

## Mercury pollution in the Tapajos River basin, Amazon Mercury level of head hair and health effects

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### Abstract

There is increasing concern about the potential neurotoxic effects of exposure to methylmercury for the 6 million people living in the Amazon, even in regions situated far away from the gold mines (garimpos), considered to be the major source of mercury pollution. In November 1998, a spot investigation on mercury contamination was conducted in three fishing villages (Barreiras, Rainha, and São Luiz do Tapajós) on the Tapajós River, an effluent of the Amazon, situated several hundred kilometers downstream from the gold-mining areas. A total of 132 fishermen and their families volunteered for the current study. As was anticipated, the total mercury levels in the head hair collected from the fishing villages were relatively high (14.1–20.8 ppm on the average) and the number of subjects with a high total mercury level over 10 ppm (the least upper bound of a normal value) was 103 (78.0%) in total, along with various symptoms, thereby suggesting wide mercury contamination in the Tapajós River basin. Moreover, in view of the absence of other diseases (e.g., alcoholism or malaria), a high intake of fish containing a methylmercury level, and high hair mercury levels in addition to the various symptoms such as sensory disturbance (especially glove-and-stocking type, which is characteristic of Minamata disease), tremor, failure in two-point discrimination, and slight balancing failure, several subjects examined were diagnosed with mild Minamata disease. The findings obtained suggest, thus, that the mercury pollution in the Amazon should be crucially observed for head hair mercury level and health in a much broader region. © 2001 Elsevier Science Ltd. All rights reserved.

**Keywords:** Mercury pollution; the Amazon; Minamata disease

### 1. Introduction

A number of research activities on the environmental impact of mercury from gold mining have concentrated on the Brazilian Amazon (Akagi et al., 1994, 1995; Boischio and Cernichiari, 1998; Branches et al., 1993; Cleary et al., 1994; Harada, 1997; Malm, 1998; Malm et al., 1990; Pfeiffer and Lacerda, 1988). The peak gold mining period in the Amazon occurred during the 1980s (Boischio and Cernichiari, 1998), and in February 1989, the Brazilian government enforced the law banning the

use of mercury (although the actual effects remain doubtful to the last). As indicated by Lebel et al. (1996) and Pfeiffer et al. (1993), however, there is increasing concern about the potential neurotoxic effects of exposure to methylmercury for the 6 million people living in the Amazon, even in regions situated far away from the gold mines (garimpos), considered to be the major source of mercury pollution. Indeed, our previous studies demonstrated that high hair mercury observed in goldminers and workers in gold shops was mostly in the inorganic form, whereas the predominant form of mercury in hair samples from fishing villages located downstream from the gold-mining areas was methylmercury (Akagi et al., 1994, 1995; Harada, 1997). Thus, it might be natural that recently, Lebel et al. (1996) detected alterations in nerv-

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ous system functions in young adults in two villages on the Tapajos River, an effluent of the Amazon, situated over 200 km downstream from the gold-mining areas. The Tapajos River region is currently the most important site of gold-mining activities in the Amazon basin (Pfeiffer et al., 1993).

Also, we periodically monitored the levels of total mercury and methylmercury in the head hair from fishermen and their families in three fishing villages (Barreiras, Rainha, and Sao Luiz do Tapajos) along the Tapajos River five times in Barreiras, three times in Rainha, and twice in Sao Luiz do Tapajos, respectively, from March 1994 till February 1998, and found that their hair mercury levels were all high during the period of investigation; that is, in Barreiras, Rainha, and Sao Luiz do Tapajos, the total mercury levels measured in the first place in the head hair were  $20.5 \pm 12.1$  (mean  $\pm$  S.D.; 1994.3;  $n=26$ ; highest value, 62.9 ppm),  $19.3 \pm 9.7$  (1994.3;  $n=16$ ; highest value, 39.0 ppm), and  $25.3 \pm 17.5$  ppm (1996.8;  $n=30$ ; highest value, 48.3 ppm), respectively, and, in February 1998, those measured lately were  $14.7 \pm 8.9$  ( $n=116$ ; highest value, 56.4 ppm),  $12.9 \pm 7.0$  ( $n=23$ ; highest value, 29.4 ppm), and  $16.0 \pm 9.2$  ppm ( $n=97$ ; highest value, 44.7 ppm), respectively (unpublished data). In addition, the predominant form of mercury in the hair samples was also methylmercury (average 87.8–96.0%) (unpublished data), although the methylmercury levels were not determined for those from Sao Luiz do Tapajos. Anxious about the future of such mercury contamination, therefore, Harada (1997) warned the Brazilian investigators as well as the local authorities concerned that Minamata disease would probably occur in the fishermen living along the Amazon basin in the not far distant future. In November 1998, the current study was thus designed to investigate the same three fishing villages by measuring their head hair total mercury levels and by a group medical exami-

nation. As the result, we could find that Minamata disease had already occurred there, albeit not serious.

## 2. Materials and methods

### 2.1. Study area

As already stated, the Tapajos River basin is considered the oldest and most productive gold-mining area (Akagi et al., 1995). The Barreiras fishing village with a population of about 1000 is located about 90 km downstream from Itaituba, the Rainha fishing village with a population of about 100 about 40 km upstream from Itaituba, and the Sao Luiz do Tapajos fishing village with a population of about 800 about 100 km upstream from Itaituba, as illustrated in Fig. 1.

### 2.2. Hair sampling

A total of 132 head hair samples were collected from fishermen and their families of the above three fishing villages where people consume locally caught fish. The details are given in Table 1. Each hair sample was cut as close to the scalp for total mercury analysis. Informed consent was obtained from all subjects, based upon the Declaration of Helsinki, 1964.

### 2.3. Group medical examinations

Moreover, of all the subjects investigated, 50 eligible subjects who had shown a high total mercury level of more than 20 ppm from March 1994 to February 1998, were examined clinically according to a questionnaire especially on fish-eating habits as well as on subjective symptoms, and a neurological test, because we had previously claimed that

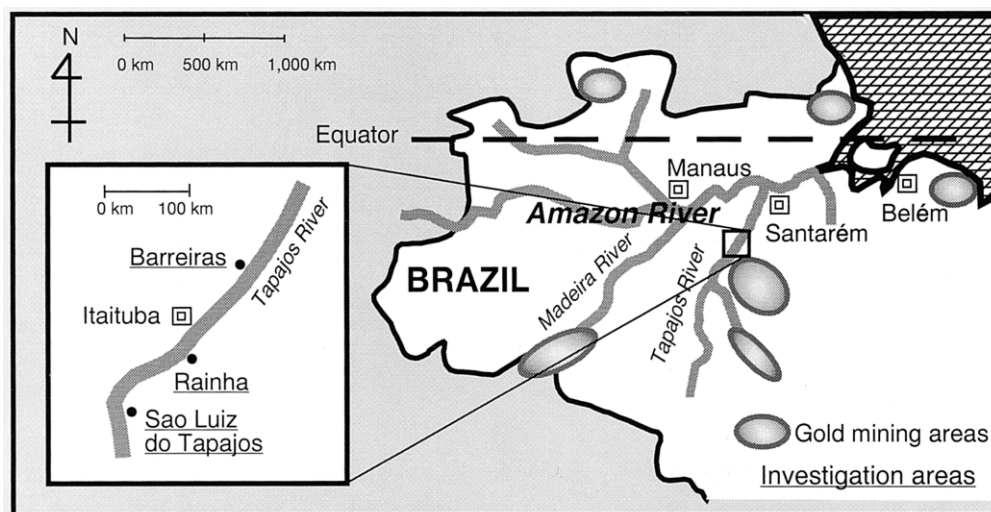


Fig. 1. Sketch map of the Tapajos River fishing villages showing the location of the study area.

Table 1

Total mercury level in head hair from the fishermen and their families living along the Tapajos River as of November 1998

Place	n	Sex		Age (years)		Total mercury (ppm)	
		Male	Female	Mean $\pm$ S.D.	Range	Mean $\pm$ S.D.	Range
Barreiras	76	32	44	28 $\pm$ 19	1–67	16.4 $\pm$ 10.6	1.8–53.8
Rainha	12	5	7	31 $\pm$ 17	7–53	14.1 $\pm$ 9.3	3.1–34.5
Sao Luiz do Tapajos	44	13	31	21 $\pm$ 13	3–47	20.8 $\pm$ 10.6	5.1–42.2
Eligible subjects examined clinically <sup>a</sup>	50	21	29	25 $\pm$ 18	3–65	23.6 $\pm$ 10.3	5.1–42.7

<sup>a</sup> These 50 subjects having shown a hair total mercury level of more than 20 ppm from March 1994 to February 1998 were involved in the above 132 individuals and examined clinically according to a questionnaire and a neurological test (Barreiras,  $n=22$ ; Rainha,  $n=4$ ; Sao Luiz do Tapajos,  $n=24$ ), with their previous mean maximum total mercury levels having been 29.1  $\pm$  9.2 ppm (20.3–71.6 ppm).

a person with a level of more than 20 ppm should be required to obtain a thorough medical examination (Doi and Ui, 1975; Harada et al., 1977; Ohno et al., 1984).

#### 2.4. Analytical procedure for total mercury

Hair samples were cut finely with surgical scissors just before analysis. Each total mercury level was measured as described previously (Ikingura and Akagi, 1996), with the precision and accuracy of this method having been repeatedly verified by interlaboratory calibration exercises (Department of Urban Engineering, Faculty of Engineering, University of Tokyo, Tokyo). There existed a high correlation between both results obtained (the correlation coefficient  $r=0.983$ ,  $n=10$ ), thereby indicating that the techniques used in the current study were reliable and accurate.

### 3. Results

As was anticipated, the total mercury levels in the head hair collected from the three fishing villages were 14.1–20.8 ppm on the average, all showing a high mercury level over 10 ppm (a more suitable threshold than 50 ppm as stated later) (Table 1). The number of subjects with a high total mercury level over 10 ppm was 57 (75.0%), 8 (66.7%), or 38 (86.4%) in Barreiras, Rainha, or Sao Luiz do Tapajos, respectively, thereby suggesting wide mercury contamination in the Tapajos River basin. As also expected, the 50 eligible subjects showed the highest total mercury level in the head hair (Table 1), with the number of subjects with a mercury level below 10 ppm being only 4 (8%). In the current study, not a few subjects were females (62.1%) and/or children from 15 years old under (32.6%). If more male adults participated in the current investigation, the total mercury levels in the head hair would probably have been higher.

As previously described, the 50 eligible subjects who had shown a hair total mercury level of more than 20 ppm were examined clinically. As to subjective symptoms, 17 of the subjects complained of numbness, 12 vertigo and dizziness, 12 headache, 11 lassitude, 9 pain in limb extremities, 7 pain in back, 7 reduction in vision, 6 trembling, 6 irritability, 5

reduction in hearing, 5 loss of memory, 5 motor disturbance, and 4 insomnia (Table 2).

Meanwhile, as to objective symptoms, we observed 16 of the subjects with sensory disturbances (7 cases of the glove-and-stocking type), 6 with disturbance in balance, 5 with disturbance in coordination, 4 with tremor, 4 with hyperreflexia, and 1 with dysarthria.

As shown in Table 2, neurological symptoms of the eligible subjects might be nonspecific, although similar symptoms are also observed in Minamata disease (Harada, 1997; Harada et al., 1998; Minamata Disease Research Group, 1968). In view of such variegated symptoms, the high hair mercury levels, and the presence or absence of other diseases, however, it seemed not to be surprising that

Table 2

Clinical symptoms of the 50 fishermen and their families having exhibited a hair total mercury level of more than 20 ppm from March 1994 till February 1998<sup>a</sup>

Symptom	n	%
<i>Subjective</i>		
Numbness	17	34.0
Vertigo and dizziness	12	24.0
Headache	12	24.0
Lassitude	11	22.0
Pain in limb extremities	9	18.0
Pain in back	7	14.0
Reduction in vision	7	14.0
Trembling	6	12.0
Irritability	6	12.0
Reduction in hearing	5	10.0
Loss of memory	5	10.0
Motor disturbance	5	10.0
Insomnia	4	8.0
Disturbance in taste (including metallic taste)	0	0.0
<i>Objective</i>		
Sensory disturbance	16	32.0
(glove-and-stocking type)	7	14.0
Disturbance in balance	6	12.0
Disturbance in coordination	5	10.0
Tremor	4	8.0
Hyperreflexia	4	8.0
Dysarthria	1	2.0
Gingivitis	0	0.0

<sup>a</sup> The symptoms were examined in November 1998.

Table 3

Hair total mercury levels and clinical symptoms of cases with mild Minamata disease and of suspected cases with Minamata disease

Case number	Age	Sex	Place	Occupation	Total mercury (ppm)						Main symptoms
					94.3 <sup>a</sup>	95.3 <sup>a</sup>	96.8 <sup>a</sup>	96.10 <sup>a</sup>	98.2 <sup>a</sup>	98.11	
<i>Mild Minamata disease</i>											
1	56	male	B	fisherman	62.9 (55.5; 88.2%)	48.8 (45.4; 93.0%)	71.5	56.3		41.8	D,G,M,N,O,P,T
2	18	male	B	fisherman/farmer					27.1	16.0	D,G,N,P,T,W
3	23	female	B	fisherwoman					30.0	35.6	D,G,N,P,T,V(?)
<i>Suspected Minamata disease</i>											
4	20	female	R	fisherwoman/farmer	21.5 (19.7; 91.6%)	25.0 (23.0; 92.0%)			15.7	26.5	G,N
5	44	female	S	fisherwoman/farmer			26.4			15.4	G,N,P,W
6	11	male	S	fisherman					21.9	31.7	C,G

B = Barreiras; C = circumoral sensory disturbance; D = disturbance in coordination; G = glove-and-stock-type sensory disturbance; M = reduction in manual dexterity; N = numbness; O = oculomotor disorder; P = failure in two-point discrimination; R = Rainha; S = Sao Luiz do Tapajos; T = tremor; V = constriction of visual field; W = muscular weakness.

<sup>a</sup> Unpublished data. Methylmercury level and the ratio of methylmercury to total mercury are given in parentheses.

several cases should be diagnosed as mild Minamata disease (Cases 1–3) or suspected Minamata disease (Cases 4–6) (Table 3) as in the following:

*Case 1:* A 56-year-old fisherman. The head hair levels of total mercury measured five times were 41.8–71.5 ppm, with the ratio of methylmercury to total mercury also being very high (Table 3). He had eaten 500–1000 g of fish daily, and had developed symptoms such as numbness, reduction in manual dexterity, butterfingering motion, and pain in shoulders for approximately 10 years. In addition, glove-and-stock-type sensory disturbance in extremities (which is the hallmark of Minamata disease; for review, see Harada, 1997), failure in two-point discrimination, tremor, oculomotor disorder, adiadochokinesis, failure in the finger-to-nose test, and balancing failure (the Mann's phenomenon) were observed. He was not alcoholic and had not contracted malaria, and worked in a garimpo for 3 months in 1960.

*Case 2:* An 18-year-old fisherman/farmer. The hair total mercury levels were 16.0–27.1 ppm (Table 3). He had eaten about 500 g of fish per day, and had complained of numbness of extremities and muscular weakness for about 2 years. Glove-and-stock-type sensory disturbance, failure in two-point discrimination, tremor, adiadochokinesis, failure in the finger-to-nose test, and failure in the knee-to-heel test were also observed. He had no drinking history. His father's total mercury level in the head hair was 21.3 ppm in February 1998, but he had died 1 month before the current examination.

*Case 3:* A 23-year-old fisherwoman. The hair total mercury levels were 30.0–35.6 ppm (Table 3). She had eaten about 300 g of fish per day, and had recently had various symptoms such as dullness, butterfingering motion, numbness, headache, reduction in vision, tinnitus, and muscular weakness. Glove-and-stock-type sensory disturbance, failure in two-point discrimination, balancing failure, failure in the finger-to-nose test, failure in the knee-to-heel test, mild tremor,

reduction in hearing, and muscular weakness were also observed. The stricture of the left visual field was suspected by a facing method. She was highly hypochondriac and had no drinking history.

*Case 4:* A 20-year-old fisherwoman/farmer. The hair total mercury levels were 15.7–26.5 ppm, with the ratio of methylmercury to total mercury being very high (Table 3). She had eaten about 500 g of fish daily, and had complained of arthralgia of extremities, irritation, dizziness, and occasional numbness. Glove-and-stock-type sensory disturbance and failure in two-point discrimination were also found. In addition, her daughter (7 years old) was suspected of fetal Minamata disease, although attempts to make a differential diagnosis from cerebral paralysis presented a great deal of difficulty, because of the loss of her clinical chart of birth. We could not find it in a hospital in Itaituba where she had been born; so that the details including body weight at birth were unclear, apart from the facts that the baby had severe cyanosis and did not cry for a week after birth. The hair total mercury levels were 9.7 (methylmercury: 9.8 ppm, 101%) and 11.4 ppm in March 1994 and in the current study, respectively. She had severe intellectual failure, hyperkinesia, microcephalia, strabismus, hypersalivation, hypotonia, or no language ability. She could not walk by herself, and needed support to walk. It should be noted that her mother's hair total mercury levels had been high throughout (more than 15 ppm) (Grandjean et al., 1997, 1999). Moreover, the daughter was a breast-fed child.

*Case 5:* A 44-year-old fisherwoman/farmer. The hair total mercury levels were 15.4–26.4 ppm (Table 3). She had eaten about 500 g of fish per day, and had shown various symptoms such as lassitude, breathlessness, headache, tinnitus, numbness, dullness, muscular weakness, and insomnia for the past 5–6 years. Glove-and-stock-type sensory disturbance, failure in two-point discrimination, mild deformation of fingers, balancing failure, and hyperreflexia of the lower limb tendons were

also observed. She was hypochondriacal and depressed, and underwent treatment of hypertension. She had no drinking history. No significant abnormality was detected in the heart by stethoscopy.

*Case 6:* An 11-year-old fisherman. The hair total mercury levels were 21.9–31.7 ppm. He had eaten about 500 g of fish daily. He sometimes had headache, but no other subjective symptoms. Glove-and-stocking-type sensory disturbance in addition to numbness around the mouth was found, but tendon reflexes were normal.

#### 4. Discussion

No measurement of methylmercury was made in the current study because, as already stated, our previous studies had revealed that the ratio of methylmercury to total mercury in the head hair from the Barreiras and Rainha fishing villages was very high, as only partially shown in Table 3. The study of Lebel et al. (1996) in other two villages on the Tapajos River confirmed this finding (72.2–93.3%).

In an earlier study, we have assumed that the upper limit of a normal hair total mercury level is 10 ppm (Harada et al., 1999) [almost equivalent to the mean value (4.6 ppm) of normal male Japanese subjects  $\pm 2$  S.D. ( $2 \times 2.4$  ppm)] (Ohno et al., 1984). Actually, Grandjean et al. (1997, 1999) showed in the Faroe Islands and the Tapajos River basin that mercury-related neuropsychological dysfunctions seemed to be present in children even below a limit of 10 ppm for mercury in maternal head hair, and that the mercury contamination that more than 80% of 246 children had hair mercury levels above 10 ppm seemed sufficiently severe to cause adverse effects on brain development, respectively. Moreover, the preliminary study undertaken in two villages on the Tapajos River demonstrates that it is possible, using a sensitive battery, to detect alternations in nervous system functions, consistent with knowledge on mercury toxicity, at levels below the currently recognized threshold of 50 ppm total mercury (Lebel et al., 1996).

In the study by Lebel et al. (1996), hair mercury levels in this population living on the Tapajos River were similar to those reported previously for groups residing in this river basin and considerably higher than those observed in fish-eating communities in Europe and Japan. The current study also confirmed these findings. For example, for control purposes, 15 native people (the Yanomani tribe) in Boa Vista (a tropical rain forest area), 16 farmers in Vila Mae de Deus (a farm village), and 46 residents in Belém (a city) were also examined in November 1989, March 1994, and August 1996, respectively, with each mean hair total mercury level being 3.6 (highest value, 7.5 ppm), 2.1 (highest value, 5.2 ppm), and 1.5 ppm (highest value, 8.9 ppm) (unpublished data), which were apparently lower than those observed in the current study. The three places investigated were remotely related to garimpos. Moreover,

even in some mercury-polluted areas along the Shiranui Sea (in which Minamata disease was discovered in May 1956), with six exceptions (highest value, 22.5 ppm), 185 fishermen and their families showed a normal total mercury level in hair ( $<10.0$  ppm) with a high ratio of methylmercury to total mercury in August 1995 (Harada et al., 1998). Also, it should be emphasized that attempts to do this type of clinical examinations in such a backward region, the Amazon region, presented difficulties, especially when dealing with nonspecific effects of past exposures that may be difficult to assess from current hair mercury concentrations.

Our field investigations including the current study have been performed not only by collecting head hair samples, but also by making a group medical examination (Harada, 1995, 1997; Harada et al., 1977, 1998, 1999, 2001; Ohno et al., 1984). In addition, in the current study, physicians of Universidade Federal do Pará, Belém, gave their medical services to the subjects investigated even when their symptoms were not considered to be attributable to mercury poisoning. For instance, as of March 1994, of the 102 fishermen and their families investigated, subjectively, 32 (31.4%) had headache, 21 (20.6%) had vertigo and dizziness, 16 (15.7%) had pain in limb extremities, and 14 (13.7%) had numbness, whereas, objectively, 11 (10.8%) had sensory disturbance, 5 (4.9%) had tremor, and 1 (1.0%) had hemiplegia; however, distal-dominant sensory disturbance (glove-and-stocking type), which is characteristic of Minamata disease, was observed only in one fisherman with a hair total mercury level of 17.0 ppm. He had worked in some garimpo up to 1991 (3 years before the examination) and returned to the Barreiras fishing village because his health had failed, probably due to inorganic mercury poisoning. Therefore, in 1994, neither fishermen nor their families examined were diagnosed with Minamata disease, although they had various symptoms in addition to high hair mercury levels (unpublished data).

On the other hand, in the current study, general sensory disturbance was observed in 16 out of the 50 subjects (32.0%) in whom hair total mercury levels had been more than 20 ppm. Of the 16 subjects, 7 (14.0%) had glove-and-stocking-type sensory disturbance (Table 2). This glove-and-stocking-type sensory disturbance may be sometimes observed for other diseases, but its incidence had been extremely high in the Minamata area where methylmercury pollution had occurred (Harada, 1995, 1997; Harada et al., 1998). For example, as of August 1995, out of 188 inhabitants of the Minamata area, 130 (69.1%) had glove-and-stocking-type sensory disturbance (Harada et al., 1998). Furthermore, considering various symptoms such as tremor, failure in two-point discrimination, and mild balancing failure in addition to the absence of other diseases (e.g., alcoholism or malaria), a high intake of fish containing a high methylmercury level (Akagi et al., 1994), and high hair mercury levels, several subjects examined could be diagnosed with Minamata disease, albeit not serious, as shown in Table 3.

Mercury pollution has been investigated only in limited regions in the large Amazon basin. Thus, the mercury pollution in the Amazon should be continuously and crucially observed for head hair mercury level and health in much broader regions.

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