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Mercury in the environment and riverside population in the Madeira River Basin, Amazon, Brazil

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Abstract

This work presents quality control results on the mercury concentrations in different environmental (river sediments, forest soils, river suspended matter and fish) and human samples from the lower Madeira River, Amazon sampled between 2001 and 2003, about 15–20 years after the nearly cessation of gold mining activities in the region, which reached its peak in the late 1980s. The study aimed to compare mercury concentrations in these environmental samples with those reported by other authors during the gold rush of the Madeira River Basin. Today, in the Madeira River the releases of mercury register a sudden reduction due the gold price fall in the international trade. However, about 100 t of Hg were released to the atmosphere and to aquatic systems in the region during the gold rush. The present survey shows that notwithstanding the reduction of Hg emissions to the Madeira River Basin from gold mining proper, concentrations in fish and humans are similar to those measured during the gold rush. Reduction in Hg concentrations is restricted to areas close to old point sources and only for abiotic compartments (air and sediments). Remobilization of Hg from bottom sediments plus re-emission from soils due to land use changes are probably responsible for keeping high Hg concentrations in biological samples.

Keywords: Mercury; Riverside populations; Fish; Sediments; Madeira River; Amazon

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1. Introduction

The gold exploration in the Amazon region relied on mercury amalgamation to separate fine gold particles from riverbank deposits and bottom sediments. In the Amazon Basin, in particular, this technique represents a simple, cheap and reliable procedure for gold exploration which produced about 2000 t of gold from 1975 to 2002, leaving behind nearly 3000 t of Hg in the region's environment (Thornton et al., 1992; Lacerda and Salomons, 1998; Lacerda, 2003a,b). The Madeira River Basin was the second most important gold mining region in the Amazon. The gold rush in this basin started around 1975 as an individual operation at riverbanks and islands during the dry season. It was fallowed by small dredges operated by divers and latter, by large mechanical dredges able to operate at higher depths directly on the river bottom. The activity reached its maximum during the late 1980s, when over 1500 dredges were working along a 300 km stretch of the Madeira River, from Porto Velho to the Bolivian border at Guajará Mirin (Fig. 1). The total gold production reached about 9.4 t $year^{-1}$, with an estimated release of 12.4 t $year^{-1}$ of Hg. Between 1979 and 1990 about 87 t of Hg were emitted, about 65% to the atmosphere through amalgam burning and bullion smelting, and about 45% discharged directly to rivers as metallic Hg, during the mining operation (Pffeifer and Lacerda, 1988; Lacerda et al., 1995). Since most of the Hg emitted to the atmosphere is deposited about 40 km from the source, most of the emission was deposited in forest soils relatively close to sources (Lacerda et al., 2004). Although gold mining in the Brazilian portion of the Madeira River Basin has decreased significantly from 1995 onwards to about 0.3 to 0.5 t $year^{-1}$ nowadays, the activities continued and even increased in the Bolivian side of the basin, and the Hg released there eventually drains into the Madeira River from its major Bolivian tributaries (Maurice-Bourgoin et al., 2000). Also, deposited Hg in soils and sediments suffers continuous transformations and interactions with environmental compartments, which eventually transform and remobilize Hg to food chains, including the increase of its bioavailability through methylation (Malm et al., 1990; Roulet et al., 1998; Lacerda, 2003a). Therefore, even with decreasing direct emissions, Hg contamination of soils, sediments and aquatic biological resources, fish in particular, is still an environmental concern to local and national environmental authorities.

For the Amazon riverside populations, fish is their main protein source with daily consumption of up to 500 g of fish (Fabré and Gonzales, 1998). According to Boischio and Henshel (2000) the daily average fish consumption among the Madeira River riverside populations reaches 243 g. Therefore, even relatively low Hg concentration in fish may result in high exposure doses for these human groups.

The above-mentioned facts support a continuous monitoring of Hg in the environment of the Madeira River Basin. This study presents a diagnostic of the current levels of Hg contamination in the environmental compartments and human populations along Madeira River between Porto Velho City (Rondônia state) and its mouth at the Amazon River close to the city of Itacoatiara (Amazonas state) (Fig. 1).

2. Material and methods

2.1. Study area

The Madeira River is a tributary of the right margin of the Amazon River. It is 1459 km of extension, totally inside the Brazilian territory, running through Rondônia and Amazon states (Fig. 1). The river has an average flow of 23,000 m^3 s^{-1} (5000 to 45,000 m^3 s^{-1}), the twentieth larger river in the world (Sedam, 2002). The Madeira River Basin drains an area of 1,420,000 km^2, divided into two different sections. The upper Madeira River, with about 360 km between the confluence of the Mamoré and Beni rivers, originated in the Andean plateau, to Porto Velho city. Along this route the Madeira River cuts off rocks and pre-Cambrians and Cenozoic sediments, with rocky outbursts in its bed responsible for forming various waterfalls and rapids (Silveira et al., 2000). The lower Madeira River, our study area, is totally navigable for an extension of 1100 km, the most important waterway of South America, between Porto Velho (RO) and Itacoatiara (AM) cities.

According to Sioli (1967) the Madeira River is classified as a white-yellowish-water river, rich in dissolved material and suspension solids. Mortatti et al. (1989) and Guyot (1993) estimated total material transport of the order of 37×10^6 t $year^{-1}$ and 40×10^6 t $year^{-1}$, respectively, but it can reach 14×10^8 t $year^{-1}$ during the rainy season. The basin soils are quite variable, presenting latossols, argissols, neossols, gleissols and cambissols, with latosols dominating about 58% of the area (Sedam, 2002).

The riverside population of the Madeira River is predominantly composed of individuals with indigenous ancestry. In lower Madeira River, these communities amount nearly 135,000 inhabitants (IBGE, 2002)

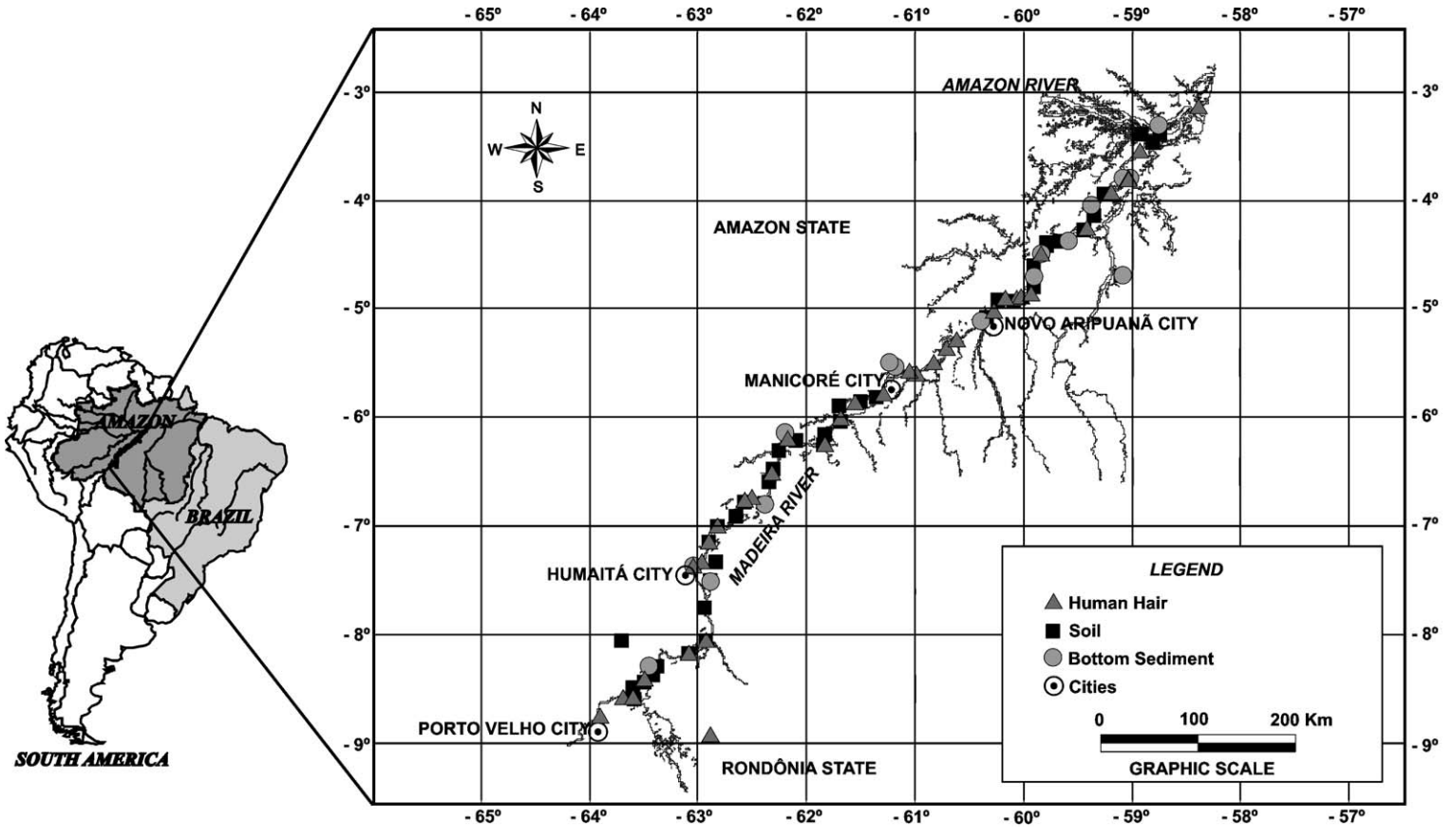

Fig. 1. Study area with collection points of the Madeira River between the Porto Velho City and the Amazon River (1100 km).

and have as major diet cassava flour and fish. They dedicate to agriculture, fishing and forest essences and fruits gathering (Correa, 2002).

2.2. Sampling and chemical analysis

The environmental sampling (bottom sediments and suspension solids, forest soils and fishes) and human samples (hair) was accomplished between 2001 and 2003 along the Madeira River between Porto Velho, RO and Itacoatiara, AM (Fig. 1).

Forest soils were collected within 2 km from the river margin using plastic shovels to remove the litter layer. After that, a 50 cm soil column was sampled and packed in plastic bags, identified and stored under refrigeration (<5 °C). In laboratory samples were sieved (74 μm; <200 mesh) prior to analysis.

Suspension solids and bottom sediments were collected in sedimentation areas of the Madeira River proper and marginal lakes that receive Madeira River waters during the rainy season. Suspension solids were vacuum-filtrated through cellulose membranes (0.45 μm of pore diameter) from 5 L polyethylene bottles whereas bottom sediments were collected with an Eckmam dredge. Bottom sediments were also sieved (74 μm <200 mesh) prior to analysis.

Fish samples were acquired in fish markets along the route and directly from local fishermen and inhabitants. In the markets only the most consumed species were selected and individuals of different sizes were sampled whenever possible. In laboratory, samples were identified, measured and a 50 to 100 g slice of the dorsal muscle of each individual was cut for analysis. Samples were kept frozen until analysis. Species identifications followed Santos et al. (1991).

The human hair samples (approximately 500 mg) were collected close to the scalp of the occipital area with the use of stainless steel scissors and kept in plastic bags at room temperature. To the volunteers of the research we applied questionnaires on alimentary habits in order to obtain details on their fish ingestion. In the laboratory the samples were washed with 20.0 mL of an EDTA 0.01% solution and dried at 40 °C.

The total Hg concentrations were quantified by atomic absorption spectrophotometer coupled with cold vapor generation FIMS-400 (Flow Injection Mercury System-Perkin Elmer). Details of sample digestion and analysis are described elsewhere (Bastos et al., 1998). Analytical control was accompanied by analysis of reagent blanks and reference certified samples (IAEA-356 and IAEA-085). Average recovery of reference standards reached 97%.

3. Results and discussion

Mercury concentrations in soils varied from 34.8 to 366.1 μg kg^{-1}, with average value of 106.9 ± 88.8 μg kg^{-1}. These values are in accordance to previous concentrations reported by Lacerda et al. (1995) and Malm et al. (1990) for the upper Madeira River, where Hg concentrations varied between 30.0 to 340.0 μg kg^{-1} and 35.0 to 300.0 μg kg^{-1}, respectively. Highest Hg concentrations (>200.0 μg kg^{-1}) occurred nearby Porto Velho City (Fig. 2) and are probably a legacy of the proximity of the large gold mining activity which existed in the area, as well as the emissions to the atmosphere from gold dealer shops, as suggested by previous results of Malm et al. (1990). High Hg concentrations in soils close to gold mining activities has also been reported by Lacerda et al. (1991) in their study in Poconé-MT region, Central Brazil, where Hg concentrations nearby the mining operations reached 270.0 μg kg^{-1}, whereas the outside the influence of such locations (about 70% of the samples) average Hg concentrations were about 30.0 μg kg^{-1}.

Downstream Porto Velho City, Hg concentrations in soil decreased to values <100.0 μg kg^{-1} to about 600 km toward the Madeira River confluence with the Amazon River. There was a considerable increase in Hg concentration in soil samples collected further than 600 km of Porto Velho City. These high Hg levels can be related to the larger deposition areas and flood plains that border the Madeira River from this portion to its mouth at the Amazon River, where soils receive alluvial sedimentation during the inundation period (Fig. 2). A similar behavior has been described by Leckler et al. (2000) for upland oxisols along the same portion of the Madeira River.

Mercury concentrations in bottom sediments are showed in Table 1. Concentrations measured in Madeira River bottom sediments varied from 32.5 to 82.2 μg

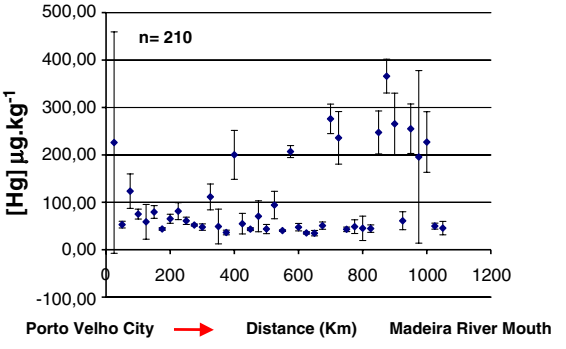

Fig. 2. Mercury concentrations in soils along the Madeira River between Porto Velho and Itacoatiara Cities (1100 km).

Table 1
Mercury concentration (μg kg^{-1}) bottom sediment from the Madeira River and marginal lakes

Local	Mean (μg kg^{-1})	SD	Min.	Max.
Cunã Lake	98.13	19.50	66.24	117.09
Puruzinho Lake	53.00	9.01	49.19	60.09
Paraíso Lake	49.21	12.87	25.99	71.79
Sto. Antônio Lake	60.34	9.00	46.91	74.74
Mergulhão Lake	90.64	15.90	73.02	109.02
Acará Lake	45.11	9.61	36.53	60.00
Matupirizinho Lake	59.93	19.36	35.14	99.06
Preto Lake	61.50	5.94	54.44	70.00
Madeira River	46.21	4.95	40.00	53.63

kg^{-1}, whereas in bottom sediments from marginal lakes Hg concentrations were higher and varied from 25.9 to 117.1 μg kg^{-1}. They are also similar to the concentrations reported by Leckler et al. (2000) who sampled along the same river section. There was no clear trend in Hg distribution in the Madeira River bottom sediments. The highest Hg concentrations found in bottom sediments of lakes are due to their hydrodynamic characteristics, being these reservoirs potential sites of fine-organic sedimentary deposition and consequent accumulation of Hg. Also, marginal lakes receive Hg-containing particles from their watershed, which may include soil-contaminated sites.

In suspension solids, Hg concentrations varied between 20.0 to 48.8 μg kg^{-1} in the Madeira River, whereas in marginal lakes they varied from 10.0 to 84.6 μg kg^{-1}, with significant seasonal suspension solids concentrations changes ranging from 200.0 mg L^{-1} (dry season) to 1115.0 mg L^{-1} (rainy season). The Hg concentrations in suspension solids showed the same distribution pattern as that of bottom sediments with higher and more variable concentrations found in marginal lakes relative to the Madeira River proper. Seasonal influences could bring Hg rich particles to the lake surface through bottom sediments remobilization, resulting in higher Hg content in the suspend solids in these lakes (Bonotto and Silveira, 2003).

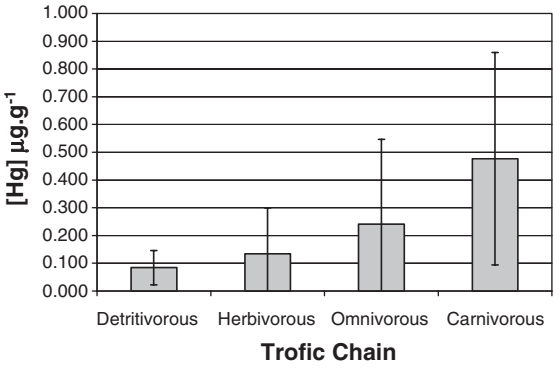

Fig. 3. Mercury concentrations in fish species of different alimentary habit.

Similar ranges of Hg concentrations in suspended matter and spatial behavior have also been described by Maurice-Bourgoin et al. (2000) for the Madeira River Basin.

The alimentary habits questionnaires answered by 660 people cited 32 fish species as locally consumed. However, only 8 fish species represent about 60% of the species consumed by riverside communities of the lower Madeira River: Pacú (*Mylossoma* sp.), with 23%, followed by Jaraqui (*Semaprochilodus ocellaris*) with 12%, Sardinha (*Triportheus albus*) with 10%, Curimatã (*Prochilodus* sp.) with 7%, Branquinha (*Curimata amazon*) with 9%, Tucunaré (*Cyclla* spp.) with 6%, Tambaqui (*Colossoma macropomum*) with 5% and Mandi (*Pimelodus* sp.) with 4% (Table 2). According to the questionnaires, fish meat consumption reached up to 250 g day^{-1} for adults and 150 g day^{-1} for children.

Of the 816 analyzed fish specimens 24% presented Hg concentrations higher than the maximum concentrations established by the World Health Organization (WHO) for human consumption (0.5 μg g^{-1}). In this group, particularly high Hg content was found in the carnivorous "piranha preta" (*Serrasalmus rhombeus*), "pescada" (*Plagioscion squamosissimus*), "surubim" (*Pseudoplatystoma fasciatum*), "tucunaré" (*Cicha* ssp.) and "barba-chata" (*Pinirampus pirinampu*).

Table 2
Fishes species of larger consumption rates in the riversides communities and Hg concentrations averages and standard deviations

Species (vulgar name)	Scientific name	Consumption (%)	[Hg] μg g^{-1}	SD	*n*
Pacú	*Mylossoma aureum*	23	0.046	0.039	23
Jaraqui	*Semaprochilodus theraponera*	12	0.099	0.068	57
Sardinha	*Triportheus albus*	10	0.231	0.252	31
Curimatã	*Prochilodus nigricans*	09	0.083	0.058	25
Branquinha	*Curimata amazônica*	07	0.095	0.045	21
Tucunaré	*Cyclla* ssp.	06	0.414	0.228	167
Tambaqui	*Colossoma macropomum*	05	0.099	0.085	26
Mandi	*Pimelodus* sp.	04	0.248	0.146	41

Among the omnivorous the "sauna" (Curimatidae) also presented Hg concentrations higher than the WHO limit (WHO, 1976). Mercury concentrations clearly varied according to fish alimentary habit with carnivorous species showing highest Hg content (Fig. 3) when compared the concentrations found for herbivorous and omnivorous species.

Out of the forty-five communities only one, the Tabocal farm community, where only two hair samples were obtained, presented average of Hg concentrations below what is considered by WHO as normal for human hair (<6.0 μg g^{-1}). In the other 44 communities the averages of Hg concentrations in human hair were higher than this limit. In 16 communities the average of Hg concentrations were higher than the limit considered by WHO (1976) to start toxicological symptoms (>16.0 μg g^{-1}) (Table 3). There was no direct relation between these high Hg concentrations and the fishes species

Table 3
Mercury concentrations (μg g^{-1}) in human hair of riverside population of the lower Madeira River

Local	Average	SD	Min.	Max.	*n*	Mediana
Calama-RO	9.02	5.78	0.50	22.48	34	8.93
Boa Vitória-RO	13.82	3.10	10.86	17.05	03	13.55
Cujubim-RO	6.30	4.00	1.55	14.67	12	6.65
Firmeza-RO	11.21	2.54	9.40	14.80	04	10.32
Itacoã-RO	11.97	4.33	5.28	16.00	06	13.93
Nazaré-RO	10.65	5.65	0.63	22.60	64	9.53
Papagaios-RO	13.72	7.71	4.76	27.22	13	12.48
Santa Rosa-RO	13.99	3.12	7.68	20.78	19	14.08
São Carlos-RO	9.51	6.36	1.84	22.83	15	8.23
Terra Caída-RO	9.61	3.61	5.01	14.61	07	10.34
Sto Ant.do Pau Queimado-RO	14.69	6.45	5.87	26.86	14	12.48
Puruzinho-AM	14.83	5.59	4.57	28.27	28	14.04
Livramento-AM	36.89	11.99	18.96	63.54	15	36.48
Valparaíso-AM	18.93	17.16	2.98	82.38	21	19.33
Auxiliadora-AM	9.34	6.72	1.12	22.78	34	5.94
Curralinho-AM	19.69	9.42	10.70	34.49	05	18.10
Nazaré do Retiro-AM	17.90	4.21	9.69	24.77	15	18.23
Novos Prazeres-AM	11.90	5.53	2.77	24.28	20	12.44
São Pedro-AM	15.77	6.76	6.61	28.00	14	15.96
Barreira do Manicoré-AM	10.82	6.89	1.45	23.04	09	11.97
Cachoeirinha-AM	14.74	8.77	1.54	37.22	14	12.11
São Lázaro-AM	9.48	8.10	2.50	23.37	06	6.72
Maracá II-AM	11.37	2.74	8.57	15.69	06	10.82
Vista Nova-AM	25.69	3.09	21.40	28.54	04	26.41
Vista Alegre-AM	16.02	5.19	7.28	26.28	17	16.26
Bom Suspiro-AM	16.29	7.39	6.43	30.06	12	15.59
Carará-AM	18.13	9.19	4.18	34.71	39	15.79
Miriti-AM	22.34	10.04	6.70	50.37	16	21.03
São Sebastião (Lago Lúcio)-AM	12.84	4.10	6.61	18.52	17	12.22
Boca do Carapanatuba-AM	10.45	4.76	3.43	19.22	18	9.89
São Sebastião do Tapuru-AM	62.76	30.76	20.43	150.00	18	60.60
Moanenses-AM	12.73	6.06	3.26	20.49	13	14.85
Três Casas-AM	33.07	23.21	5.62	70.70	09	24.26
Boa Ventura-AM	16.55	10.85	4.73	35.79	07	13.15
Auará Grande-AM	15.97	5.66	6.21	24.98	19	15.48
Fazenda Tabocal-AM	1.00	0.71	0.50	1.50	02	1.00
Remanso-AM	18.16	7.70	8.36	29.02	12	19.75
Arapapá-AM	16.56	3.54	10.43	21.33	07	16.33
Axinim-AM	8.65	5.20	3.27	23.02	13	7.17
Espírito Santo-AM	12.47	4.03	3.51	21.28	18	13.30
Santa Maria-AM	9.28	3.57	6.70	16.84	07	7.55
Caiçara-AM	10.04	4.71	1.94	17.98	23	10.45
Paquiqué-AM	9.23	1.73	7.49	11.57	06	8.99
Uricurituba-AM	9.09	4.25	0.36	19.12	46	9.53
Santa Rosa II-AM	11.65	3.58	5.81	16.89	12	12.03
Average	15.22	9.60	5.99	150.00	713	12.48

consumed by these communities, since species preference varied according to species relative abundance in fishery catches, but about 78% of these communities consume fish two times per day all week long (Table 4).

Table 5 presents a comparison between the ranges of values found in bottom sediments, suspension solid, soil, fish and human hair measured in this study with the comprehensive review of Malm et al. (1990), which reported values relative to the peak period of gold mining activity in the Madeira River and close to point of Hg sources. As can be observed, Hg concentrations in sediment, soil and suspended solids are lower when compared with the 1990 survey. However, only part of our data originated from samples collected in the same area of the Madeira Basin. If comparisons are made with other studies of that period but including samples of soils and sediments from site not directly affected by gold mining, including some collected at the lower basin, concentrations are similar to those found in this study. For example, Lacerda et al. (1987) and Martinelli et al. (1988) reported bottom sediment concentrations ranging from 0.05 to 0.28 μg g^{-1} and 0.03 to 0.35 μg g^{-1}, respectively. For the middle 1990s Silveira et al. (2000) reported Hg concentrations in bottom sediment of 0.14 to 0.21 μg g^{-1}, not significant different from the concentrations reported here. Therefore, the decrease in Hg concentrations in these abiotic compartments may have happened mostly from sites close to sources at the higher Madeira River Basin where the peak of gold mining occurred. On the other hand, fish and human hair Hg concentrations are very similar between the late 1980s survey of Malm et al. (1990) and our values, notwithstanding the large reduction of the activity witnessed by 2000 and the fact that most of our fish samples come from the lower Madeira River Basin, when the present sampling took place. The Hg concentrations in fish and human hair presented here are also similar to concentrations summarized by Lacerda and Salomons (1998) for the gold rush period. Similarly, in a study of the upper Madeira River in Bolivia, Maurice-Bourgoin et al. (2000) reported about 86% of fish sampled to present Hg contamination up to 4 times the WHO limit for human consumption. Concentrations in piscivorous fish from the Beni River ranged from 0.33 to 2.30 μg g^{-1}. Also, riparian populations living along this river also presented relatively high average Hg concentration in hair (9.81 μg g^{-1}). These results suggest no reduction in Hg levels in biological compartments of the Madeira River Basin.

Table 4
Fish intake frequency for riverside populations of lower Madeira River

Consumption	Individual	Consumption (%)
Zero meals per week	05	0.70
Infants	07	1.00
One meal per week	10	1.50
Two meals per week	26	4.00
Three meals per week	49	7.50
Four meals per week	29	4.40
Five meals per week	16	2.40
Six meals per week	04	0.60
Seven meals per week	512	77.90
Total	658	100

Table 5
Hg concentration ranges (means values ± standard deviation) in bottom sediment, suspension solid, soil, fish and human hair between the years 1990 and 2000

Samples	(Malm et al., 1990) (μg g^{-1})	This work (μg g^{-1})
Bottom sediment	0.13 ± 0.11 (0.03–0.35)	0.06 ± 0.02 (0.03–0.12)
Suspension solid	0.50 ± 0.13 (0.35–0.58)	0.04 ± 0.02 (0.02–0.05)
Soil	0.39 ± 0.10 (0.27–0.54)	0.11 ± 0.09 (0.04–0.37)
Fish	0.92 ± 0.75 (0.10–2.10)	0.34 ± 0.36 (0.01–2.52)
Human hair	9.20 ± 14.0 (0.22–40.0)	15.22 ± 9.60 (0.36–150.0)

4. Conclusions

As a result of the present survey we show that notwithstanding the reduction of Hg emissions to the Madeira River Basin from gold mining proper, large amounts of Hg are presently being mobilized in the region. Reduction in Hg concentrations seems to be restricted to areas close to old point sources, and only for abiotic compartments. Remobilization of Hg from bottom sediments plus re-emission from soils due to land use changes are probably responsible for keeping Hg emissions at high levels. These processes have been recently accelerated by the opening of the Madeira River waterway and the "soya race" which resulted in large deforestation in Rondônia and Amazon states. Land use changes, in particular forest conversion to pasture by slash and burning, have been recently suggested a major cause of the maintenance of high Hg concentrations in the Amazon ecosystems either through Hg vaporization from burning the forest biomass, re-emission from denuded soil surfaces and increasing erosion (Lacerda, 1995; Roulet et al., 2000; Godoy et al., 2002; Lacerda et al., 2004). Also, while gold mining in Brazil dropped faster after the 1990s, it steadily increased in Bolivia and Peru, including tributaries of the Madeira River Basin. Finally, the biogeochemical processes typical of Amazon River

systems and marginal lakes are able to remobilize Hg due to efficient methylation and eventual bioaccumulation and biomagnification processes within food webs. The high concentrations measured in the local population highlights the necessity of a continuous monitoring program for the region, notwithstanding the present low level of anthropogenic Hg sources for the region.

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