

EFFECTS OF RECENT HUMAN COLONIZATION ON THE PRESENCE OF MERCURY IN AMAZONIAN ECOSYSTEMS[#]

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Abstract. Three Hg sources were characterised and mass balance calculations were used to determine their relative contributions to the contamination of the Amazonian environment. About an order of magnitude more Hg is emitted to the atmosphere by goldmining activity than by the burning of forest biomass. However, anthropogenic atmospheric Hg cannot account for the high Hg burdens found in terrestrial ecosystems: deposition of Hg from goldmining sources is estimated to account for less than 3% of the Hg present in the surface horizons of soils. We propose that erosion of deforested soils following human colonization constitutes a major disturbance of the natural Hg cycle. Deforestation thus increases soil Hg mobilisation by runoff, which may explain the increase of Hg burdens in Amazonian aquatic ecosystems in newly colonized watersheds.

Keywords: Amazonia, mercury, soil erosion

1. Introduction

Several studies have shown that the fish diet of the human population living along the shore of the Tapajós River, up to hundreds of kilometers downstream from goldmining areas, may expose individuals to sufficient quantities of methylmercury (MeHg) to result in nervous system dysfunctions (Cleary *et al.*, 1994; Akagi *et al.*, 1995; Lebel *et al.*, 1996, 1997). It is commonly accepted, with no supporting scientific evidence, that the source of this MeHg contamination is the release of Hg by the goldmining activities upstream (Pfeiffer *et al.*, 1993; Cleary *et al.*, 1994; Akagi *et al.*, 1995; Malm *et al.*, 1995). The ongoing Amazonian gold rush is thought to release 130 tons of Hg/yr, roughly half to the rivers and the other half to the atmosphere (Pfeiffer *et al.*, 1993).

[#] The present investigation is part of an ongoing study, the CARUSO project (IDRC-UFPA-UQAM), initiated to determine the sources, fate, and health effects of MeHg in the Lower Tapajós area.



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However, other sources may contribute to Hg contamination of the environment. Recently, Veiga *et al.* (1994) estimated that the burning of forest biomass releases 90 tons Hg/yr into the atmosphere and suggested that these emissions represent the major source of atmospheric Hg over the Amazon region, contributing significantly to the contamination of aquatic ecosystems. This estimate was disputed by Lacerda (1995) who calculated that atmospheric Hg emissions from biomass burning amount to less than 17 tons Hg/yr. However, both these estimates are based on assumed Hg concentrations, not on actual values observed in the various compartments of the Amazonian forest ecosystem.

The impact of slash and burn agriculture is not limited to the combustion of the forest biomass. Deforestation and cultivation usually lead to increased soil erosion and leaching. Since the finding by Roulet and Lucotte (1995) that oxisols accumulate significant quantities of Hg associated with iron oxy-hydroxides, recent work in the Tapajós Basin has focussed on the geochemistry of Hg in these soils (Roulet *et al.*, 1996) and on determining the source of waterborne Hg in the Tapajós River (Roulet *et al.*, 1998). It was proposed that the erosion of deforested oxisols causes an increase of Hg contamination of aquatic systems in the Tapajós Basin and in other disturbed catchments.

Because detailed data were lacking, no study has compared the various sources of Hg contamination to the Tapajós Basin. In order to assess the relative importance of the three possible sources of Hg mentioned above (goldmining, biomass burning, soil erosion), this study applies a mass balance approach to extensive Hg data on the various components of the Amazon forest. The relative contributions of goldmining activities and forest burning to atmospheric Hg (and its subsequent deposition) are estimated for the entire Amazonian basin. At the regional scale, the effects of goldmining, biomass burning and soil erosion on the Hg burdens in forest ecosystems of the Tapajós region are assessed, with a view to determining their relative contribution to the contamination of aquatic ecosystems.

2. Materials and Methods

Sampling was carried out in 17 sites during 5 sampling campaigns in 1992, 1994 and 1995. These sites were representative of three South American equatorial rain-forest zones (Figure 1): dense forest on clayey and hydromorphic oxisols in French Guyana (3 sites), dense forest on ultisols in the Tocantins Basin (1 site), and both dense and open forest on clayey and podzolized oxisols in the Tapajós Basin (13 sites). In order to assess regional scale variation of Hg accumulation in the forest ecosystem, with regard to the impact of goldmining activities, sampling stations in the Tapajós region were distributed over a transect along the shore of the river, extending from 50 to 350 kilometers downstream from the goldmining camps. Forest ecosystem compartments sampled were live tree and shrub leaves, live tree branches and twigs, roots and rootlets, leaf and woody litter, as well as coarse

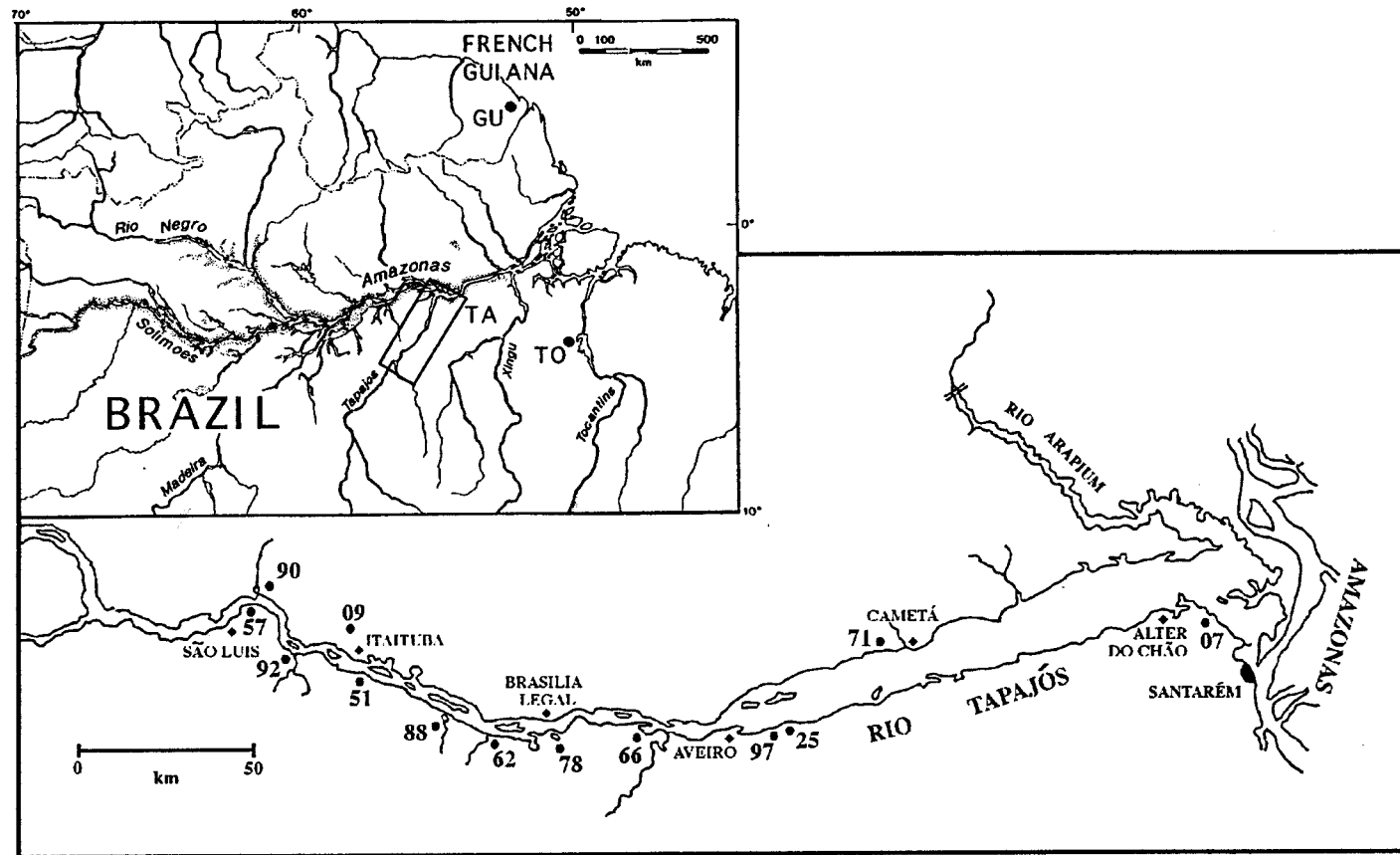


Figure 1. Study area and sampling sites. GU: French Guiana forest site near Cayenne; TO: Tocantim river site; TA: transect of forest sites along the Tapajós Valley (forest stations are illustrated in the detailed map).

humus. In each site, leaf and wood samples of 6 to 31 tree (and/or shrub) species were randomly collected within a 20-m radius. Soil cores were taken in each site using a 15-cm diameter PVC tube and sliced at 1-cm intervals (over a depth of 75 cm).

All samples were frozen within 2 hours after collection. In the laboratory, they were freeze-dried before being grounded and homogenized with a glass rod. For Hg analysis, samples weighing between 100 and 200 mg were digested in a 10:1 solution of nitric and hydrochloric acids for 6 hrs at 120°C. Hg concentrations were then determined by cold vapor atomic fluorescence spectrometry (Bloom and Fitzgerald, 1988). Details of the procedure may be found in Louchouart *et al.* (1993).

Mean total Amazonian forest total biomass (M) was recently estimated by Fearnside *et al.* (1993) in order to calculate the emission of greenhouse gases from forest fires. They calculated a mean value of 301 t/ha for above-ground living plant biomass, and of 385 t/ha for total biomass (including necromass) (see Table II). For mass balance calculations, total forest biomass values of Fearnside *et al.* (1993) were apportioned to each forest compartment using data on biomass composition in neotropical forests from Jordan (1989), Lescure *et al.* (1990) and Klinge *et al.* (1975). As for the soils, true density was determined by weighing a subsample of each 1-cm thick layer and following the method of Grondin *et al.* (1995).

In order to estimate the amount of mercury emitted to the atmosphere by a primary forest fire (Q_{Hg}), we used the equation developed by Seiler and Crutzen (1980) to estimate carbon emissions from a forest fire:

$$Q_{\text{Hg}} = A \cdot \text{RE} \cdot \sum(\text{CE}_i \cdot [\text{Hg}]_i \cdot M_i) = A \cdot \sum \text{EF}_i \quad (1)$$

(1) where A, is the surface area of forest fire, RE, the efficiency of Hg release to the atmosphere from the complete combustion of biomass, CE, the efficiency of biomass combustion, [Hg], the mercury concentration, M, the biomass, EF, the Hg emission factor for biomass combustion, and i, the forest biomass compartment considered. To calculate the cumulative Hg emission from biomass burning since the 1960's, the following equation was used:

$$Q_{\text{Hg}} = A \cdot \text{CE} \cdot \text{RE} \cdot \sum([\text{Hg}]_i \cdot M_i) = A \cdot \text{EF} \quad (2)$$

where CE is the mean cumulative combustion efficiency applied to the total biomass.

The efficiency of Hg release to the atmosphere (RE) from biomass combustion was assumed to be constant (among all biomass compartments) and to represent 90% of the initial mercury content (Veiga *et al.*, 1994).

The combustion efficiency (CE) of Amazonian primary forests is relatively low. Rainforest fires leave large quantities of standing and fallen incompletely burned plant biomass. The combustion efficiency of a first fire varies among the leaf, cauline and root components of plant biomass (Table II). The overall combustion

efficiency of a first fire averages only 28% of total above-ground biomass (Fearnside, 1991; Fearnside *et al.*, 1993). Leaf litter is completely burned, while no more than 30% of both woody litter and rootlet biomass is consumed (Seiler and Crutzen, 1980). However the mean cumulative combustion efficiency of three fires over the normal 10-year slash and burn cycle reaches 40% of the initial above-ground biomass (Fearnside, 1991).

3. Results

3.1. BURNING OF THE TROPICAL FOREST AS A SOURCE OF ATMOSPHERIC Hg

The mean Hg concentrations in the various compartments of a typical Amazonian forest were calculated as the averages of data from the three forests sampled in Brazil and French Guiana (Table I).

The Hg emission factor for tropical rain forest combustion is 273 g/km² for initial burning of primary forest using Equation (1), and 370 g/km² if one considers the cumulative impact of slash and burn agriculture (cycles of 3 fires over 10 years) with Equation (2) (Table II).

Estimates of the total surface area of forest burned (A) in Brazilian Amazonia since the onset of human colonization range from 270 000 km² (Fearnside, 1991) to 400 000 km² (Myers, 1991). Cumulative Hg emissions calculated from the data shown in Table II using Equation (2) amounted to 149–197 tons over the period 1960–1994. Estimates of the mean annual deforestation rate in the 1980s range from 22 000 km²/yr (Fearnside, 1991) to 34 000 km²/yr (Myers, 1991). Applying Equation (1) to the data shown in Table II, the annual atmospheric Hg emission from the initial burning of primary forest is estimated to be 6 to 9 t Hg/yr.

3.2. GEOGRAPHICAL DISTRIBUTION OF Hg CONCENTRATIONS IN FOREST VEGETATION, LITTER AND SOILS

Since the translocation of Hg from roots to the aerial parts of trees is negligible (Beaufort, 1977; Gobdol-Hutterman, 1988; Maserti and Ferrara, 1991), leaf Hg originates largely from atmospheric sources. Lindberg and Harris (1985) have observed that the interception of Hg in precipitation appears to be very efficient in the tropical forest canopy. Hg data in leaves and coarse humus may thus be used to assess the influence of Hg dispersion from a point source at the local and regional scale (Tamura *et al.*, 1985; Fukuzaki *et al.*, 1986). The spatial distribution of Hg concentrations in leaves and coarse humus (Figure 2) sampled in the Tapajós valley showed no contamination gradient with increasing distance from the goldmining point sources. The Hg concentrations observed in this region of intense goldmining activities are of the same order of magnitude as those found in the Tukurui (Tocantins) region, and in north-eastern French Guyana where goldmining activities

TABLE I
Mercury concentrations in Amazonian forest vegetation (ng/g d.w.)

Stations	Fresh leaves			Fresh stems, twigs twigs and branches			Roots			Rootlets (root mat)			Leaf litter			Wood litter			Coarse humus		
	mean	range	n	mean	range	n	mean	range	n	mean	range	n	mean	range	n	mean	range	n	mean	range	n
GU 1	152 ±41	53-226	23	15 ±4	9-19	7	33 ±27	10-72	4	129 ±17	117-141	2	140 ±35	97-197	6	34 ±19	15-53	3	204 ±34	153-248	6
GU 2	151 ±68	51-291	22	23 ±2	21-25	4	48		1	106 ±31	74-136	3	131 ±20	110-159	5	21 ±9	15-34	4	165 ±28	145-185	2
GU 3	123 ±47	51-220	20	22 ±8	16-28	2	36 ±6	28-41	4	107 ±53	61-173	4	85 ±23	60-121	5	28 ±8	19-34	3	102 ±27	83-121	2
TO 1	93 ±23	63-128	8	–			–			128 ±16	113-142	4	114 ±40	67-162	8	–			152 ±21	121-165	4
TA 07	56 ±34	18-134	11	13 ±9	8-27	4	23 ±8	17-19	2	31 ±4	26-35	4	56 ±4	51-61	4	26 ±26	7-42	5	63 ±8	53-78	7
TA 09	67 ±24	39-111	9	15 ±8	5-26	9	10		1	56		1	74 ±4	68-79	6	25 ±11	26-37	8	72 ±9	60-83	6
TA 25	101 ±59	22-162	6	19 ±8	12-32	7	12 ±4	9-14	2	84 ±50	35-154	4	100 ±11	84-109	4	30 ±5	14-32	4	113 ±14	110-136	5
TA 51	40 ±21	9-77	16	16 ±4	11-22	6	–			–			93 ±28	73-112	2	30 ±14	20-40	2	89 ±13	76-106	4
TA 57	64 ±35	17-163	31	–			–			–			–			–			83 ±5	78-87	3
TA 62	84 ±42	25-157	11	–			–			36		1	83 ±4	80-86	2	23		1	101 ±6	94-105	3
TA 66	57 ±27	3-114	23	–			–			–			–			–			–		
TA 71	111 ±58	49-198	7	–			–			70		1	80 ±9	74-90	3	25		1	92 ±10	75-106	6
TA 78	74 ±35	17-123	10	–			–			–			–			–			104		
TA 88	73 ±35	30-143	13	–			–			–			69 ±5	64-77	6	19 ±4	16-22	2	74 ±9	63-84	4
TA 90	94 ±48	17-66	11	24 ±14	8-48	6	–			65		1	101 ±18	75-127	8	48 ±14	28-59	4	119 ±28	89-153	5
TA 92	71 ±36	20-144	17	28 ±14	13-40	3	–			–			78		1	–			95		1
TA 97	63 ±40	16-127	13	–			–			67	40-89	3	85 ±26	58-109	3	42 ±19	26-66	4	113 ±33	90-136	2
Mean	99			27			26			88			106			30			–		

GU: French Guiana, TO: Tocantins, TA: Tapajos

TABLE II
Mercury emission factors from Amazonian forest fires

	% of biomass or litter ^a	Biomass or Necromass (10 ³ t/km ² d.w.) Mi	Hg concentration (ng/g d.w.) [Hg] _i	Hg burden (g/km ²) Mi.[Hg] _i	Combustion efficiency ^b (%) CE _i	Hg release efficiency (%) RE _i	Hg emission factor (g/km ²) EF _i =Mi.[Hg] _i .CE _i .RE _i
Fresh vegetation							
trunks	51	19.6	17	334	21	90	63
branches	17	6.5	17	111	49	90	49
leaves	2	0.8	99	76	100	90	69
total above-ground	70	27		521			181
root mat	4	1.5	88	136	30	90	37
roots	15	5.8	26	150	0	90	0
total below-ground	20	7.3		286			37
Dead vegetation							
litter							
leaves	1	0.4	106	41	100	90	37
wood	1	0.4	30	12	30	90	3
total litter	2	0.8		52			40
dead wood of litter layer							
stems	4	1.5	17	26	30	90	7
branches	2	0.8	17	13	30	90	4
standing dead wood	4	1.5	17	26	20	90	5
total necromass	10	4.6		118			55
Total vegetation (dead + fresh)							
		38.5		925			Initial Emission Factor ΣEF _i = 273
		38.5		925	40	90	Cumulative Emission Factor EF= 370

^a from the biomass composition of neotropical rain forest determined by Jordan (1989); Lescure *et al.* (1990); Klinge *et al.* (1975).

^b combustion efficiencies from Uhl *et al.* (1988); Ward *et al.* (1992); Fearnside *et al.* (1983).

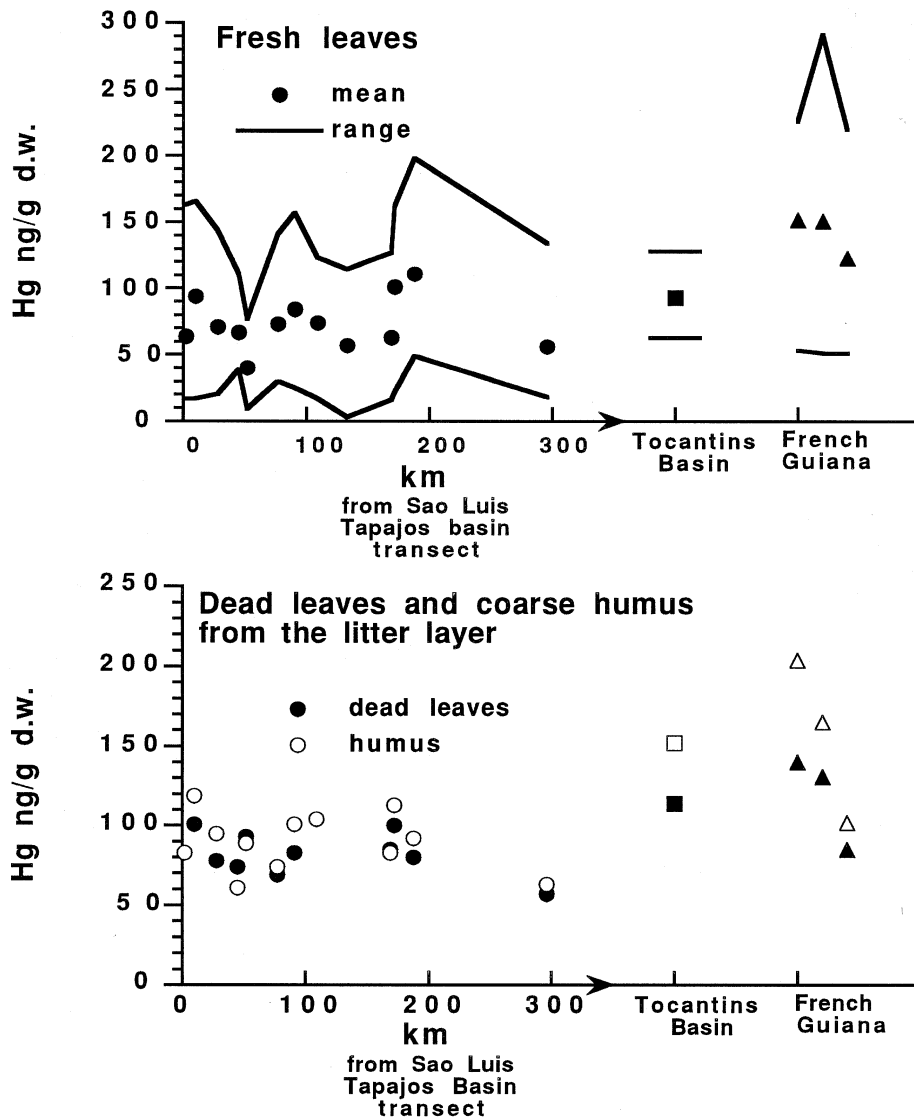


Figure 2. Geographical distribution of mercury concentrations in fresh leaves, litter and rough humus. The x-axis represent the distance in km from the beginning of the transect (São Luis, about 50 km downstream the first goldmining camp) along the Tapajós Valley. Hg concentration datas from others Amazonian sites are illustrated on the same axis for comparison.

are much less developed. Hg concentrations along the Tapajós valley transect suggest that atmospheric deposition intercepted by leaves and accumulated in humus is uniform at the regional scale.

3.3. INFLUENCE OF GEOCHEMISTRY AND PEDOLOGY ON THE Hg ACCUMULATION IN SOILS

Most of the Hg in the oxisols of Brazilian Amazon and French Guyana accumulate in the mineral horizons, associated with iron and aluminium oxy-hydroxides (Roulet and Lucotte, 1995; Roulet *et al.*, 1996). In the Tapajós Basin, topographic location determines soil type as well as the oxy-hydroxide and Hg content of the soil (Figure 3). There is a transition from clayey oxisols on the plateau to surface-leached oxisols and podzolized oxisols on the valley slope, typical of soil system overlying the Alter-do-Chão sedimentary formation (Lucas *et al.*, 1984). According to the geochemical model of Lucas *et al.* (1996), sandification and podzolization processes control the evolution of soils on slopes. These processes leads to the depletion of iron and aluminum oxy-hydroxides (Fe_{cdb} and Al_{cdb}) as well as of the associated Hg (Roulet *et al.*, 1996) in soils on the slopes of the Tapajós Basin.

The dense mineral horizons of Amazonian soils show high concentrations of Hg (Table III). Hg burdens in the upper 20 cm vary between 10 000 and 30 000 $\mu\text{g}/\text{m}^2$ in the Tapajós region, between 60 000 and 70 000 $\mu\text{g}/\text{m}^2$ in the very clayey and oxy-hydroxide-rich soils in French Guiana and 25 000 $\mu\text{g}/\text{m}^2$ in the Tocantins Basin ultisols. These burdens are ten times greater than those observed in the upper layer of temperate and boreal soils (Grondin *et al.*, 1995; Grigal *et al.*, 1995; Lee *et al.*, 1995).

4. Discussion

4.1. Hg EMISSION TO THE ATMOSPHERE IN AMAZONIA

Taking into account the impact of cumulative fires in slash and burn agriculture, the cumulative emissions to the atmosphere by biomass burning of Hg amounted to 149 to 197 tons (4 to 6 tHg/yr) over the period 1960–1994. The current annual atmospheric Hg emission for the first burning of primary forest is estimated at only 6 to 9 t Hg/yr.

Since the late 1970s, 1500–3000 tons of Hg are estimated to have been released into the environment by goldmining activity in Amazonia (Pfeiffer *et al.*, 1993). Atmospheric emissions account for an estimated 65 to 83% of this total, amounting to 1000 to 2500 tons over 15 years. This represents a mean annual emission rate from goldmining sources of 65 to 170 tons Hg/yr, in general agreement with the mean total annual atmospheric input of 78 t Hg from goldmining estimated by Lacerda and Marins (1997). Goldmining Hg represents 67% of the total use of

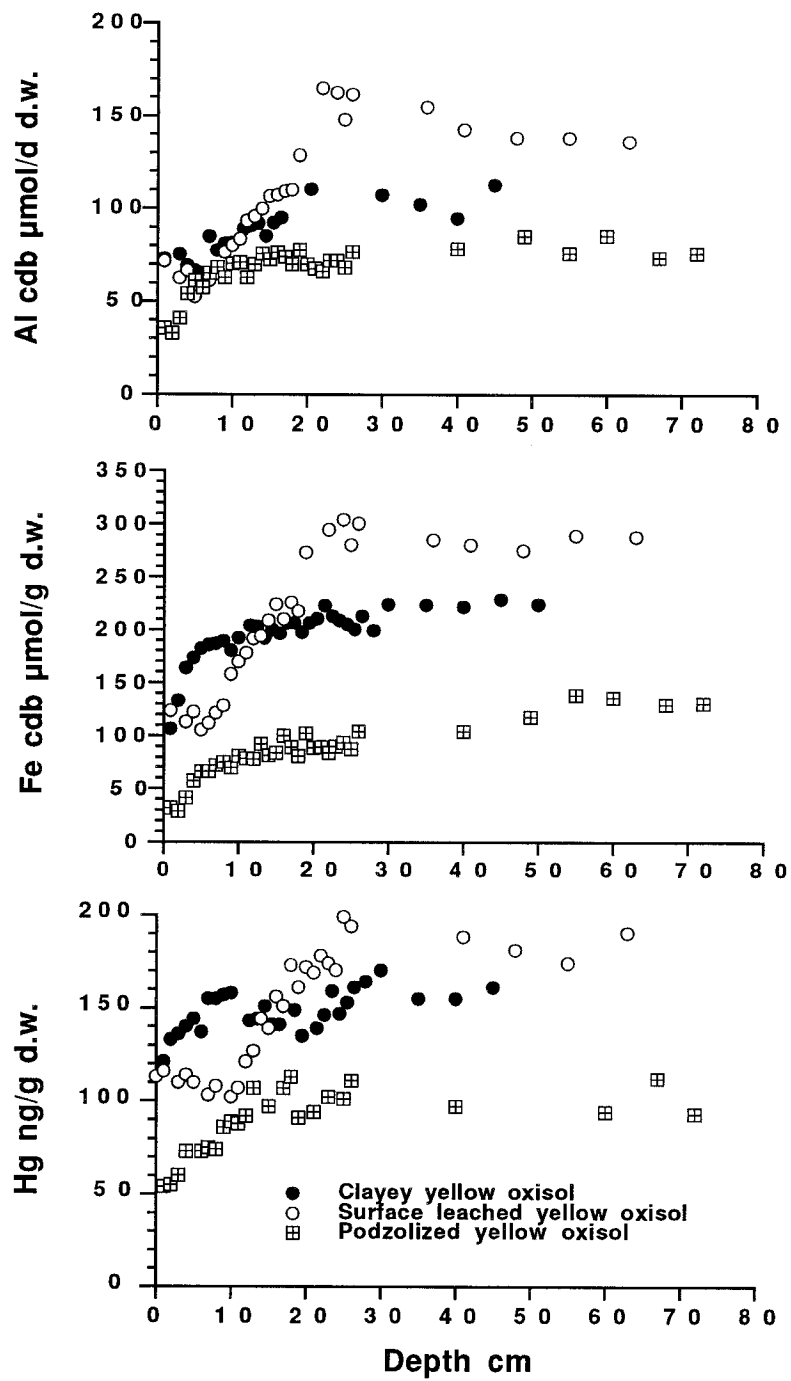


Figure 3. Profiles of Al_{cdb}, Fe_{cdb} and Hg concentrations in typical soils from the Tapajós Valley slopes.

TABLE III
Mercury concentrations and burdens in Amazonian soils

Stations	Soil type	Hg concentration ng/g d.w.						Hg burden 10 ³ μg/m ²			
		Organic horizon			0–20 cm		20–60 cm		Organic horizon	0–20 cm	0–60 cm
		mean ± s.d.	range	thickness cm	mean ± s.d.	range	mean ± s.d.	range	cm	cm	cm
GU 1	clayve yellow oxisol	199	–	1	270 ± 35	199–318	–	–	0.9	69	–
GU 2	clayve yellow oxisol	219	–	1	240 ± 13	219–264	–	–	1.1	63	–
GU 3	hydromorphic oxisol	60 ± 6	54–65	4	67 ± 9	54–80	–	–	1.5	13	–
TO 1	ultisol	71			94		130		1.7	24	76
TA 51	clayve yellow oxisol	137 ± 28	83–169	11	137 ± 27	83–169	156 ± 13	131–166	4.5	18.9	113.7
TA 62	clayve yellow oxisol	132 ± 10	124–143	3	134 ± 7	124–144	136 ± 14	117–158	0.5	32.6	126.5
TA 71	clayve yellow oxisol	93	–	1	96 ± 12	82–118	109 ± 5	104–118	0.3	22.5	84.1
TA 78	clayve yellow oxisol	125 ± 20	104–144	4	130 ± 12	104–150	144 ± 7	132–155	0.9	21.4	102.7
TA 92	clayve yellow oxisol	127 ± 8	121–133	2	143 ± 11	121–162	153 ± 10	136–170	0.1	25.1	103.6
TA 25	surface leached yellow oxisol	116	–	1	132 ± 25	102–173	183 ± 15	165–212	0.2	33.8	180.8
TA 97	surface leached yellow oxisol	180 ± 26	143–207	6	132 ± 35	86–207	157 ± 31	118–187	1.0	18.4	110.1
TA 57	podzolized yellow oxisol (ultisol)	–			83 ± 18	54–113	101 ± 6	94–111	–	25.6	97.3
TA 88	podzolized yellow oxisol (ultisol)	–			62 ± 12	46–82	93 ± 11	77–113	–	12.9	58.9
TA 66	podzolized yellow oxisol (ultisol)	105	–	1	43 ± 11	22–60	70 ± 14	54–88	0.1	9.9	59.7
TA 07	podzolized yellow oxisol (ultisol)	84 ± 8	75–101	8	74 ± 15	45–101	69 ± 14	54–88	2.5	10.3	41.5
TA 09	podzolized yellow oxisol (ultisol)	84 ± 8	78–90	2	48 ± 14	34–80	77 ± 28	41–133	0.4	9.9	68.6

GU: French Guiana (adapted from Roulet & Lucotte, 1995), TO: Tocantins (adapted from Aula *et al.*, 1994), TA: Tapajós.

Hg in Brazil (Lacerda and Marins, 1997), and 86 to 96% of the total atmospheric emission from anthropogenic sources (goldmining and forest burning combined) in Brazilian Amazonia.

4.2. DEPOSITION OF ANTHROPOGENIC AIRBORNE Hg ON FOREST SOILS

Spatial distribution of Hg in topsoil and vegetation near goldmining centers and gold shops show that Hg concentrations decrease rapidly with distance from the source (Lacerda *et al.*, 1991; Malm *et al.*, 1991, 1995; Pfeiffer *et al.*, 1993). Bulk deposition and sedimentation of Hg are higher near Brazilian gold mines than in remote location (Table IV). Great caution is needed with the sediment interpretations for the calculation of atmospheric deposition; they do not distinguish the relative contribution of goldmining, forest burning, and natural sources to atmospheric Hg. Nor do they distinguish the specific geochemistry of the sediment and the influence of the climate on its deposition and release from terrestrial surface. Von Tümpling *et al.* (1996) observed significant bulk precipitation deposition from fire emission only during the dry season in a remote area of Pantanal where setting fire to grasslands and periodic wetlands is a common and important agricultural practice. Mason *et al.* (1994) have estimated a bulk deposition of $12.7 \mu\text{g}/\text{m}^2/\text{yr}$ for pristine continental areas between 10°N and 10°S , of the same order as the recently measured $21.5 \mu\text{g}/\text{m}^2/\text{yr}$ for remote rain forest in Amapá State (Fostier *et al.*, 1997). This suggests that most of the Hg emitted to the atmosphere is deposited near its source and the rest is dispersed by diffuse long-range transport (more than 50 km). None of these estimates take into account the effect of volatilization and the influence of the transport of natural aerosols (Artaxo *et al.*, 1996; Hacon *et al.*, 1995) on the Hg burdens in Amazonian ecosystems.

Our estimate for the entire Amazon region is based on the above observations and concerns only the deposition of Hg originating from goldmining activities. Speciation of Hg emitted to the atmosphere is a determining factor for global budget balance (Masson *et al.*, 1994). There is no study of the speciation of Hg in the Amazonian atmosphere. All the ionic Hg is normally deposited locally (Masson *et al.*, 1994). For the present estimate we arbitrarily decided that 50% of the Hg emitted to the atmosphere is deposited near its sources and that the other 50% undergoes long-range atmospheric transport and is uniformly dispersed over the entire Amazon basin. In order to set an upper limit for goldmining Hg atmospheric long distance deposition on terrestrial ecosystems, we may assume that 33–85 tons of Hg from goldmining activities are deposited uniformly on the entire surface of the Brazilian Amazon (states of Acre, Amapá, Amazonas, Maranhão, Mata Grosso, Pará, Rondônia, Roraima and Tocantins). It can be noted that this calculation seems conservative since Artaxo *et al.* (1996) have observed an exportation of Hg through the atmosphere from the Amazon Basin to the South Atlantic and the tropical Pacific Oceans. If one considers Amazonia as a closed system with a surface area of $5 \cdot 10^6 \text{ km}^2$, the maximum deposition rate of Hg derived from goldmining activities

TABLE IV
Bulk mercury deposition rates in Amazonian environment

Methods	Locations	Deposition rates $\mu\text{g}/\text{m}^2/\text{yr.}$	Notes	References
<i>Bulk atmospheric deposition</i>				
	Pantanal, southern Brazilian Amazon, 1–10 km from gold mines	151	direct measurement	von Tümpling <i>et al.</i> (1996)
	Pantanal, southern Brazilian Amazon, remote 110 km from gold mines	25	direct measurement during the rainy season and beginning of dry season (attributed to gold mining)	von Tümpling <i>et al.</i> (1996)
	Pantanal, southern Brazilian Amazon, remote 110 km from gold mines	40	direct measurement during the dry season (attributed to grassland and wetland biomass combustion)	von Tümpling <i>et al.</i> (1996)
	Amapá, northeastern Brazilian Amazon, remote undisturbed rain forest, 100 km from gold mines	21	direct measurement, remote from gold mining	Fostier <i>et al.</i> (1997)
	Pristine continental areas between 10°N and 10°S	13	estimation from oceanic measurement	Mason <i>et al.</i> (1994)
<i>Bulk sediment accumulation</i>				
	Pantanal, southern Brazilian Amazon, near gold mines	90–120	no chronological or geochemical data	Lacerda and Salomons (1991)
	Mato Grosso, Alta Floresta, southern Brazilian Amazon, near gold mines	20/210	respectively background-remote/near emission point sources, no geochemical data	Gomes <i>et al.</i> (1996)
<i>Calculation</i>				
	Brazilian Amazon, gold mining Hg contribution to remote rain forest (more than 50–100 km from gold mines)	7–17	see text	this study

is estimated to be 7–17 $\mu\text{g}/\text{m}^2/\text{yr}$, amounting to 100–250 $\mu\text{g}/\text{m}^2$ over the period of 1979–1994. This is consistent with the value measured by Fostier *et al.* (1997) in a remote rain forest of Amapá State (Table IV).

4.3. RELATIVE CONTRIBUTIONS OF ANTHROPOGENIC AND NATURAL SOURCES TO SOIL Hg BURDENS

In an undisturbed Amazonian forest catchment, Fostier *et al.* (1997) observed a positive input/output budget of Hg suggesting that it is being recycled within the forest and/or retained by the soil system. Rapid turnover of organic matter at the surface of tropical soils limits the amount of Hg accumulated in the organic horizon, which is much lower than in the underlying mineral horizons (Table III). Fluxes of atmospheric Hg into the soil may be controlled by humification and mineralization rates, as well as by the depth of organic matter penetration (Roulet and Lucotte, 1995). Working on Amazonian forest oxisols, Trumbore (1993) showed that over a 10–15 year period, most surface-derived organic matter is found in the upper 20 centimeters. Supposing they were completely mixed into the upper 20 centimeters of our oxisols, the 100–250 $\mu\text{g}/\text{m}^2$ cumulative deposition of atmospheric Hg transported over long distance from goldmining sources (over the past 15 years) would account for less than 3% of the soil surface layer Hg burden in the Tapajós Basin. The contribution from forest fire emissions would be lower by about an order of magnitude.

4.4. RELEASE OF Hg BY SOIL EROSION

Deforestation and cultivation of oxisols in the Tapajós region also causes depletion of soil Hg (Roulet *et al.*, 1996). The removal of plant cover and the destruction of root systems lead to the loss of soil structure, allowing heavy rains to scour the soil. The thin organic horizon as well as the upper centimeters of the mineral soil may thus be carried away (Roulet *et al.*, 1996). Increased oblique runoff through cultivated soils on slopes may result in the depletion of iron oxy-hydroxides and Hg in the mineral horizons as fine clay particles are selectively eroded (Roulet *et al.*, 1996). This leads to increased sandification of these soils, which may thus be likened to an acceleration of the natural process of soil transformation resulting from oblique runoff. Soil impacts due to the loss of protective forest cover may be particularly severe on steep slopes: gully erosion is observed in many sites, with the loss of the upper 30–50 cm of soils. These soil disturbances may export considerable amounts of particulate material and associated Hg to the aquatic environment.

Our soil data show that the loss of the organic horizon (thickness ranged from 1 to 11 cm) is expected to mobilize 200–4600 $\mu\text{g Hg}/\text{m}^2$ (Table III). The selective erosion of the 30–50% fine clay content (based on oxy-hydroxides profiles, Roulet *et al.*, 1996) would mobilize an average 500 $\mu\text{g Hg}/\text{m}^2/\text{cm}$ of eroded soil, whereas

gullying and scouring would transfer 500–3000 $\mu\text{g Hg/m}^2/\text{cm}$ of eroded soil to the aquatic environment.

5. Conclusions

Although Hg release from goldmining activity is commonly held responsible for the Hg contamination of aquatic ecosystems remote from mining centers, a cause-effect relationship between these two phenomena has yet to be shown.

At the scale of the entire Amazon basin, the contribution of goldmining to atmospheric Hg dominates largely over that of forest fires, with respective emissions of 65–170 and 6–9 t Hg/year. The latter estimate supports the conclusion of Lacerda (1995) that the contribution of fires to anthropogenic Hg emissions to the atmosphere is relatively small.

At the regional scale of the Tapajós basin, Hg burdens observed in forest ecosystems indicate that the cumulative deposition onto the soils of atmospheric Hg transported long distances from goldmining sources is negligible relative to the amount of Hg accumulated in the soils. With an estimated total flux of 100–250 $\mu\text{g Hg/m}^2$ over 15 years, this deposition accounts for less than 3% of the Hg burden in the upper 20 cm soil layer. For the sake of comparison, a slash and burn cultivation cycle directly emits 370 $\mu\text{g Hg per m}^2$ of forest burned. After a fire, unburned forest biomass decomposes slowly, releasing an additional 550 $\mu\text{g Hg/m}^2$ at the surface of soils.

Thus, while goldmining activity results in severe direct Hg contamination of aquatic ecosystems and humans at the local scale, near goldmining centers, long-distance atmospheric transport of Hg cannot account for the Hg burdens found in the various compartments of Amazonian forest ecosystems.

Our data show that an important effect of human colonization may be the erosion of deforested soils releasing an estimate 500 to 3000 $\mu\text{g Hg/m}^2$ per cm of soil eroded. This source presumably causes a significant Hg contamination of Amazonian rivers. Eroded soils may lose up to 30 000 $\mu\text{g Hg per m}^2$, most of which presumably reaches the aquatic environment in particulate form (Roulet *et al.*, 1998). In newly colonized Amazonian environments soil erosion may be the major process increasing the loading of natural Hg to adjacent aquatic ecosystems.

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