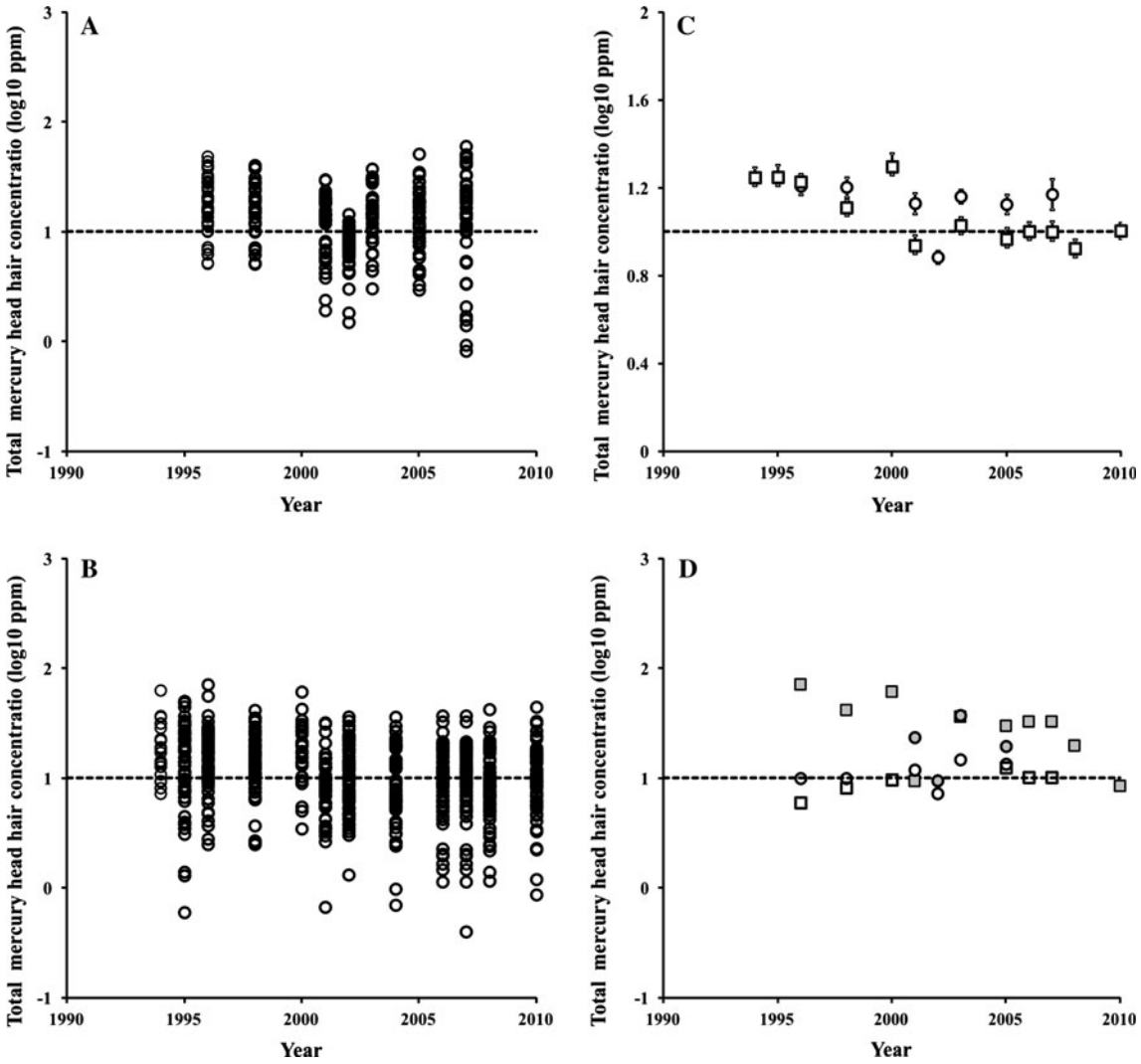

Fig. 1 Temporal evolution of total mercury concentration from 1994 to 2010 in two communities of the Tapajós river basin. Scatter plots of the total mercury concentration in the Tapajós communities from 1994 to 2010 in São Luiz do Tapajós (**a**) and Barreiras (**b**). It was observed that more than half of samples had mercury concentration higher than 1 μg/g (log units, *dotted line*). **c** Median (± SE) from São Luiz do Tapajós (*circles*) and Barreiras (*squares*). **d** Total mercury concentration for two subjects from Barreiras (*circles*) and São Luiz do Tapajós (*squares*) along the years of the current study. *Symbols* represent different subjects in **a**, **b**, and **d** as well as median for the whole population examined in **c**

communities of the Tapajós river basin, suggest that is necessary to keep these communities under health vigilance.

Accumulated MeHg in head hair represents 80 %–95 % of total mercury (National Academy of Science, 2000). Mercury concentrations in the head hair are proportional to those obtained from blood, but amplified times 250 (Cernichiari et al., 1995). Blood mercury concentration, particularly blood organic mercury concentration, reflects the frequency and amount of consumed fish as well as the mercury concentration in fish (Mahaffey and Mergle 1998). Although our purpose in the present study does not include the analysis of the amount of ingested fish, there are studies showing that riverside communities along the Tapajós river basin have a high ingestion of fish in the food (Lebel et al. 1997; Passos and Mergler 2008). In a study conducted in communities near São Luiz do Tapajós, it was observed that the population feeds mainly highly mercury contaminated piscivorous and omnivorous species during the raining season, while their diet was based in herbivorous fish species during the dry season (Lebel et al. 1997).

We collected hair samples during the raining season in the first 3 years of this project, but during the remained years we collected them during the dry season. Barreiras and São Luiz do Tapajós have some common features once they depend of the river for their biological survival, economy, and transportation (Pinheiro et al. 2001). Moreover, they live in poor health and hygiene conditions for

Table 3 Hair mercury concentration (log units) measured in non-exposed communities

Population	n	Mean (μg/g)	Min (μg/g)	Max (μg/g)	References
Caxiuanã, Pará, Brazil (Amazon river)	214	0.93	−0.22	1.65	
Lago Grande, Pará, Brazil	316	0.59	−0.39	1.06	
Santana do Ituqui, Pará, Brazil	321	0.63	−0.39	1.06	
Tabatinga, Pará, Brazil	499	0.72	0.43	1.23	
Pindobal grande, Pará, Brazil (Tocantins river)	43	0.51	−0.09	0.85	
Panacauera, Pará, Brazil (Tocantins river)	23	0.91	0.14	1.4	
Panacauera, Pará, Brazil – only women	20	0.55	0.11	0.77	Pinheiro et al. (2008)
Panacauera, Pará, Brazil – only children	36	0.45	−0.4	0.97	Pinheiro et al. (2007)
Barão do Melgaço, Mato Grosso, Brazil	114	0.31	0.42	0.88	
Cananéia, São Paulo, Brazil – only children	105	−0.31	−2	0.52	
San Jorge river basin, Colombia	94	0.69	–	–	Olivero et al. (2002)
Napo river valley, Coca community, Ecuador	45	0.27 (urban community)	−1.52	1	Webb et al. (2004)
Napo river valley, Añangu community, Ecuador	27	0.94 (rural community)	0.34	1.31	Webb et al. (2004)
Napo river valley, Pañacocha community, Ecuador	27	0.72 (rural community)	0.16	1.13	Webb et al. (2004)
Barreiras, Brazil		1.02	−0.39	1.85	Current study
São Luiz do Tapajós, Brazil		1.07	−0.08	1.78	Current study

n number of samples, *SE* standard error, *Q1* first quartile, *Q3* third quartile

the supply of drinking water and waste destination. Although São Luiz do Tapajós is nearer from gold-mining areas than Barreiras, the levels of mercury exposure across time are similar in both communities, suggesting that similar features such as preference and frequency of ingestion of piscivorous fish species and lacking of nutritional antioxidant compounds (fruits and nuts) would have influenced the exposure scenario. For instance, it has previously been shown that the consumption of fruits obtained in the region where these communities are located, has favorable effects to reduce the mercury levels in head hair samples (Lebel et al. 1997).

We concluded that mercury exposure in the Tapajós river basin occurring for a long time, from 1994 to 2010, has reached levels higher than the regulatory levels or higher than the general population (WHO 1990). Clinical and toxicological monitoring of this population as well as future studies involving other communities are recommended.

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