Gold-Mining Activities and Mercury Contamination of Native Amerindian Communities in French Guiana: Key Role of Fish in Dietary Uptake

N. Fréry,¹ R. Maury-Brachet,² E. Maillot,¹ M. Deheeger,³ B. de Mérona,⁴ and Alain Boudou²

¹French National Institute of Public Health Surveillance, Saint-Maurice, France; ²Laboratory of Ecophysiologie et Ecotoxicologie des Systèmes Aquatiques, Arcachon, France; ³INSERM, Paris, France; ⁴Institut de Recherche pour le Développement, Laboratoire d'Hydrobiologie, Cayenne, French Guiana

In 1994, the French National Public Health Network reported significant mercury exposure of native Amerindians in French Guiana. In 1997, a study was conducted in the Wayana community to quantify the dietary intake and to identify the fish species contributing the most to the contamination. The study was completed by an impregnation analysis based on Hg determination in hair samples. The methodology used was a detailed familial dietary study associated with Hg measurements in fish and some game. The study was conducted over 7 days in two different seasons in the four most populated Wayana villages on the upper part of the Maroni River (521 people; 70% of the Wayana population in French Guiana). Analysis was based on data on consumption obtained from 165 people in a 1–14 day period (i.e., 940 persons \times days) and involved 270 fish samples from 48 species. Total Hg and monomethylmercury (MMHg) were also determined in hair samples (235 samples for total Hg). The results confirm mercury exposure of the Wayana population related to a diet rich in fish, which are relatively highly contaminated for certain species (up to 1.62 mg/kg fresh weight or 8.1 mg/kg dry weight in skeletal muscle). Results from hair samples showed that 57% of the Amerindians had Hg levels above the World Health Organization (WHO) safety limit (10 µg/g); all those over 1 year of age had a Hg intake greater than the WHO safety limit (200 µg MMHg/week for a 60-kg male). Hg concentrations in fish muscle were closely linked to the feeding regime and position of fish in the food webs. Overall, 14.5% of the fish collected exceeded the 0.5 mg/kg (fresh weight) safety limit. Four carnivorous species accounted for no less than 72% of the metal ingested by the Wayana families, although these represented only 28% of the consumed fish biomass. In conclusion, this study revealed excessive exposure to mercury in the Wayana population in French Guiana related to the consumption of contaminated fish. Key words. Amerindians, dietary uptake, fish, French Guiana, gold mining, hair, mercury. Environ Health Perspect 109:449-456 (2001). [Online 1 May 2001] http://ehpnet1.niehs.nih.gov/docs/2001/109p449-456frery/abstract.html

Gold-mining activities have been responsible for discharges of mercury into the environment as a consequence of using the mercury amalgamation method, where the gold obtained from river sediments, soils, or groundwater rocks is separated from elemental mercury (Hg°) by open circuit heating. In Brazil, for example, it is estimated that more than 130 tons of mercury are used per year, on the basis of 1.4 kg Hg for 1 kg of gold (1,2). More than 6 million people participated in gold prospecting in the Amazon region alone during the gold rush at the end of the 1880s (1). This volatile form of mercury is discharged into the atmosphere (55%) and aquatic biotopes (45%) (1). Within the biogeochemical cycle of the metal, Hg° can be oxidized in inorganic mercury (HgII) and then methylated by biotic (bacterial methylation) and/or abiotic (humic acids as methyl group donors for example) processes (3).

Studies in the Amazon Basin have shown numerous examples of contamination of hydrosystems by mercury. Concentrations measured in aquatic organisms were different according to their position within the food webs: predatory species, particularly the piscivorous species, have the highest levels of contamination due to the biomagnification of the methylated form of the metal. In a survey conducted between 1987 and 1990, approximately one-third of the carnivorous fish caught downstream from the Madeira River gold-mining area had mercury levels > 0.5 μ g/g fresh weight (fw), which is the safety limit (4).

Riverside populations of the Amazon tend to present high Hg levels in hair because fish is their main protein source. For example, the average of hair Hg in family members of fishermen from the Duas Bocas Lake shores (Amapa State, Brazil), directly influenced by Hg releases from gold mining, was 28 μ g/g. The World Health Organization (WHO) suggests a 10 µg/g safety limit, above which adverse effects on brain development are likely to occur (5). At such levels of Hg impregnation, health effects are measurable, with reduced peripheral visual field profiles, reduced color discrimination capacities, and reduced performance in psychomotor and neuropsychologic tests (6-9).

In French Guiana, available data are currently limited. Since the end of the last century, gold mining has been carried out at numerous sites and still continues to be practiced by international mining companies and smaller groups on terrestrial sites or directly in the rivers. Official gold production during 1857-1992 was about 200 tons, but accurate data are sparse, and a large amount of illegal production occurred (10). A study of Hg impregnation of the whole Guianan population was conducted in 1994, with the analysis of the metal in hair samples (11). Results did not show high levels of Hg, except for in the native Amerindian communities, particularly in the Wayanas living in the upper reaches of the Maroni River. This contamination probably reflects past and current gold-mining activities in this area, linked to the diet of these populations, of which fish is a main component. This population's health is a major concern for France because the Wayana is an ethnic group that may be vulnerable due to their particular way of life, such as Cree in Quebec or people from the Feroe Islands.

This possible health risk led the French National Institute of Public Health Surveillance (InVS/ex-RNSP), in collaboration with the National Scientific Research Center and the health authorities, to launch several studies including a dietary study. The study is part of a vast program initiated in Guiana to analyze the biogeochemical cycle

We are grateful to the authorities in French Guiana—Direction Départementale des Affaires Sanitaires et Sociales, Direction des Services Véterinaires, Conseil général, Centre d'Aide et d'Intervention Technique, de Maripassoula—for their assistance during the setting up and the followup of the two missions. In the villages, health workers, instructors, the "Grand Man" (A. Twenké), and all the Wayana communities participated with much enthusiasm and willingness. We also thank J.M. Jegu (Museum National d'Histoire Naturelle/Institut de Recherche pour le Développement) for his assistance in determining several fish species, S. Cordier for helpful comments on the manuscript, and C. Sourceau for proofreading assistance.

This work was supported by the E.C. Feder funds, via the Prefecture de Cayenne, the French Ministry of the Environment, and the National Scientific Research Center (Programme Environnement, Vie et Sociétés, "Mercury in Guiana").

Received 22 June 2000; accepted 14 November 2000.

Address correspondence to A. Boudou, Laboratory of Ecophysiologie et Ecotoxicologie des Systèmes Aquatiques, UMR CNRS 5805, Université Bordeaux 1, Place du Dr Peyneau, 33120 Arcachon, France. Telephone: (33) 5 56 22 39 20. Fax: (33) 5 56 54 93 83. E-mail: a.boudou@ecotox.u-bordeaux.fr

of mercury and to estimate the sanitary and environmental impact of the metal. In this paper, we present the study conducted in the native Wayana community, which has the principal goal of quantifying the dietary Hg intake and identifying the species of fish contributing the most to the human contamination. To do this, we conducted an impregnation study, based on Hg and MMHg determination in hair samples.

Methods

Characteristics of the Wayana populations living in the upper part of the Maroni River. The Wayana Amerindian population belongs to a large cultural and linguistic group who lives between the Orenoque River in Venezuela and the Amazon Basin (Brazil, Guiana, Surinam). The Wavanas live in the South of French Guiana along the upper Maroni River, mostly in the four villages selected for the study. The study subjects represented 70% of the whole French Wayana population (Antecume Pata, Twenke, Taluhen, and Cayode on the Tampok River; three sites) (Figure 1). The villages are accessible only by pirogues or helicopter, and access is controlled by prefectorial authorization. In March 1997, the total population in the four villages was 521, 286 men and 235 women: 54.3% of the population were under 20 years of age and only 5.4% were 60 years of age or over. The remaining French Wayana population lives in smaller villages or hamlets, with the same way of life as in the four villages, organized around clearing, hunting, fishing, and gathering, which provide all dietary needs.

Methodologic basis for the dietary survey and calculation of mercury intake. In August 1996, information meetings were organized in the four villages with the Wayana chiefs, health agents, and teachers concerning the future studies on the mercury problem, and a study of feasibility was performed within the population. To set up the nutritional study, information meetings were organized with the inhabitants, with simultaneous translation into Wavana. as well as in schools with the teachers. Although the Wayana language is spoken by everyone, French is mainly spoken only by the youngest people (generally under 30 years of age). For the last 20-25 years, primary education has been conducted in schools within the villages. It was therefore possible to identify in most households at least one person who was able to speak French, willing to participate, and willing to be our correspondent within the household.

The study was conducted over 7 days during two different seasons (March and November 1997; high and low waters, respectively), which are distinguished by changes in the diet, particularly fish and game. We selected 23 households according to geographic and feasibility criteria as well as the households' willingness to participate (so they were not randomly included): 10 in Twenke, 3 in Taluhen, 5 in Cayode, and 5 in Antecume Pata. Except for the problem of language comprehension, the main logistic difficulties were the frequent absence of the inhabitants and the scattering of families within the village. We selected households according to the diversity in the number of household members and their ages. Consumption data for these households, corresponding to 165 subjects studied over a period of 1-14 days (variations due to logistic difficulties cited above), allowed us to obtain 940 daily intakes (i.e., person \times day).

Each day the following was recorded in the studied households: *a*) fish consumed, including name of the species, number, size, total weight, and weight after evisceration, without bones; and *b*) the distribution of the household members participating in the meals (by sex, age, and weight). The record of fish consumed, which is approximately 250 kg of fish for the whole study, allowed us to calculate the amount of Hg ingested by the households each day because we calculated the mean Hg concentration according to the fish species and their weight category from the ecotoxicologic study (see below).

Each day the amount of fish consumed by each person within each household was calculated using the standard portions. The standard portion (SP) was the average of the daily amount of fish consumed by category of age (eight classes) and sex. SPs were calculated from a smaller number of persons selected according to their age and sex in different villages, but mainly in Twenke for logistic reasons. SPs corresponded to 227 daily intakes obtained by weighing each food consumed by an individual of each particular sex and age category.



Figure 1. Location of the four Wayana villages—Cayode, Twenke, Taluhen, Antecume Pata— in three sites of the upper part of the Maroni River, French Guiana.

The proportion of fish consumed each day by an individual belonging to a given sex and age category was obtained after dividing the corresponding standard portion by the sum of the standard portion corresponding to each member of the household on that day.

Proportion of fish consumed = $SP_{sa}/Sum_h (SP_{sa})$,

where h is household, s is sex, and a is age. Each day, the individual consumption of mercury within a household was obtained by its proportion of consumption multiplied by the amount of Hg ingested by the family that day:

The average daily intake for an age group was obtained by adding the different intakes of that group obtained along the study because we wanted to obtain a global intake, independent of the season. However, we checked that the number of records was about the same at the two periods and corresponded to about the same percentage of children and adults. In parallel with the nutritional survey, morphometric profiles (weight, size) were taken for the study subjects of the four villages, and precise ages were obtained from health notebooks. The amount of Hg consumed per kilogram of body weight ($\mu g Hg/kg \times day$) was calculated for each daily consumption, then the average was presented by age group. This calculation from the weight of each individual gives a more accurate result than from the average weight in this age group.

The dietary intake of mercury was quantified from the amount of fish consumed. The days when meat was eaten were entered as 0 ng Hg/day for different reasons: a) the game species consumed during the two periods were not representative of all the species consumed during the year; b) it was not easy to estimate the weight of game that was already shared between different villages or households; and c) to avoid overestimating the mercury intake related to a diet exclusively based on fish (generally, Hg fish concentration >> Hg game concentration). Thus, 65.5% of the persons \times days only included fish, 24.5% were mixed days (fish and meat), and 10% were exclusively based on meat. Nevertheless, a small number of game samples were also collected (doe, peccary, tapir, monkey, armadillo), and total Hg was analyzed in different organs, in particular the skeletal muscle and liver.

Other methods of estimation (generally grosser) could not be used in this context;

this is the case of the 24-hr memory method, which involves the difficulty of remembering or mistaking the amount consumed during the day, or the global method on the scale of the entire village, which consists of measuring all food entering the village (hunting, fishing). This method would have required a larger number of investigators because the location of the villages on islands allows multiple arrivals. In addition, the use of the "double portion" due to nonexcessive food intake could not be considered.

Hair sampling and mercury determination. Mercury in hair is a good indicator of Hg blood levels, particularly when an individual is exposed to organic mercury of dietary origin. In humans, monomethyl mercury (MMHg) is distributed between blood and hair in a ratio of 1:250 (5).

Hair was collected from 235 persons, in the occipital region where hair growth is constant. It was sampled at the root with scissors, leaving a maximum of 6 cm (due to exogenous contamination); the weight of the final sample should be a minimum of 30 mg. Samples were stored in envelopes. Total Hg determinations were carried out by the Center of Toxicology of Quebec, Quebec, Canada), a reference laboratory for this type of analysis. Hair samples were not washed before analysis because this practice has been shown to be inefficient; Hg is so tightly bound to the hair that it is impossible to remove the metal without damaging the hair. Samples (20 mg) were digested for 48 hr in nitric acid at room temperature. An aliquot was then introduced into an alkaline reducing solution $(SnCl_2 + CdCl_2)$. Mercury was determined by cold vapor atomic absorption spectrometry. Reliability of the results was monitored through Health Canada's interlaboratory comparison program for the determination of Hg in human hair.

Fishing techniques, collection of fish samples, and mercury determination. Together with the collection of a limited number of fish samples during the food enquiries, we organized fishing expeditions in close collaboration with native residents of the four villages in order to use their fishing techniques [nets, harpoons, *nivrées* (rotenone poisoning), and fish lines] and to obtain representative samples of the fish species consumed. All the fish caught were used for Hg analysis: small fish (< 15 g, fw) were entirely preserved; larger fish were dissected on site to collect samples from different organs (dorsolateral skeletal muscle, liver, gill arches, kidneys, stomach, intestine, brain, and gonads) for more complete ecotoxicologic studies on Hg uptake routes and organotropism according to the trophic status. We measured standard length and weight of each fish. Due to severe climatic conditions and the absence of a

continuous electricity supply in the villages, we used a freezer powered by a generator, which provided satisfactory storage for the biological samples. The temperature for shipping the samples from Cayenne to France was ensured by using dry ice, with final storage in the laboratory at -20° C. For total Hg determination, biological samples (< 0.5 g, fw) were first digested by pure nitric acid (3) mL) in a pressurized medium (borosilicate glass tubes) at 95°C for 3 hr. Digestates were then diluted up to 20 mL with ultra-pure water. After mixing, subsamples were used for Hg determination with a SETAC M6000 spectrometer (Varian, Victoria, Australia); this flameless atomic absorption spectrometer was optimized for Hg dosage. An intercalibration exercise was carried out with the CNEVA (Centre National d'Etudes Vétérinaires et Alimentaires, Paris, France) on eight samples of fish muscle. Results showed good levels of agreement between the two laboratories (R =0.98; data not shown).

We selected a limited number of fish muscle samples for MMHg determination, using Bloom's procedure (12) and incorporating several improvements by using a dual circuit on a semi-automatic system: MMHg was determined by cold vapor atomic fluorescence spectrometry (Brooks Rand Ltd., Seattle, WA, USA), after KOH/methanol digestion at 70°C followed by aqueous phase ethylation (NaBEt₄), isothermally controlled gas chromatography at 95°C and 800°C electrothermal atomization. Detection limits for total Hg and MMHg were based on three standard deviations from blank measurement: detection limits for biota on a dryweight basis were 1.4 and 0.5 ng/g, respectively. All biota concentrations are reported on a dry-weight basis (60°C over 2 days). The average dry:fresh weight ratio for fish muscle samples was 0.20 ± 0.01 .

Data treatment. The data collected were treated using descriptive and analytical statistics. The collection and analysis of the data were conducted with Epi-Info (Centers for Disease Control and Prevention, Atlanta, GA, USA), Excel (Microsoft, Redmond, WA, USA), and SAS (SAS Institute, Cary, NC, USA) software. The hair Hg distribution corresponds to the concentrations obtained from all the people in March (n =235). A study of seasonal variations was conducted on a subset of 87 persons in March and November for measurement of total Hg. The fish data were analyzed by multifactorial correspondence analysis with three variables: Hg total concentration in the muscle, fish species, and trophic level of the species (herbivorous, detritivorous, omnivorous, fructicarnivorous, carnivorous) for each individual fish collected. This multivariate analysis was followed by Ward's hierarchical clustering using factorial coordinates. These data treatments were carried out using the SPAD 3.5 software (13). We performed a two-way analysis of variance (ANOVA) to reveal the effects of the two variables, food regime and fish species, on Hg concentrations in the muscle. Based on a one-way ANOVA (site factor), a comparative analysis of Hg concentrations among the three sites (Antecume Pata, Twenke/Taluhen, and Cayode) was carried out for the species *Pseudoancistrus barbatus* (Statistica, version 5; StatSoft, Maisons-Alfort, France), on length-adjusted Hg concentrations.

Results and Discussion

Population exposure to mercury. The distribution of total Hg concentrations in the 235 hair samples analyzed is presented in Figure 2. The average concentration was 11.4 ± 4.2 $\mu g/g$ (mean \pm SD). This value corresponds to high exposure; 57.4 % of the subjects had Hg levels above the safety limit determined by WHO (10 μ g/g), with a maximal value of $27.2 \mu g/g$. These results confirm the data obtained in 1994 on a more limited number of samples in the same Amerindian villages (11). In 1994, the average hair mercury concentration was approximately $3 \mu g/g$ in the entire native Guianese population and 1.7 $\mu g/g$ among French people from the European continent living in French Guiana.

Different factors influenced the hair mercury concentrations: age, hair length, body fat, and place of residence, but not sex (data not shown). The median concentrations were the same across the different age groups, except for children under 1 year of age, who had lower Hg levels. Nevertheless, the distribution of Hg among the Wayana children under 1 year of age was well beyond that of the non-Amerindian inhabitants of Guiana; almost all Amerindian children this age had Hg levels 5 μ g/g. Mercury concentrations in hair were weakly but significantly correlated with the body mass index (r = 0.18, p < 0.01). The concentrations were also weakly correlated with the length of the hair sample (r =0.13, p < 0.05). Results show a good correlation between Hg concentrations in the two seasons (r = 0.68, p < 0.001). The difference in mercury was not statistically significant, although, on average, the concentrations were approximately 7% higher in November than in March. Differences were also observed between the study sites. In people living in Cayode, located on the Tampok River, Hg concentrations were slightly higher than in the people living farther upstream along the Maroni River ($12 \pm 3.5 \mu g/g$ at Cayode, 11.1 \pm 4.2 µg/g at Twenke/Taluhen, and 11.2 µg/g at Antecume Pata). The average Hg concentrations for Cayode remain statistically higher than the others, even after taking into

consideration age, hair length, and body fat (p < 0.05). We note that recent gold-mining activities are mostly on the Tampok River and its terrestrial basin.

Significant Hg levels have been found in populations living in regions not exposed to human polluting activities, but who eat large amount of fish. In Papua-New Guinea, Hg levels in the hair around 4.2 μ g/g were reported in villagers on the coast, whereas those living 25 km inland had values around 1 μ g/g (*14*). Feng (*15*) reported an average concentration of 4.6 μ g/g in the inhabitants of Tokushima, Japan, on the coast.

Several studies have shown that Hg levels in hair are higher in residents of areas contaminated by mercury than in residents of uncontaminated regions. Our results are in agreement with data from the Amazon Basin in Brazil. Hair mercury concentrations from four riverside communities in the southeastern part of the Amazon (Rio Tapajos and Rio Amazonas) had overall geometric means of 11.0 mg/g and 11.6 mg/g for children aged 7–12 years (n =351) and their mothers (n = 135), respectively (9). Lebel et al. (6), Guimaraes et al. (16), and Lacerda and Salomons (1) revealed similar concentrations but with wide variations depending on the site, proximity to gold-mining zones, and population characteristics, particularly the relative importance of fish in the diet. For example, inhabitants in isolated communities, on the lakes of the Amapa coastal plain in the Amazon, have average concentrations of Hg in hair ranging from 6.8 μ g/g (gold-mining village with diversified diet) to 35 µg/g (Duas Bocas Lake residents) (16). It is interesting that the limnologic criteria were similar in the diverse areas, but the origin and importance of their Hg sources were different.

Analysis of Hg chemical forms in hair samples from 27 individuals living in the

four Amerindian villages shows that 8.5% of the metal burdens are in the inorganic form. Mercury in human hair samples was predominantly in the form of MMHg, even near gold-mining areas (1,17). This high percentage of the methylated form must be linked to the exposure conditions of individuals, based mainly on the ingestion of fish muscle, as this tissue is characterized as having a high percentage of MMHg, close to 95%, especially in carnivorous species (18,19).

Contamination levels in fish. During the two study periods, 270 individual fish from 48 species were collected (Table 1). Total Hg concentrations measured in the muscle samples showed marked differences between species, according to their feeding regime and their position in trophic networks. Herbivorous species, such as Acnodon oligacanthus and Myleus rubripinnis, were characterized by low average Hg concentrations of 49 and 65 ng/g [dry weight (dw)], respectively. Conversely, piscivorous species, such as Pseudoplatystoma fasciatum and Hoplias aimara, had average contamination levels in the muscle of 4,700 and 2,910 ng/g (dw), respectively. Overall, 14.5% of the fish analyzed exceeded the safety limit applied in the United States, Canada, and Brazil of 500 ng/g (fw) or 2,500 ng/g (dw).

The multifactorial correspondence analysis highlights an obvious separation of the five feeding regimes considered (herbivorous, detritivorous, fructicarnivorous, omnivorous, and carnivorous). These are associated with different levels of Hg in muscle (Figure 3). The two-way ANOVA confirmed a significant effect of the two variables "food regime" and "fish species" on the Hg concentrations in the dorsal muscle (p < 0.001). The hierarchical clustering allows the definition of five classes:



Figure 2. Distribution of mercury concentrations in hair among the Wayana population (235 samples).

r. . . .

- Class 1: Ninety-four percent of the carnivorous fishes belong to this class, and 98% of individual fish from this class are carnivorous. Eighty-six percent of individual fish from this class have Hg concentrations in muscle between 1,425 and 8,736 ng/g (dw). In addition, all specimens from five species are included in that class: *Hoplias aimara* (n = 21), *Cynodon meionactis* cf. *gibbus* (n = 16), *Serrasalmus humeralis* (n = 10), *Ageneiosus brevifilis* (n = 8), and *Potamotrygon hystrix* (n = 6).
- Class 2: Two species, *Myleus rhomboïdalis* (n = 7) and *Serrasalmus striolata* (n = 6), belong to this class. They have a fructicar-nivorous feeding regime.
- Class 3: Ninety percent of fish assumed to be omnivorous belong to this class. Their Hg concentrations in the muscle range from 256 to 556 ng/g (dw). Five species are exclusively associated with this class: *Doras micropoeus* (n = 18), *Platydoras costatus* (n = 18)

11), Semaprochilodus varii (9), Aequidens guianensis (n = 9), and Brycon pesu (n = 6).

- Class 4: Sixty-one percent of detritivorous fish are included in this class, and their Hg concentrations in muscle range from 116 to 242 ng/g (dw). Two species are exclusively associated with this class: *Pseudoancistrus barbatus* (n = 45) and *Hemiancistrus median* (n = 53).
- Class 5: One hundred percent of herbivorous fish are included in this class, which is characterized by Hg concentrations in muscle of 8 to 115 ng/g (dw). Two species are exclusively associated with this class: *Myleus rubripinnis* (n = 19) and *Acnodon oligacanthus* (n = 10).

Despite the clear pattern shown by this analysis, the difficulty in identifying the feeding regime of fish must be taken into account. Basic knowledge of fish communities in French Guiana is relatively poor, and feeding regimes can change with numerous factors such as age and developmental stage, hydrologic conditions, and habitat characteristics.

To estimate the Hg intake, the average concentration measured in the muscle samples was retained for all nonpiscivorous species. For *Hoplias aimara*, two weight classes were defined, < 1,700 g (fw) and \geq 1,700 (fw), with an average Hg concentration of 1,960 and 3,960 ng/g (dw), respectively. For the majority of species collected, contamination levels in fish muscle were correlated to a limited extent to fish weight, even when the weight ranges were generally great (Figure 4). For carnivorous species such as Hoplias aimara or Cynodon meionactis, a positive correlation was observed, but the weight impact is clearly less marked than in results obtained with predatory fish collected in numerous hydrosystems in the Northern Hemisphere (United States, Canada, Sweden) (20-22). On the other hand, our data are in agreement with the results from fish collected

Table 1. List of the 48 fish species collected in the Haut-Maroni, French Guiana	a, ranked according to their relative contributions ((%)) to total Hg	intake.
--	---	-----	---------------	---------

Family	Species	Wayana name	No. of fish	[Hg] muscle ng/g, dw (mean ± SD)	Percent of total flesh consumed	Percent Hg intake
Pimelodidae	Pseudoplatystoma fasciatum	Huluwi	1	4,700	7.5	27.02
Erythrinidae	Hoplias aimara (> 1,700 g)	Aimara	21	3,965 ± 1,045	7.2	22.02
Ageneiosidae	Ageneiosus brevifilis	Mitala	8	2,640 ± 1,332	5.5	11.12
Doradidae	Doras micropeus	Agonosu	18	1,167 ± 318	11.2	9.98
Serrasalmidae	Serrasalmus rhombeus	Piraïe/Pene	3	1,870 ± 1,000	4.5	6.43
Erythrinidae	<i>Hoplias aimara</i> (< 1,700 g)	Aimara	21	1,955 ± 925	3.5	5.19
Doradidae	Platydoras costatus	Hoké	11	518 ± 206	8.1	3.2
Characidae	Cynodon meionactis (cf. gibbus)	Haïkané	16	4,407 ± 2,041	0.9	2.9
Curimatidae	Prochilodus reticulata	Kulumata	4	432 ± 97	7.9	2.6
Serrasalmidae	Myleus tometes/M. rhomboidalis	Coumaru/Watau	7	103 ± 122	15.9	1.25
Characidae	Cynopotamus essequibensis	Elémaké/Olémaké	2	1,440 ± 525	1	1.13
Characidae	Astyanax/Moenkhausia spp.	Otululu/Opi/Yaya	5	1,040 ± 415	0.9	0.74
Anostomidae	Leporinus friderici	Talani	3	630 ± 421	1.5	0.7
Electrophoridae	Electrophorus electricus	Alimina	1	495	1.8	0.68
Curimatidae	Semaprochilodus varii	Alumasi	9	398 ± 79	1.9	0.58
Sternopygidae	Sternopygus macrurus	Mitoë/Mapala	2	1,110 ± 745	0.6	0.5
Anostomidae	Leporinus fasciatus	Ciaomouné	3	1,524 ± 426	0.4	0.46
Loricariidae	Pseudoancistrus barbatus	Pële	45	155 ± 53	3	0.36
Sciaenidae	Pachypops furcraeus	Kupi/Masao	1	370	1.3	0.36
Characidae	Brycon falcatus	Molokoïme	1	217	2	0.34
Anostomidae	Leporinus lebaili (cf. despaxi)	Walak	1	865	0.5	0.31
Pimelodidae	Pimelodus ornatus	Liku	1	1838	0.2	0.31
Rhamphichthyidae	Rhamphichthys rostratus/Hypopomus artedi	Mapalaime	2	1,108 ± 746	0.3	0.3
Characidae	Triportheus rotondatus	Kampuluka	1	541	0.5	0.22
Cichlidae	Geophagus harreri	Hawa Hawa	1	256	1	0.19
Cichlidae	Cichla ocellaris	Matawalé	2	148 ± 12	1.4	0.16
Serrasalmidae	Acnodon oligacanthus	Laku	10	49 ± 25	3.6	0.14
Hemiodidae	Hemiodopsis huraulti	Walé Walé	3	514 ± 300	0.3	0.13
Serrasalmidae	Myleus rubripinnis	Pasina	19	65 ± 60	2.1	0.1
Characidae	Brycon pesu	Enké/Entke	6	622 ± 349	0.2	0.09
Hemiodidae	Bivibranchia bimaculata/Hemiodus unimaculatus	Opui/Epui	6	250 ± 240	0.3	0.08
Pimelodidae	Pimelodella gracilis	Kasibë/Kawayéma	5	280 ± 85	0.4	0.08
Cichlidae	Crenicichla saxatilis	Kolopimpé	1	715	0.1	0.05
Cichlidae	Aequidens maroni/Geophagus surinamensis /Guianacara owroefi	Awalipa	9	385 ± 290	0.2	0.05
Loricariidae	Hemiancistrus medians	Mili	7	151 ± 41	0.5	0.05
Serrasalmidae	Serrasalmus humeralis/S. striolatus	Piraïe/Pene (< 100 g)	16	405 ± 40	0.2	0.05
Anostomidae	Leporinus maculatus	Halanaé/Kalanalé	2	316 ± 35	0.2	0.04
Curimatidae	Curimata cyprinoides	Pohakë	3	301 ± 40	0.2	0.04
Characidae	Bryconops affinis	Wiwi	4	269 ± 65	0.2	0.03
Loricariidae	Hypostomus plecostomus	Kawawa	2	173 ± 98	0.1	0.02
Loricariidae	Loricaria sp.	Lapipi	2	65 ± 5	0.2	0.01
Potamotrygonidae	Potamotrygon hystrix	Sipali	6	950 ± 471	0	0

from several gold-mining sites in the Amazon Basin (16,23,24). It must be emphasized that age determination in tropical fish is difficult, even impossible, using traditional techniques based on scales or otolites, because of minor variations between seasons. The relation between fish weight and length was well defined for the different species collected, with no significant distinction between fish populations from the four sampling sites in the Haut-Maroni area (Figure 4).

The comparison of the length-adjusted Hg concentrations in the fish species *Pseudoancistrus barbatus* among the three sites, based on a one-way ANOVA, confirmed the highest contamination at the Cayode site that was previously observed for hair mercury concentrations [Cayode (163 ng/g, dw) > Antecume Pata (132 ng/g, dw) > Twenke/Taluhen (109 ng/g, dw); p < 0.05].

Monomethyl mercury determination on a limited number of muscle samples from carnivorous species showed that $98 \pm 5\%$ of the metal accumulated in this tissue was in the methylated form. This result is in agreement with the data published by Akagi et al. (*25*) on fish samples from the Madeira River in Brazil (MMHg, 98%).

It is difficult to make a comparative analysis of fish contamination levels from the data published on the Amazon Basin, which actually contains more than 1,300 freshwater species. Moreover, fish species in Brazil do not exist in French Guiana and vice versa. Nevertheless, from the review published by Lacerda and Salomons (1), Hg concentrations measured in the Haut-Maroni area are similar to the majority of Amazon data in gold-mining areas, except for the Madeira and Tapajos rivers, where maximum Hg concentrations of 15,000 µg/g (fw) were reported.

Mercury determinations made on a limited number of game samples (12 in all) show that contamination levels of muscle tissue are lower than those observed in aquatic species. For the majority of terrestrial species (doe, monkey, peccary, and tapir), concentrations are lower than 50 ng/g (dw). Only two armadillos showed concentrations close to 700 ng/g (dw). According to Lacerda and Salomons (1), Hg concentrations in terrestrial vegetation are low, with exceptions occurring in the vegetation growing on railings due to the selection of metal-tolerant forms. Thus, contamination levels in terrestrial food webs are much lower than those observed in the aquatic environment, but more data are needed for a better comprehension of the fate of Hg in terrestrial systems. However, piscivorous terrestrial species, such as caimans or certain bird species (kingfishers), can show high levels of contamination (26). These species are either not eaten by the Wayanas or are at

present rare in the areas near the Haut-Maroni villages. Moreover, this dietary study has shown the marked predominance of fish consumption in the diet, compared to game: in Wayana families, only 10% of the person × days data are based on meals composed exclusively of meat.

Estimate of dietary intake of mercury via fish. According to the International Programme on Chemical Safety (5), the



Figure 3. Plan 1-2 of the multifactorial correspondence analysis based on the 270 fish collected, with three variables: Hg concentration in the muscle (ng/g, dw), fish species, and trophic levels. Conc, range of concentrations.



Figure 4. Relationships between total weight and total length (left) and mercury concentration in the muscle and total weight (right) for three fish species, (*A*) *Hoplias aimara* (n = 21), (*B*) *Pseudoancistrus barbatus* (n = 45), and (*C*) *Myleus rubripinnis* (n = 19). R = 0.99 for (*A* left); R = 0.47 for (*A* right); R = 0.99 for (*B* left); and R = 0.98 for (*C* left).

daily intake of MMHg by humans, regardless of the source, is approximately 2.4 µg, and the intake of total mercury is $6.7 \mu g$ (higher when dental amalgam is present). The Wayana population is well beyond these values: adults consume 40–60 µg total Hg per day, and the elderly ingest approximately 30 µg (Table 2; Hg intake per day and per week). As for children, the youngest ingest approximately 3 µg/day, taking into account nursing; those from 1 to 3 years, approximately 6 μ g/day; those from 3 to 6 years of age, approximately 15 μ g/day; and those from 7 to 15 years of age, between 28 and 40 µg/day. Moreover, these results are underestimates because they do not take into consideration the Hg intake from game. Mercury intake was estimated from the fish Hg content and the average daily consumption of fish for the different ages (Table 3). In periods of abundance, the amount of fish consumed could be very high [more than 600 g/day (fw) in some adult males], whereas in times of scarcity these amounts could be relatively low, and more cassava seemed to be eaten to provide sufficient caloric intake. Average daily consumption was highest in men between 15 and 45 years of age (around 340 g/day, fw). The Wayanas are a perfect example of a "fishing civilization," like Brazilian populations living in isolated villages on river banks or lake shores in the Amazon Basin: recent investigations on family members of fishermen from Duas Bocas Lake shore (State of Amapa, Brazil) show that most subjects reported taking 14 fish meals per week (16). Similar diets have been described among the Cree Indians in Northern Quebec, resulting in high levels of Hg impregnation due to the consumption of fish caught around hydroelectric reservoirs (27).

The Food Agricultural Organization/ WHO Expert Committee on Food Additives recommends that the weekly intake not exceed 300 µg total Hg, with a maximum of 200 µg MMHg, which represents 3.3 µg/kg for a 60-kg male (5). Approximately 95% of MMHg is absorbed through the intestinal barrier (28); the biological half-life of organic Hg in humans is estimated at between 70 and 80 days (29). Dietary intake of Hg in the Wayana population is above this safety limit. The Hg intake data are significantly correlated with the corresponding Hg hair data (r = 0.36, p < 0.01, data not shown). The value of 200 µg/L blood (or 50 µg/g hair) was associated with a low risk (5%) of severe neurologic disorders in the adult. Nevertheless, fetuses and young children can present neurologic disorders at exposure levels at which adults are not affected. WHO recommends instituting epidemiologic studies in chidren exposed *in utero* to levels of MMHg in maternal hair of the order of $10-20 \ \mu g/g$ to establish the effects on health by means of psychomotor and behavioral tests.

We found that four carnivorous species corresponding to 28% of the dietary intake of fish contribute to 72% of the amount of Hg consumed. These species are Pseudoplatystoma fasciatum (27% of the Hg dietary intake), Hoplias aimara (27%), Ageneiosus brevifilis (11%), and Serrassalmus rhombeus (6.5%). Cynodon meionactis, which along with P. fas*ciatum* has the highest Hg concentrations, was hardly eaten at all (especially by children) because these fish contain a large number of bones. These data are extremely important for the implementation of recommendations for reducing Hg intake via the ingestion of fish. The two species that are consumed in the greatest amounts, Myleus rhomboidalis/tometes (12.7%) and *Doras micropeus* (11.2%), are not contaminated to a very high degree (100 and 1,160 μ g/g, dw, respectively).

In parallel with our dietary study, each child from the four villages was given a neurologic examination and neurobehavioral development tests (*30*). Results revealed significant links between impregnation levels in the children and the presence of some neurologic and behavioral deficits. Gold-mining activities seem to play a role in the contamination of this population, although they are not easily identified because it is difficult to estimate the contribution of past and recent gold mining. Nevertheless, our results showed that hair and fish Hg levels were significantly higher in Cayode, the isolated village where gold mining now occurs, than in other villages. In addition, the results from a study on Hg exposure of the French Guiana population conducted in 1994 (11) showed that Amerindians from Camopi on the Oyapok River (border of Brazil) who live in an area without gold mining and eat as much fish as the Wayanas from the Upper Maroni region had hair Hg levels two times lower than the Wayana population. Nevertheless, the role of natural Hg in this contamination is still under discussion, especially the metal accumulated in soils and transferred to the aquatic systems via natural and anthropogenic erosion (2).

Conclusion

This study showed the Wayana population from French Guiana to have high exposure to mercury at levels exceeding WHO recommendations. Levels of mercury, evaluated through measurements in hair and food, are related to high consumption of fish, especially carnivorous species that have high concentrations of mercury. These results are of the same order of magnitude of those found in similar situations for countries such as Brazil, where gold mining is particularly developed. At the present time, a close collaboration between local populations, public health authorities, and scientists should allow the development of concrete recommendations as well as the adoption of measures, in order to solve this problem without causing human and environmental perturbations, in

Table 3. Average daily consumption of flesh by

 Wayana family members, according to age and sex.

Table 2. Average daily and weekly mercury uptake by Wayana family mem	bers via fish consumption
according to age and sex.	

			Mercury				
	Average		Mean ± SD	Median	Mean		
Age group	age	No.	(µg/day)	(µg/day)	(µg/week)		
< 1 year	0.84	19	1.2 ± 1.0	1.0	8.4		
1-3 years	1.8	55	6.6 ± 6.1	4.2	46.4		
3–6 years	5.4	96	14.3 ± 17.1	9.1	100.7		
7–10 years	8.7	166	27.7 ± 40.8	14.0	191.6		
10–14 years	12.4	83	37.7 ± 43.1	16.1	263.6		
15–25 years							
Men	19.3	102	47.9 ± 50.9	28.3	335.0		
Women	20.3	124	41.4 ± 59.9	23.2	289.5		
25–45 years							
Men	37	101	61.3 ± 66.0	34	428.8		
Women	33.5	89	41.3 ± 50.1	21.1	288.8		
> 45 years							
Men	58.1	63	28.8 ± 56.3	14.4	210.5		
Women	56.6	42	29.1 ± 33.2	12.9	203.9		

Average daily consumption ± SD Age group No. (SP, g/day, fw) 20 ± 9 <1 year 5 1-3 years 14 47 ± 18 3-6 years 27 116 ± 69 7-10 years 34 173 ± 79 11-14 years 29 195 ± 110 15-25 years 16 307 ± 185 Men Women 20 262 ± 161 Total 282 ± 171 36 26-45 years 27 Men 372 ± 193 Women 27 262 ± 138 Total 54 317 ± 175 > 45 years 19 Men 162 ± 158 9 Women 140 ± 116 Total 28 155 ± 144 SP, standard portion.

parallel with a health monitoring system to evaluate the effectiveness of recommended preventive actions.

REFERENCES AND NOTES

- 1. Lacerda LD, Salomons W. Mercury from Gold and Silver Mining: a Chemical Time Bomb? Berlin:Springer, 1998.
- Roulet M, Lucotte M, Farella N, Serique G, Coelho H, Sousa Passos CJ, Jesus da Silva E, Scavone de Andrade P, Mergler D, Guimaraes JRD, et al. Effects of recent human colonization on the presence of mercury in Amazonian ecosystems. Water Air Soil Pollut 112:297–313 (1999).
- Morel FMM, Kraepiel AML, Amyot M. The chemical cycle and bioaccumulation of mercury. Annu Rev Ecol Syst 29:543–566 (1998).
- Malm O, Branches FJP, Akagi H, Castro MB, Pfeiffer WC, Harada M, Bastos WR, Kato H. Mercury and methylmercury in fish and human hair from the Tapajos River basin, Brazil. Sci Total Environ 175:141–150 (1995).
- WHO. Methylmercury. Environmental Health Criteria 101. Geneva:World Health Organization, 1990.
- Lebel J, Mergler D, Lučotte M, Amorim M, Dolbec J, Miranda D, Arantes G, Rheault I, Pichet P. Evidence of early nervous system disfunction in Amazonian populations exposed to low levels of methylmercury. Neurotoxicology 17:157–168 (1996).
- Nielsen A, White RF, Nielsen U, Cleary D, Grandjean P. Psychomotor performance of methylmercury exposed children from two Amazonian villages. In: Proceedings of the International Conference Health Effects of Mercury Exposure, 22–25 June 1997, Torshavn, Feroe Islands. 1997. Odense, Denmark:Odense University, 1997:59.
- Grandjean P, Weihe P, White RF, Debes F, Araki S, Murata K, Sorensen N, Dahl D, Yokoyama K, Jorgensen PJ. Cognitive deficit in 7-year-old children with prenatal exposure to methylmercury. Neurotoxicol Teratol 19:417-428 (1997).
- Grandjean P, White RF, Nielsen A, Cleary D, de Oliveira Santos EC. Methylmercury neurotoxicity in Amazonian children downstream from gold mining. Environ Health Perspect 107:587–591 (1999).
- 10. Picot JC, Foucher JL, Wagner R. Or, mercure et environnement: production aurifère et mercure utilisé de l'origine à

nos jours. BRGM R37837. Cayenne, French Guiana:Bureau de Recherches Géologiques et Miniéres, 1993.

- Cordier S, Grasmick C, Paquier-Passelaigue M, Mandereau L, Weber JP, Jouan M. Mercury exposure in French Guiana: levels and determinants. Arch Environ Health 53:299–303 (1998).
- Bloom NS. Determination of picogram levels of methylmercury by aqueous phase ethylation, followed by cryogenic gas chromatography with cold vapour atomic fluorescence detection. Can J Fish Aquat Sci 46:1131–1140 (1989).
- Lebart L, Morineau A, Lambert T, Pleuvret P. SPAD version 3; Système pour l'analyse des données. Saint-Mandé, France:Centre International de Statistique et d'Informatique Appliquées, 1996.
- 14. Suzuki T. Advances in Mercury Toxicology. New York:Plenum Press, 1991;459–483.
- Feng Q, Suzuki Y, Hisashige A. Hair mercury levels of residents in China, Indonesia and Japan. Arch Environ Health 53:36–43 (1998).
- Guimaraes JRD, Fostier AH, Forti MC, Melfi JA, Kehrig H, Mauro JBN, Malm O, Krug JF. Mercury in human and environmental samples from two lakes in Amapa, Brazilian Amazon. Ambio 28:296–301 (1999).
- Malm O, Guimaraes JRD, Castro MB, Bastos WR, Viana JP, Branches FJP, Silveira EG, Pfeiffer WC. Follow-up of mercury levels in fish, human hair and urine in the Madeira and Tapajos basins, Amazon, Brazil. Water Air Soil Pollut 97:45–51 (1997).
- Bloom NS. On the chemical form of mercury in edible fish and marine invertebrate tissue. Can J Fish Aquat Sci 49:1010–1017 (1992).
- Boudou A, Ribeyre F. Mercury in the food web: accumulation and transfer mechanisms. In: Mercury and Its Effects on Environment and Biology, Vol 34 (Sigel A, Sigel H, eds). New York:Marcel Dekker, 1997;289–320.
- Brouard D, Doyon JF, Schetagne R. Amplification of mercury concentrations in lake whitefish downstream from the La Grande 2 Reservoir, James Bay, Québec. In: Mercury Pollution: Integration and Synthesis (Watras CJ, Huckabee JW, eds). Boca Raton, FL:Lewis Publishers, Inc., 1994;369–380.
- Lindqvist O. Mercury in the Swedish Environment. Dordrecht, Netherlands:Kluwer Academic Publishers, 1991.

- Wiener JG, Spry DJ. Toxicological significance of mercury in freshwater fish. In: Interpreting Environmental Contaminants in Animal Tissues (Heinz G, Beyer N, eds). Boca Raton, FL:Lewis Publishers, Inc., 1995;36–52.
- Roulet M, Lucotte M, Dolbec J, Gogh YF, Pelaja JRP, Silva DS, Lebel J, Mergler D, Guimaraes JRD. Mercury bioaccumulation in two fish communities on the Tapajos River, Brazilian Amazon. In: Proceedings of the 5th International Conference Mercury as a Global Pollutant, 23-28 May 1999, Rio de Janeiro, 1999. Rio de Janeiro, Brazil:Center for Mineral Technology, 1999;232.
- Meili M, Malm O, Guimaraes JRD, Padovani C, Forsberg BR, Viana JP, Silveira EG. Bioaccumulation of mercury in tropical river food webs (Amazon): similar patterns as in boreal lakes. In: Proceedings of Fourth International Conference on Mercury as a Global Pollutant, August 1996, Hamburg, Germany, 1996. Geesthacht, Germany:GKSS-Forschungszentrum, 1996;123.
- Akagi H, Malm O, Branches FJP. Human exposure to mercury due to goldmining in the Tapajos river basin, Amazon, Brazil: speciation of mercury in human hair, blood and urine. Water Air Soil Pollut 80:85–94 (1994).
- Aula I, Braunsweiller H, Leino T, Malin I, Porvari P, Hatanaka T, Lodenius M, Juras A. Levels of mercury in the Tucurui reservoir and its surrounding area in Para Brazil. In: Mercury Pollution: Integration and Synthesis (Watras CJ, Huckabee JW, eds). Boca Raton, FL:Lewis Publishers, Inc., 1994;21–40.
- Verdon R, Brouard D, Demers C, Lalumière R, Laperle M, Schetagne R. Mercury evolution (1978–1988) in fishes of the La Grande Hydroelectric Complex, Québec, Canada. Water Air Soil Pollut 56:405–417 (1991).
- Aberg B, Ekman R, Falk U. Metabolism of methylmercury compounds in man: excretion and distribution. Arch Environ Health 19:478–484 (1969).
- Man Chan H. Metal accumulation and detoxification in humans. In: Metal Metabolism in Aquatic Environments (Langston WJ, Bebianno MJ, eds). London:Chapman & Hall, 1998;415–438.
- Cordier S, Garel M. Risques neurotoxiques chez l'enfant liés à l'exposition au méthylmercure en Guyane française. Paris:Institut Veille Sanitaire, 1999.