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Mercury Contamination in Humans in Upper Maroni, French Guiana Between 2004 and 2009

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Abstract We measured hair mercury concentration in Amerindians between 2004 and 2009 in Upper Maroni, French Guiana. Hair samples were collected from 387 residents (males: 153, females: 234). Average hair mercury concentration was high (males: 9.4 ppm, females: 9.9 ppm). We examined fish consumption by 37 residents. There was a significant correlation between hair mercury concentration and fish consumption. We also measured mercury concentration in polluted fish in upper reaches of the Maroni River. Muscle mercury concentrations were high in the fish. These results indicate that current high hair mercury concentrations in Amerindians remain linked to fish consumption.

Keywords Mercury pollution · Human exposure · Upper Maroni · French Guiana

Many Amerindians live along the upper reaches of the Maroni River, French Guiana. In this area, clandestine gold mining has contaminated numerous terrestrial and aquatic sites. Although the French government has issued reports on gold mining in the region, they are outdated. At the end of the twentieth century, human exposure to mercury in the region was detected by the measurement of hair mercury (Cordier et al. 1998; Fréry et al. 2001). A clinical survey carried out in French Guiana showed a significant correlation between mercury contamination levels and neurological impairment. The average hair concentration of mercury in Amerindian children in Upper Maroni was 12.0 and 12.6 ppm in hair of males and females, respectively, and many were afflicted by neurological disorders such as poor coordination of the legs and decreased performance in the copying section the Stanford–Binet intelligence test (Cordier et al. 2002). The same team recently published data on Amerindian children in Upper Maroni and found a decreased level of mercury in hair with a mean of 10.5 ppm in 2007 (Chevrier et al. 2009). Nevertheless, one should not hastily conclude that the levels of mercury in hair had declined in these communities between 2002 and 2007. Indeed, another study showed that the mercury concentrations in hair had increased in the Upper Maroni communities between 1997 and 2005 from 11.4 to 13.1 ppm (Quénel et al. 2007). Therefore, facing these two conflicting studies, we wanted to address the question of whether hair mercury levels are increasing, decreasing, or are remaining constant with time.

Materials and Methods

In the period 2004–2009, socio-demographic information was collected using a self-administered questionnaire survey in Cayode, Twenke/Taluwen, Antecume Pata and Elahe, including questions on age, sex. In addition, in May

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and September 2008, a food frequency questionnaire survey of fish consumption was administered in Cayode and Twenke/Taluwen villages; there were 37 subjects. Information collected included daily intake quantities, and species of fish consumed. Four sampling sites for human hair were located in Upper Maroni villages of Cayode, Twenke/Taluwen, Antecume Pata, and Elahe (Fig. 1). Hair samples from 387 village inhabitants (males: 153, females: 234) were taken from 2004 to 2009, particularly at least once a year from almost the same subjects in Cayode. Samples were cut from the occipital area close to the scalp. In March 2009, we sampled fish (Fig. 1). There were 2 sampling sites for fish, in the Maroni River near Twenke/Taluwen village, and in the Tampok River, a branch of the Maroni River near Cayode. Flesh samples were taken from the dorsal sides of 6 fish species (*Pseudoplatystoma fasciatum*, *Serrasalmus rhombeus*, *Platydoras costatus*, *Ageneiosus brevifilis*, *Doras micropeus* and *Astyanax/Moenkhausia* spp.) and stored in a refrigerator.

Samples were analyzed for total mercury and MeHg at the National Institute for Minamata Disease (NIMD), Japan using reliable and sensitive techniques following the procedures of Akagi and Nishimura (1991) and modified by Akagi et al. (1995). Hair samples were measured for total mercury and MeHg; fish samples were subjected to analysis of total mercury only. The precision and accuracy of these techniques have been verified repeatedly by inter-laboratory calibration (Matsuo et al. 1989; Malm et al. 1995) that has included analysis of reference standards (e.g., IAEA 086 and DORM2). Total hair mercury was determined directly by the oxygen combustion-gold amalgamation procedure using a Mercury Analyzer, MA2000 (Nippon Instruments, Japan). The limit of detection (LOD) was 0.17 ng/g calculated from the standard deviation of six blanks. MeHg was extracted from the sample by adding 2 N HCl and heating at 100°C for 5 min. After heating, the HCl extract was further extracted with toluene. MeHg in the toluene extract was determined by gas chromatography coupled with electron capture detection (GC-ECD, Yanaco G3800, Japan). The LOD was 0.01 ng/g calculated from the standard deviation of six blanks. The fortified sample (IAEA086) recovery was 102% ± 2.2% (mean ± SEM, n = 4). To control measurement quality, a standard reference material IAEA 086 certified values of mercury: 0.573 ± 0.039 ppm (mean ± SEM) as total mercury, 0.258 ± 0.022 ppm as MeHg) was included in the analyses. Our qualification data were 0.59 ± 0.01 ppm as total mercury, 0.26 ± 0.01 ppm as MeHg (mean ± SEM, n = 7). Fish samples were analyzed for total mercury concentration without drying. Total mercury concentration was determined by cold vapor atomic absorption spectrometry (CV-AAS). Briefly, fish muscle was inserted into a volumetric flask, followed by the addition of a mixture of nitric and perchloric acids (1:1), sulfuric acid, and water. The flask was heated at 200°C for 30 min. After cooling to room temperature, distilled water was added to the digest, rendering the sample ready for mercury analysis by CV-AAS. The LOD was 0.07 ng/g calculated from the standard deviation of six blanks. The fortified sample (DORM2) recovery was 100 ± 2.5% (mean ± SEM, n = 4). To control the quality of measurements, a standard reference material, DORM2 [certified value of total mercury is 4.64 ± 0.26 ppm (mean ± SEM)], was included in the analyses. Our qualification data were 4.70 ± 0.15 ppm as total mercury (mean ± SEM, n = 7).

Results and Discussion

Human subjects were from 1 to 53 years old. The frequency distribution of total mercury concentration in the 387 hair samples (males: 153, females: 234) is presented in Fig. 2. The average concentration was high (males: 9.4 ppm, females: 9.9 ppm) compared with that in a region

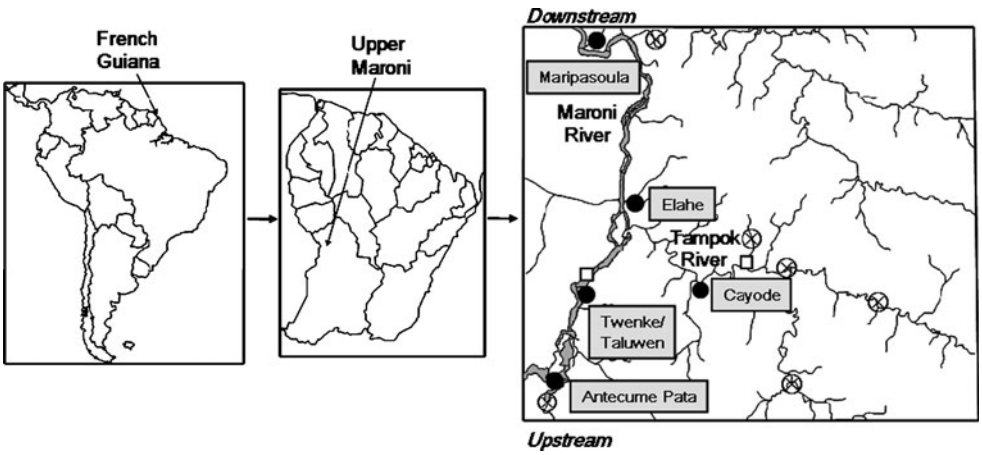

Fig. 1 Study area in Upper Maroni, French Guiana. Sampling sites for human hair (*filled circle*) and fishes (*open square*) are shown in the right panel. Gold mining sites (*circled times*) are also indicated

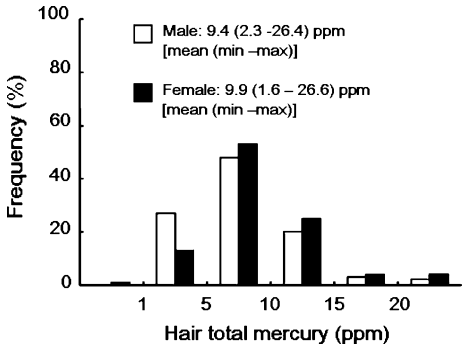

Fig. 2 Frequency distribution of total mercury concentrations in Amerindian hair. Hair samples were collected from 387 residents (males: 153, females: 234)

Table 1 Time course of total hair mercury levels in Cayode from 2004 to 2009

Date (year)	No. of subjects	Hair total mercury (ppm) [mean (min–max)]
2004	37	12.4 (6.4–22.0}
2005	32	12.8 (6.4–26.6}
2006	39	10.2 (3.0–13.5)
2007	24	9.6 (3.4–20.7)
2008	37	7.2 (2.8–17.9)
2009	25	12.8 (4.7–24.6)

devoid of gold-mining activity (2.6 ppm, n = 77), located on the Atlantic coast in French Guiana (Cordier et al. 2002). Our results compare well with previous reports in the same mercury-contaminated area of French Guiana. Average hair mercury concentrations of 11.4 ppm (n = 235) in 1997 (Fréry et al., 2001), 12.2 ppm (n = 90) in 1997 and 1998 (Cordier et al. 2002), 13.1 ppm (n = 130) in 2005 (Quénel et al. 2007), 10.5 ppm in children's hair (n = 65) and 12.9 ppm in maternal hair (n = 58) in 2007 (Chevrier et al. 2009) has reported. In the present study, 30% of the subjects had mercury levels above the safety limit determined by WHO (10 ppm), and the maximum value was 26.6 ppm (Fig. 2). In Cayode village where almost the same subjects were sampled at least once a year, however, hair mercury concentration decreased from 2006 to 2008, but increased again in 2009 (Table 1). Among 52 individuals that showed highly contaminated hair samples (≥10 ppm) and living in Amerindian villages, we found that 94.5% of the mercury was in the methylated form. Hair MeHg is an indicator of dietary mercury exposure (Wilhelm et al. 1996; Drasch et al. 2001; Legrand et al. 2007) which is aggravated by the proximity of gold-mining areas (Malm et al. 1997). Our results show that dietary mercury still remains the major source of contamination. Importantly, Amerindians living in the Upper Maroni are not gold-miners, and most probably they became contaminated through consumption of fish with high muscle mercury concentrations.

Overall, mercury concentrations in fish muscle did not exceed 0.5 ppm wet weight (Table 2). However, the mercury concentrations were high in the fish species *P. fasciatum*, *S. rhombeus*, and *P. costatus* (averages of 0.33, 0.40, and 0.32 ppm wet weight, respectively) and are in the range of what has been reported in carnivorous species in the Amazonian basin (Berzas Nevado et al. 2010). Since fish flesh contains about 70%–75% of water, the mercury concentrations in these species range between 1.0–1.4 ppm on a dry weight basis. These values are commensurate to those reported for the same species in 1997 (Fréry et al. 2001). Although we attempted to include the piscivorous species *Hoplias aimara* in our analysis, we were unable to catch specimens in March 2009. Previously, mercury concentrations in the range of 2–5 ppm dry weight have been reported in this species (Fréry et al. 2001; Durrieu et al. 2005; Maury-Brachet et al. 2006) making about 0.5–1.25 ppm wet weight, and indeed a very high concentration of mercury. Our questionnaire survey showed that the Amerindians have a preference for mercury-contaminated fish species, especially piscivorous species such as *P. fasciatum* and *H. aimara*. Furthermore, we calculated daily mercury intakes by multiplying average fish consumption by mercury concentrations found in fish flesh (given in Table 2). In the case of *H. Aimara* species, the mercury concentrations were taken from Fréry et al. 2001

Table 2 Mercury concentrations in the flesh of 6 fish species collected in Upper Maroni, French Guiana in 2009

Family	Species	Amerindian name	No. of fish	Length (cm) [mean (min–max)]	Total mercury in muscle (ppm) [mean (min–max)]
Pimelodidate	*Pseudoplatystoma fasciatum*	Hulluwi	6	46.8 (32.5–58.7)	0.33 (0.24–0.44)
Serrasaimidae	*Serrasalmus rhombeus*	Piraie (Pene)	3	43.3 (38.2–47.3)	0.40 (0.34–0.46)
Doradidae	*Platydoras costatus*	Hoké	1	23.4	0.32
Ageneiosidae	*Ageneiosus brevifilis*	Mitala	1	26.8	0.18
Doradidase	*Doras micropeus*	Agonosu	1	18.3	0.11
Characidae	*Astyanax/Moenkhausia* spp.	Yaya (Otululu, opi)	1	16.7	0.18

Table 3 Fish consumption and daily mercury intake in Cayode and Twenke/Taluwen villages in 2008

Species	Average of fish consumption (g/day) [mean (min–max)[a]]	Calculated daily mercury intake (mg/day) [mean[b]]
Pseudopaltystoma fasciatum	59.8 (0–150)	19.7
Hoplias aimara	52.7 (0–150)	47.4
Serrasalmus rhombeus	49.2 (0–150)	19.7
Platydoras costatus	44.0 (0-150)	14.1
Ageneiosus brevifilis	36.2 (0–150)	6.3
Doras micropeus	28.1 (0–50)	3.1
Astyanax/Moenkhausia spp.	17.6 (0–100)	3.2

[a] The seven most preferred fish species were listed

[b] Daily mercury intakes were calculated by multiplaying the average fish consumption by mercury concentrations found in fish flesh (given in Table 2). In the case of *Hoplias aimara*, the mercury concentrations were taken from Fréry et al. 2001

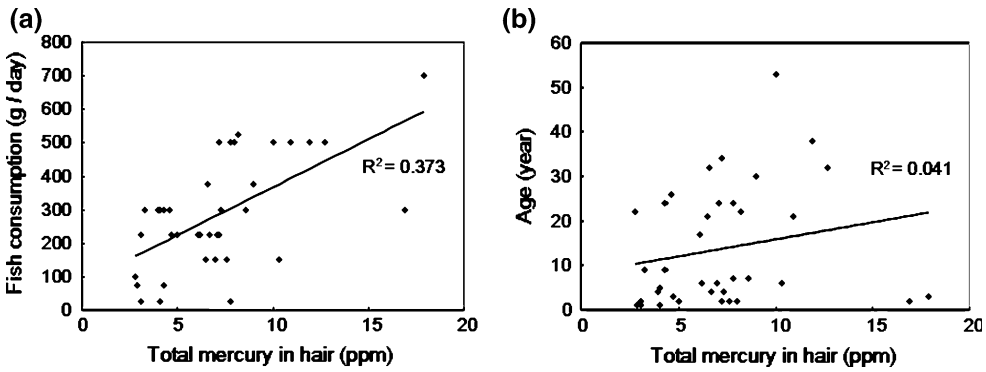

Fig. 3 Correlation between total mercury concentration in hair and daily fish consumption (**a**) and age (**b**) among 37 subjects in Cayode and Twenke/Taluwen villages in 2008

(Table 3). These results show that the major source of mercury intake was *H. Aimara*. Moreover, total daily amount of fish consumed was positively correlated with total hair mercury ($R^2 = 0.373$, $p < 0.05$ by regression analysis) (Fig. 3a), while the age of subjects was not matched to the total hair mercury ($R^2 = 0.041$) (Fig. 3b). In this study, we found that the preferred fish species were *P. fasciatum* > *H. aimara* > *A. brevifilis* > *D. micropeus* > *S. rhombeus* > *P. costatus*. Eleven years ago the recorded preferred fish species were ranked as follows: *Myleus torretes* > *D. micropeus* > *H. aimara* > *P. costatus* > *Prochilodus reticulata* > *P. fasciatum* > *A. brevifilis* > *S. rhombeus* (Fréry et al. 2001). Apart from *M. torretes* and *P. reticulata*, which are not heavily loaded with mercury, the piscivorous fish are still favored and highly represented in the Amerindians' diet, indicating that current high hair mercury concentrations in Amerindians remain linked to fish consumption.

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