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Gold Mining as a Source of Mercury Exposure in the Brazilian Amazon

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Amalgamation has been used for more than 4500 years in mining processes. Mercury has been extensively used in South America by Spanish colonizers for precious metal recovery. It is estimated that between 1550 and 1880, nearly 200,000 metric tonnes of mercury was released to the environment. During the present gold rush, Brazil is first in South America and second in the world in gold production (with 90% coming from informal mining or *garimpos*). At least 2000 tonnes of mercury has been released to the environment in the present gold rush. From the mid 1980s, environmental research has been carried out in impacted Amazon rivers, later followed by human exposure studies. The river basins studied were the Tapajós, Madeira, and Negro, but also some man-made reservoirs and areas in central Brazil. The analyses mainly involved sediments, soil, air, fish, human hair, and urine. The results show high variability, perhaps related to biological diversity, biogeochemical differences in the river basins, and seasonal changes. High mercury values also occur in some areas with no known history of gold mining. The results available document a considerable impact on environmental mercury concentrations and frequent occurrence of human exposure levels that may lead to adverse health effects. © 1998 Academic Press

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INTRODUCTION

Artisanal goldmining is using simple processes to extract gold from secondary or primary ore bodies. Usually secondary deposits or very fine gold particles need mercury for good gold recovery. The amalgamation process was known by the Phoenicians and the Carthagians (2700 BC), who commercialized mercury from the Almadén mines in Spain. Caius Plinius in his "Natural History" (50 AD) describes the mining technique for gold and silver with an amalgamation process that is similar to the one used today in many gold mining areas. Final recovery of fine gold particles extracted was always done through heating or burning of the amalgam with high mercury emissions to the atmosphere in many tropical countries, especially in South America but also in Asia and Africa where a new gold rush is occurring (Lacerda and Salomons, 1997). In South America this procedure has ben used in countries like Brazil, Venezuela, Colombia, Bolivia, French Guyana, Guyana, Ecuador, and Peru since the 1980s.

The current gold rush in South America is the consequence of an increase in the gold prices during the 1970s by a factor of 8 to 10, but also has a strong association with social and economic difficulties, including unemployment, poverty, etc. (Pfeiffer and Lacerda, 1988).

Mercury was extensively used in South America (mainly Mexico, Peru, and Bolivia) by Spanish colonizers for silver and gold recovery. It is estimated that, from the middle sixteenth century (1550) to the middle nineteenth century (1880), nearly 200,000 tonnes of mercury was released to the environment (Nriagu, 1993). A release of 400 tonnes is estimated for colonial Brazil during the same period (Lacerda, 1997).

Since the 1980s, Brazil has ranked as first in South American gold production with annual production from 100 to 200 tonnes per year during the past 20 years 70 (with to nearly 90% coming from informal mining or *garimpos*), followed by Colombia, Venezuela, Peru, and Bolivia. This production would correspond to about 2000 to 3000 tonnes of mercury released into the Brazilian Amazon environment during the present gold rush. These estimates are approximate, since formal gold production and mercury imports were not well registered. Table 1 shows some estimates of mercury released to the

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TABLE 1
Estimated Mercury Input to the Environment Due to Gold and Silver Mining

	Period	Total input (tonnes)	Annual input (tonnes per year)
Spanish Colonial America	1554–1880	196,000	600
All North America	1840–1900	60,000	1,000
Colonial Brazil	1800–1880	400	5
Brazilian Amazon	1979–1994	2,300	150
Venezuela	1988–1997	360	40
Colombia	1987–1997	240	30
Bolivia (Pando Department)	1979–1997	300	20
Philippines	1985–1997	200	26
Tanzania	1991–1997	24	6
China	1992–1997	480	120

Note. Data from Nriagu, 1993; Lacerda and Salomons, 1997; Veiga, 1997, and estimates from the author.

environment due to mining. If total mercury releases are considered, colonial environmental discharges were about 600 tonnes per year, while currently averaging about 150 tonnes per year. Mining of gold and silver during the colonial period utilized mainly primary ores but the current gold rush in the Amazon affects mainly secondary gold deposits (colluvial or alluvial gold reserves) in soils or river sediments, where mercury releases during mining directly involve water bodies.

The total number of miners is probably between 200,000 and 400,000 within the Brazilian Amazon. Half as many are thought to exist in Colombia and Ecuador. For all Latin American countries combined, the estimate is between 543,000 and 1,039,000 miners (Veiga, 1997). During the peak of the gold rush the estimated number of miners in Brazil alone was above 1,600,000 (Pfeiffer and Lacerda, 1988).

Studies on mercury transport to water bodies, its transformation to methylmercury, and the contamination of food chains, including human beings, are quite recent in South American countries. In the middle 1980s environmental research began in the Amazon river basins.

Investigations were mainly performed with sediments, soil, air, fish, human hair, and urine samples. The most well studied river basins are the Tapajós, Madeira, and Negro, but some man-made reservoirs and some areas in central Brazil have also been studied. These results will be briefly summarized here.

RESULTS

Different samples from distinct goldmining areas have helped in understanding the tropical mercury cycle and identified human exposure pathways and possible risks. As a general result in the case of mercury vapor, resulting from burning or reburning of gold amalgams, the main group of humans exposed are the gold dealers in shops rather than the gold miners (*garimpeiros*), who work outdoors. In the case of methylmercury, the riverine populations constitute the group at risk, as they depend on fish as their main source of protein.

The first critical human exposure to mercury results from the burning of the amalgam, when a significant fraction of mercury emissions occur mainly to the atmosphere. Table 2 summarizes the main data on mercury in air from different studies. Very

TABLE 2
Mercury Concentration ($\mu g\ m^{-3}$) in Air from Urban, Rural, or Occupational Exposures

Origin/situation	Average	*N*	Range	Reference
Rondônia – SW Amazon				
Urban, far from reburning areas	—	7	< 0.02– < 0.66	Malm *et al.*, 1991
Urban, nearby reburning areas	2.80	8	0.45–7.50	
Occupational exposure during reburning in the dealer's shops	71.40	7	17.50–107.20	
Occupational exposure during burning in the field with various retorts	91.70	6	< 10.24–296	
Occupational exposure during burning in the field without retorts	15,499	6	< 42.29–59,600	
Outlets with contrary water reflux for mercury recovery system	1,280	5	< 120–5,162	
Air samples close to waterfalls	—	3	< 0.14–0.50	
Poconé City (urban area) – Central Brazil				
Urban, nearby reburning areas	—	10	< 0.14–1.86	Marins *et al.*, 1991
Above tailings	0.08			Tumpling *et al.*, 1995
Alta Floresta City – South Amazon				
Occupational exposure during reburning in the dealer's shops	5.14	86	0.07–40.60	Hacon *et al.*, 1995
Urban, far from reburning areas	0.61	152	0.01–5.79	

high air mercury concentrations are found when burning amalgam in open air. Fortunately this occurs at a low frequency (about once a week) in connection with gold mining. High mercury air values were found also in indoor areas in gold dealer shops even without reburning activities. In our studies (Malm *et al.*, 1995b) an average value of 71.50 $\mu g \cdot m^{-3}$ was obtained, i.e., above the 50 $\mu g \cdot m^{-3}$ maximum limit established by WHO (1991) for occupational exposure. A more recent study (Hacon *et al.*, 1995) showed lower average values, perhaps due to decreased production rates.

Inhaled metallic mercury is readily absorbed through lungs (more than 85%) and after some time in the blood stream, part is oxidized and accumulated in the kidneys (WHO, 1991). The mercuric ion is excreted through urine, which is the best indicator of metallic mercury exposure and inorganic mercury body burden (WHO, 1991).

Reburning of amalgams (bullion) in gold dealer shops in cities and villages contaminates indoor areas and the vicinities. The highest average values were observed in urine samples from people working under indoor conditions with little ventilation or in reburning rooms with air conditioning. Urine from gold shop workers contained mercury mostly in inorganic form (Akagi *et al.*, 1995a). Higher values were also observed in gold traders compared with miners (Cleary *et al.*, 1994) in several goldmining areas along the Tapajós basin.

Of 78 urine samples collected from gold shop workers from Santarém during 1991 and 1992 some showed extremely high concentrations (up to 1200 μg/L), very much above the limit of 50 μg/L established by WHO (1991) (median value 115 μg/L), but in 1994 (with median value of 5.2 μg/L) the concentrations were considerably lower. The reduction of mercury values is a likely consequence of the mining decline, thus reducing the number of primitive gold shops, but perhaps also is the result of recommendations by a local physician, Dr. Fernando J. P. Branches. In Alta Floresta the values were significantly lower (up to 70 μg/L; median value of 11.7 during the period from 1991 to 1995), but this is a young city with newer and better ventilated shops (Malm *et al.*, 1997).

Fish is the main source of methylmercury to humans. Piscivorous or carnivorous fish from several goldmining areas show higher mercury concentrations followed by fish from lower trophic levels such as omnivorous, detritivorous, and herbivorous species.

The former should be monitored as indicators of aquatic system contamination as well as for public health reasons (Pfeiffer *et al.*, 1993). There are around 200 different fish species of commercial importance in each river basin. Fishing activities as well as fish fat and protein contents (Val and Almeida-Val, 1995) vary significantly according to the season (Malm *et al.*, 1995a; Lebel *et al.*, 1997).

Fish from pristine areas usually present levels lower than 0.2 μg mercury $\cdot g^{-1}$ wet wt in rivers. In contaminated areas predatory fish can reach levels of 2 to 6 $\mu g \cdot g^{-1}$ or even more. The piscivorous and carnivorous group from the main mined Amazon river basins usually present mercury average values above the 0.5 $\mu g \cdot g^{-1}$ maximum limit established by the Brazilian Legislation (Brasil, 1975). Table 3 summarizes mercury data on carnivorous and piscivorous fish from Amazon areas.

In some piscivorous/carnivorous fish species, a tendency for increasing mercury concentrations with increasing fish weight was observed. Even for this group, a high variability was observed even in the same species of fish collected at the same time in the same net (Meili *et al.*, 1997). A mercury seasonal

TABLE 3

Mercury Concentrations in Carnivorous or Piscivorous Fish from Dierent Amazon Areas

Origin	No. of species	No. of samples	Average ($ng \cdot g^{-1}$)	Range ($ng \cdot g^{-1}$)	Reference
Madeira River	50	370	850	165–3920	Malm *et al.*, 1997
Madeira River	22	154	665	60–3960	Gali, 1997
Madeira River	—	251	638	11–500	Barbosa *et al.*, 1995
TapajósRiver	23	118	498	25–5960	Malm *et al.*, 1997
Tapajós River	12	212	499	46–2200	Uryu, 1996
Tapajós River	19	73	511	132–1354	Lebel *et al.*, 1997
Tapajós River	09	85	723	120–3580	Hacon, 1996
Negro river	18	113	780	226–4231	Malm *et al.*, 1994
Tucuruí Reservoir	8	121	1300	200–5900	Porvari *et al.*, 1995
Balbina Reservoir	6	27	371	49–1103	Malm *et al.*, 1996b,

Note. N total = 1524.

TABLE 4
Total Mercury Concentrations in Hair from Different Amazon Areas

Origin	No. of samples	Average ($\mu g \cdot g^{-1}$)	Range	Reference
Madeira river	169	8.98	0.22–71	Malm *et al.*, 1996
Madeira river	242	17.2	303	Barbosa et al., 1995; Boischio *et al.*, 1995
Tapajós river	432	16.76	0.7–176	Malm *et al.*, 1996; Akagi *et al.*, 1995b
Tapajós river	96	13.2	1.0–142	Lebel *et al.*, 1997
Kayapó Indians	419	8.00	37	Barbosa *et al.*, 1995
Negro river	154	75.5	171	Forsberg *et al.*, 1995
Tucuruí reservoir	125	35	0.9–240	Leino and Lodenius, 1995
Balbina Reservoir	58	5.78	1.15–26	Malm *et al.*, 1995b
Yanomami Indians	162	3.61	1.40–8.14	Castro *et al.*, 1991

Note. N total = 1782.

variability was suggested for some piscivorous species, but more data are still needed for several species in different river basins or reservoirs.

Human hair is accepted as the best indicator for assessment of contamination in populations exposed to methylmercury (WHO, 1990). Table 4 presents some main results on mercury in human hair samples from several areas. Again, in general there is good agreement among the different investigations, and the total number of samples is quite impressive. Several groups present average values within the range associated with a risk to pregnant women (WHO, 1990). At the same time a high variability (as in fish) was observed in hair mercury even within a family or a group living in the same household. In general, a relation between hair mercury and fish ingestion is observed (Grandjean *et al.*, 1993), but variability in the same family may be high. High variability was also observed among strands of human hair (Akagi *et al.*, 1994; Kehrig *et al.*, 1997) from the same house, which could be explained by seasonal changes but also by the high variability observed in fish.

DISCUSSION

Recent mercury emissions in the Amazon are important because of the new environmental circumstances of mercury releases rather than because of their magnitude. The complex ecosystem associated with tropical rainforest river basins with its enormous biodiversity is the new challenge for the understanding of the mercury cycle and for evaluating mercury health risks.

In Brazil estimation of releases of mercury per area or river basin does not correlate with the levels found in biota. The Tapajós river basin has been prospected for a longer time than the Madeira (30 and 15 years, respectively), but mercury concentrations in fish of the same species and weight range are higher in the latter basin. Elevated concentrations are found in the Tapajós river basin only in its upper reaches. The Negro river, with no significant history of gold mining activities, presents quite high values in fish samples.

Since no input of mercury is known for sure, the high levels of mercury in the Negro river samples could be due to certain natural processes of contamination some centuries ago. Where the mercury released during the Spanish Colonial period is now still a question. The importance of forest burning on mercury emissions has also been a matter of discussion (Veiga, 1997).

Changes in fish alimentary habits in riverine populations are a factor that must be considered in human risk evaluation. Nevertheless, clinical evaluation (if possible showing changes from the past) must support any decision in this respect. Attention must be paid to avoiding political and dishonest bad uses of this information.

Atmospheric mercury investigations are necessary for a better understanding of the dispersion and deposition of mercury in the surroundings of urban and rural areas in the Amazon (Malm *et al.*, 1996a). Further, better knowledge on mercury methylation and the rate controlling factors is essential for understanding food chain contamination in this tropical environment.

Concentration of methylmercury in sediments, water, and fish is probably influenced by several parameters, such as mercury concentration, microbiological activity, organic matter, presence of methyl group donors, pH, redox potential, and O_2, among others.

The experiments done so far indicate detectable methylation in bottom sediments of rivers such as the Madeira and Tapajós and higher rates in forest streams. The highest methylation rates were recently found in environments that are typical of tropical aquatic systems, such as the surface of seasonally flooded forest soils and especially the root zone of the "floating meadows" formed by the aquatic vegetation, where up to 35% of added mercury chloride was converted to methylmercury after short-term experiments (Guimarães *et al.*, 1997).

The high variability of mercury values observed in carnivorous or piscivorous fish is making it difficult to generate advice on fish ingestion to the local population. Perhaps a small number of top predator species could be blacklisted, but decisions must be made area by area and reviewed regularly.

High mercury values were observed in several individual cases as well as in some populations. If neurological damage occurs (Lebel *et al.*, 1996), its extent deserves to be documented to provide a deeper understanding of health risks and as a basis for dietary recommendations.

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