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Is the Negro River Basin (Amazon) impacted by naturally occurring mercury?

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Abstract

In order to investigate the major sources and cycling of mercury in the Negro River Basin (Amazon), total metal measurements were carried out in soils, in river and lake waters, in the atmosphere, and in bulk precipitation during the period 1995 throughout 1998. Median values of 1.3 ng m^{-3} in the atmosphere, 172 μg kg^{-1} in soils, 4.6–7.5 ng l^{-1} in three different lakes, 4.5 ng l^{-1} in 17 different Negro River tributaries and 20.3 μg m^{-2} $year^{-1}$ in bulk precipitation were found. Mercury concentrations in rivers and lakes waters, as well as in soils and bulk precipitation were high, considering the scarcity of anthropogenic point sources in the region. Mercury leaching from soil, the largest regional reservoir of this metal, was considered to be the major pathway to mercury enrichment in the region.

Keywords: Amazon; Mercury; Cycling; Soil; Water

1. Introduction

Mercury contamination in the Brazilian Amazon region has been mostly associated to gold mining activities. This informal type of mining is carried out in the so-called *garimpos*, where metallic mercury is used to amalgamate gold. The Hg–Au amalgam is then burned to purify the gold, causing loss of mercury into the atmosphere. It has been estimated that the amount of mercury released in the Amazon since early 1970s, when the modern gold rush began, has already reached 2000 tons (Malm, 1998). However, the link between mercury contamination and gold mining is not always clear in the Amazon, and has

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been under constant debate in the scientific community. Silva-Forsberg et al. (1999), e.g. found high levels of mercury in fish and in the hair of the riparian population in the upper Negro River Basin, a region where gold mining activities are rare and under constant surveillance of the Federal Police.

Mercury contamination in remote areas has already been reported in the literature. In most cases, it has been attributed to natural sources (Rasmussen, 1994), or to long-term atmospheric transport of anthropogenic emissions (Mason et al., 1994; Hanisch, 1998; Mercury Study Report to Congress, 1998). In the Negro River Basin, identification of individual mercury sources has been difficult due to the lack of reliable data on mercury concentrations in the key environmental matrices.

To elucidate the sources and the fate of mercury and subsidize further ecotoxicological studies in the Negro River Basin, a monitoring program was established in 1995 in this region, centered on sampling water, sediment, air, rainwater, hair from riverine populations, fish and soils. The major objective was to generate the first reliable database of mercury concentrations in these key reservoirs in the northern part of the Brazilian Amazon, a remote region with little documented history of gold mining activities.

2. Methods

2.1. Study area

The Negro River Basin spreads over an area of 690×10^3 km^2, representing approximately 14% of the total area of the Brazilian Amazon. The basin is characterized by a landscape with predominate elevation between 0 and 100 m above sea level (IBGE, 1998) and an average rainfall of 2000 mm annually, with well-defined seasonal behavior. The Negro River covers an extension of approximately 1700 km, with an average flow of 29 000 m^3 s^{-1} in Manaus, where after meeting the Solimões River, they form the Amazon River (Fig. 1). The figure also shows the sampling sites used during these 3 years: soil sampling sites are marked as (♦), lakes as (⌘), rivers as (▲), and atmosphere as (☆).

2.2. Cleaning of glassware

Teflon® bottles and other sampling glassware were rinsed with MilliQ® water, soaked in hot (70°C) 4 mol l^{-1} HCl for 48 h, washed with MilliQ® water, and filled with sub-boiling distilled HCl containing 5 ml l^{-1} of a BrCl solution, prepared as described by Bloom and Crecelius (1983). After 24 h, bottles were rinsed several times with MilliQ® water, dried in a class 100 fume hood, filled with 100 ml of high purity 0.02 mol l^{-1} HCl and stored in three consecutive plastic bags until sampling. This is basically the same procedure recommended by Vermette et al. (1995).

2.3. Water sampling in lakes and rivers

Surface samples were collected along the Negro River and in various tributaries, between Santa Isabel do Negro River (S 00°25′ W 64°30′) and Manaus (S 03°03′ W 60°06′). In lakes, samples were collected at different depths. Samples were collected and handled using ultra-clean techniques including shoulder-length polyethylene gloves, hot acid pre-treatment of flasks, and triple plastic wrapping, taking into account precautions similar to the ones used for measurement of mercury in open sea water (Gill and Fitzgerald, 1985, 1987; Mason and Fitzgerald, 1991). The acid solution was discharged far from the collection point and care was taken to avoid sampling water over which the boat had passed, to prevent contamination of the water sample. The bottles were rinsed at least five times prior to filling, then filled and triple-bagged. Some bottles were returned to the laboratory for blank check of the HCl filling solution.

The determination of reactive mercury was carried out according to the following procedure: 2 ml of a $SnCl_2$ solution (containing 20% (w/v) of

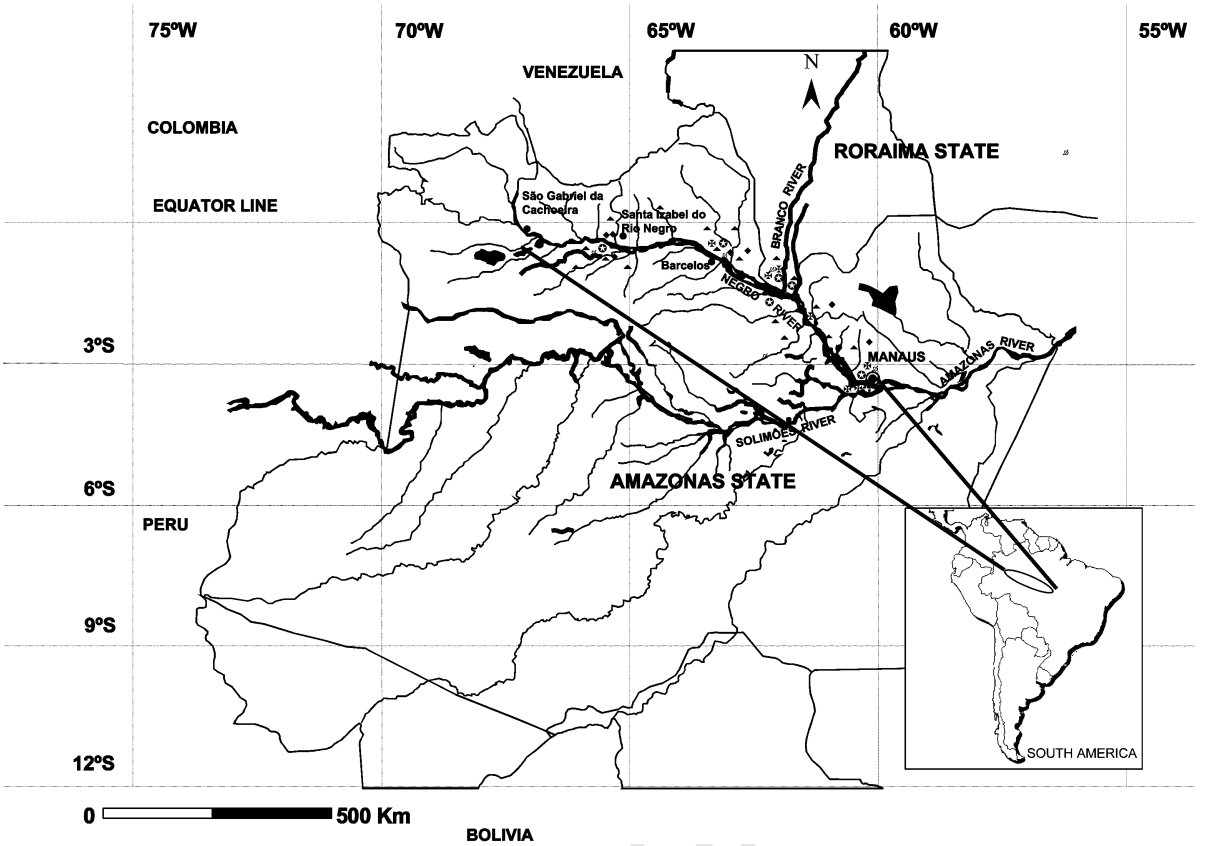

Fig. 1. Sampling sites: soils (♦), lakes (⊕), rivers (▲) and atmosphere (☆).

$SnCl_2$ and 10% (v/v) concentrated HCl, and bubbled with gold filtered argon for 45 min) were added to 100 ml of a non-filtered sample. The sample was then purged with argon (300 ml min^{-1}) for 15 min. The mercury released was amalgamated in a gold sand column, de-sorbed by heating, and detected using a Brooks Rand® CVAFS equipment (Bloom and Fitzgerald, 1988). A detection limit of 0.04 ng l^{-1} (blank plus three times the standard deviation) has been observed. For total mercury, 100 ml or less of sample were first wet-oxidized with BrCl (Bloom and Crecelius, 1983).

2.4. Mercury in bulk deposition

Bulk deposition was collected at 1 m height using a 23-cm-diameter glass funnel connected to a 1-l bottle using glass tubing. This collection device, except for the glass funnel, was housed inside a 200-mm-internal diameter PVC tube. All samples were preserved with 3 ml of BrCl solution added prior to sample collection. No overflow problems were observed and all events were collected in an entire form.

2.5. Mercury in soils

Soil samples from seven different sites (representing six different types of soil) were collected at various depths using steel hand augers and preserved in plastic bags without air at −18°C. For mercury determination, samples were digested as follows: 1–2 g of wet soil sample were suspended in 10 ml of MilliQ® water, followed by additions of 5 ml of concentrated sulfuric acid, 2.5

ml of concentrated nitric acid and 15 ml of 5% (w/v) potassium permanganate solution. After 15 min, 8 ml of 5% (w/v) potassium persulfate solution was added and the sample was heated for 2 h at 95°C. After cooling, excess of permanganate was destroyed by adding 6 ml of 12% (w/v) hydroxylamine hydrochloride solution, and the final volume was adjusted to 50 ml. Standard solutions and a blank used in the calibration procedure were prepared following this same procedure. Elemental mercury was determined after reduction with $SnCl_2$ by CVAAS using the flow injection method (Pasquini et al., 1988; Canela and Jardim, 1997). Results represent the mean of ≥ 2 replicate analyses, expressed as micrograms total Hg per kilogram of dry soil. Determination of total Fe and Al were conduced using X-ray fluorescence (Spectrace 5000) using bulk solid samples ground to fine homogeneous powder.

2.6. Mercury leaching from soil

Five different surface soil samples, and five soil samples collected below 40 cm were suspended in MilliQ® water to simulate a possible phase transfer between soil and water due to weathering processes. Aliquots of soil (0.4 g dry base) were suspended in 100 ml of MilliQ® water in Erlenmeyer flasks with Teflon® stoppers and placed in an orbital shaker at 120 strokes min^{-1} for 2 h. After 7 days of quiescent equilibrium, 30 ml aliquots were removed from each flask and centrifuged. Total Hg concentration in the supernatant solution was determined in the sub-samples by CVAFS after a BrCl oxidation. After withdrawing the samples, the total volume of the soil suspension was again adjusted to 100 ml and the whole procedure was repeated for another 7 days.

2.7. Mercury in the atmosphere

A known volume of air, typically 0.1–0.3 m^3 was pumped through a gold sand quartz filled column, using a battery-operated air pump, continuously monitored by a flowmeter. Air samples were collected 1 m above the soil or water surface at flow rates ranging from 500 to 800 ml min^{-1}. Reference curves for CVAFS determination were constructed by injecting different volumes of saturated mercury vapor in the system (Dumarey et al., 1985).

3. Results and discussion

The results presented in this paper were obtained in 8 excursions during the period of September 1995 throughout November 1998. The Negro River is well known for its unique physicochemical characteristics of the water, showing a very high content of dissolved organic matter (up to 20.7 mg C l^{-1}). Water collected in this basin shows an average pH of 4.86, with values as low as 3.78 having been measured just after the wet season. The so-called 'black waters' also show very little dissolved solids and average conductivity values approximately 15 $\mu S\ cm^{-1}$, as previously described by Williams (1968) and Kuchler et al. (1994). The following results obtained in these campaigns were used to elucidate not only the stock of mercury in different reservoirs in the river basin but also the inter-reservoir fluxes.

3.1. Mercury in lake and river waters

The monitoring of mercury in lake water was centered in seven lakes, five of them characterized as black waters (Cristalino (S 03°08′ W 60°07′), Tupé (S 03°03′W 60°15′), Maependi (S 02°10′W 61°03′), Iara (S 01°20′W 62°03′), Nazaré (S 00°58′W 62°55′), and two from white waters, Araça (S 01°14′, W 61°50′) and Calado lake (S 03°15′, W 60°34′). The values obtained for total and reactive mercury are presented in Table 1. Taking into consideration average values of mercury concentration in non-polluted surface waters worldwide, they can be considered at least three or four times higher than the ones expected for such remote areas (Amyot et al., 1997; Vandal et al., 1998). Mercury concentration is also considered high when compared to the ones obtained for both Lake Michigan (Sullivan and Mason, 1998) and Adirondack lakes (Driscoll et al., 1994). In a recent survey conducted in preserved areas

in São Paulo State to evaluate mercury background concentrations, it was found that for Óleo lake (S 21°35′ W 47°50′) the mean concentration of total mercury (ng l^{-1}) was 1.3 ± 0.6 with a median of 1.2 ng l^{-1} ($n = 6$).

In order to evaluate the total mercury concentration in the aquatic system of the Negro River Basin, as well as the content of dissolved organic carbon, 16 major tributaries were sampled, from Santa Isabel do Negro River down to Manaus, covering an extension of nearly 600 km. The results are presented in Table 2.

Despite the fact that this basin is characterized by waters rich in humic material, a poor correlation ($R^2 < 0.2$) between both total and reactive Hg and total organic carbon (TOC) concentrations was found. In this context, it is important to emphasize that for these samples, values of TOC are very close to the ones obtained for DOC. This result differs from the ones reported for the Adirondack lakes (Driscoll et al., 1994, 1995), in waters that resemble the Negro River in terms of DOC and total Hg. It has been postulated that the aqueous mercury cycle is coupled to naturally occurring organic matter (Meili, 1991). However, it is important to bear in mind that DOC measures the stock of organic carbon present in the water body, which may not necessarily be related to the availability of mercury binding sites, specially in these water bodies, where the ratio DOC/metals is very high.

When compared with other rivers worldwide, the concentrations of mercury found in the Negro River and its tributaries are in the same range as those reported for Lake Michigan tributaries (Hurley et al., 1998) and other rivers in areas with high population density as well as industrial activities (Mason and Sullivan, 1998). Taking into consideration the anthropogenic activities in these two regions, it is possible to conclude that riverine concentrations found in this work are high. Considering the median Hg concentration in the Negro River as 4.5 ng l^{-1}, and assuming an average flow of 29 000 m^3 s^{-1}, the total amount of mercury exported by the river at Manaus (just before the Amazon River) reaches 4.1 tons per year.

Table 1
Total and reactive mercury in six different lakes in the Negro River Basin and in the Solimões River basin[a]

Sampling site	*N*	Mean	Amplitude	Median
$Hg_{reactive}$				
Cristalino lake	20	1.3 ± 0.8	0.1–2.9	1.4
Tupé lake	22	1.5 ± 0.9	0.3–4.2	1.3
Maependi lake	14	1.7 ± 0.4	1.3–2.3	1.5
Iara lake	11	1.9 ± 0.8	0.8–3.6	1.6
Nazaré lake	4	1.9 ± 0.7	1.7–2.3	2.0
Calado lake	16	1.9 ± 0.1	0.6–6.7	1.1
Araçá lake	7	1.0 ± 0.6	0.3–1.5	1.2
Hg_{total}				
Cristalino lake	20	4.9 ± 2.2	0.8–7.4	5.9
Tupé lake	24	6.8 ± 3.5	1.8–17.1	7.0
Maependi lake	14	8.0 ± 2.2	5.6–13.3	7.0
Iara lake	11	6.8 ± 1.2	4.9–9.0	6.4
Nazaré lake	4	7.7 ± 0.6	7.1–8.6	7.6
Calado lake	14	5.2 ± 0.3	1.2–12.3	4.6
Araçá lake	7	5.3 ± 1.0	3.81–6.57	5.2

[a]All values are in ng l^{-1}.

Table 2
Total concentration of mercury and dissolved organic carbon in Negro River tributaries

Tributary	TOC (mg C l^{-1})	Hg_{total} (ng l^{-1})
Demene (below River Aracá)	14.0	7.4 ± 0.6
Demene (above River Aracá)	12.5	11.4 ± 0.6
Aracá	13.2	3.8 ± 0.6
Negro (near Barcelos city)	9.1	2.3 ± 0.6
Urubaxi	10.9	15.0 ± 0.3
Maruia	8.1	7.3 ± 0.5
Tea	18.0	8.0 ± 0.2
Uneixi	15.4	5.7 ± 0.3
Daraá	20.7	11.5 ± 0.3
Aiuanã	11.8	13.1 ± 0.3
Juapiris	–	4.5 ± 0.5
Unini	–	3.5 ± 0.2
Jaú	11.6	3.3 ± 0.3
Cananaú	13.1	3.3 ± 0.4
Cuieiras	9.0	2.9 ± 0.3
Branco	5.1	2.0 ± 0.4
Maependi	15.4	4.1 ± 0.4
Median value	12.5	4.5

Table 3
Atmospheric total depositional fluxes of mercury

Place	Deposition ($\mu g\ m^{-2}\ year^{-1}$)	Reference
Chesapeake Biological Laboratory, MD, USA	13	Mason et al., 1997
Hart-Miller Island, MD, USA	25	Mason et al., 1997
Lake Michigan, USA	< 10	Hoyer et al., 1995
Lake Champlain, USA	< 10	Burke et al., 1995
Vermont, USA	9.26	Burke et al., 1995
Wisconsin, USA	10.3	Burke et al., 1995
Florida, USA	15–28	Landing et al., 1995; Guentzel et al., 1995
Suburban Seattle, USA	15	Bloom, 1996
Pacific Coast of Washington State, USA	7	Bloom, 1996
North Central Wisconsin, USA	12.5–23	Morrinson et al., 1995
Quebec, Canada	7.6	Poissant and Pilote, 1998
Mean value of North America	8.86 ± 2.06	Poissant and Pilote, 1998
Wisconsin, USA	11.8	Vandal et al., 1991
Amazon region, far from gold mining regions, median value	20.3	This work

3.2. Mercury in the atmosphere

It is worth mentioning that atmospheric sampling was carried out at height of 1 m above nude soil or forest area, as well as over flooded forest. These different sites have to be considered because the precipitation regime in the Amazon is responsible for flooding up to 20% of the total area of the basin just after the wet season, dropping to only 4% in the dry season. At this moment it is not possible to identify the individual contribution (and scavenging) due to volatilization from naked or covered soils or water bodies, or from canopy deposition. As pointed out by Lindberg et al. (1998), mercury emissions from soils are very dependent not only on temperature, turbulence, humidity and vertical mixing, but also on sampling height from the soil, specially in a forest area. Based upon the mercury figures presented in Tables 3 and 4, and assuming 12 km of troposphere, the Negro River Basin shows an atmospheric mercury pool of nearly 10.8 tons. Assuming a very simple and conservative model of emission and deposition, and taking into consideration the mercury burden obtained for the atmosphere, an uniform layer of 12 km in the troposphere, and the washout of mercury due to the total precipitation of 2068 mm $year^{-1}$ (Ferreira, 1991), the average atmospheric residence time for mercury in the Negro River Basin is 0.8 year. This residence time is in accordance with the range (0.5 up to 2 years) presented by Lindqvist (1994), and the value of 1 year presented in the EPMAP (1994). Mason et al. (1994) calculated the current concentration of mercury in the troposphere as 1.6 ng m^{-3}, and pointed out that the value in the pre-industrial atmosphere was 0.5 ng m^{-3}. In this work the median concentration of gaseous mercury was found to be 1.3 ng m^{-3}, which is in accordance with Mason's estimate, and approximately three times higher than the value from the last century. This fact indicates the influence of the global cycle of mercury over the Amazon.

The determination of total mercury carried out in 6 different samples of bulk deposition collected over the basin during both wet and dry seasons showed a median value of 9.8 ng l^{-1}. For this reservoir, the relative small number of samples allows only a preliminary analysis. Taking into account a mean annual rainfall of 2068 mm in the basin, one can estimate a total mercury deposi-

tion of 20.3 μg m^{-2} $year^{-1}$. This value is well above the majority of the values presented in Table 3 for other localities and in the same range as industrialized and temperate areas. It is important to remember that the average temperature in the Negro River Basin is 26.6°C (Ferreira, 1991), which favors mercury emission from both soils and water bodies, when compared with temperate areas in the world. This deposition represents a total load of 14 tons per year over the Negro River Basin.

3.3. Mercury in soils

Table 5 summarizes the major results obtained for the six different types of representative soils from the basin. Several points can be addressed concerning the parameters investigated. Firstly, the vertical distribution of mercury in the Negro River Basin indicates that these soils are naturally rich in mercury. This statement is based upon the data presented in Table 6, that shows a comprehensive survey of mercury concentration in soils collected worldwide. Recently, Roulet et al. (1998a) also found high levels of mercury in Amazonian soils from the Tapajós River Valley.

A significant correlation between the total concentrations of mercury, aluminum and iron in these soils was found, as presented in Figs. 2 and 3. Higher concentrations of mercury in deep layers of soil can be associated to a possible mechanism of surface leaching of Hg, followed by retention onto both iron and aluminum oxy-hydroxides in mineral horizons, as already presented by Roulet and Lucotte (1995) and Roulet et al. (1998a).

The carbon content in soil profiles shows higher concentrations in the first centimeters (up to 11%), decreasing to values of approximately 0.5% at 1 m depth, what is considered a normal pattern in poor soils such as the ones found in the Amazon. A poor correlation between organic content and total mercury was found. Lack of evidence associating mercury to organic carbon content in tropical rain forest soil has also been observed in the Tucuruí region (Aula et al., 1994), in French Guiana (Roulet and Lucotte, 1995) and Tapajós region (Roulet et al., 1998a).

Experiments on mercury leaching conducted under laboratory conditions using surface sample soils suspended in water showed an average leaching capacity of 1.5 μg of mercury per kg of soil in 14 days of contact. The experimental conditions used in this experiment can be considered

Table 4
Atmospheric total gaseous mercury concentrations

Place	Concentration (ng m^{-3})	Reference
Atlantic Ocean (southern hemisphere)	0.7–2.5	Slemr and Langer, 1992
Atlantic Ocean (northern hemisphere)	1.0–4.8	Slemr and Langer, 1992
Global mean	1–4	WEC, 1989
Northern hemisphere	1.5–2.0	EPMAP, 1994
Urban areas, worldwide	4.0	Nriagu, 1990
Rural areas, worldwide	1.6	Nriagu, 1990
Remote areas, worldwide	0.38	Nriagu, 1990
Scandinavia	2–4	Petersen et al., 1995
Global background values	1–4	Petersen et al., 1996
North Pacific	1.77	Fitzgerald et al., 1991
Wisconsin, USA	1.57	Fitzgerald et al., 1991
Tennessee, USA	2.15	EPRI, 1994
Scandinavia	2.5–2.8	Iverfeldt, 1991
New York, USA	2.1–2.7	Olmez and Ames, 1997
Amazon region, far from gold mining region	1.3	This work

Table 5
Total mercury and other elements in soils from the Negro River Basin

Type of soil	Depth (cm)	Horizon	C (%)	N (%)	Fe_2O_3 (%)	Al_2O_3 (%)	Hg total ($\mu g\ kg^{-1}$)
Allic Plinthudult	0–8	A11	2.45	0.21	4.7	10.5	91 ± 4
Allic Plinthudult	8–25	A12	0.79	0.12	5.3	13.1	91 ± 8
Allic Plinthudult	25–55	B21	0.68	0.15	7.8	23.2	225 ± 8
Allic Plinthudult	55–95	B22	0.43	0.15	12.1	30.3	241 ± 79
Allic Aquic Paleudult	0–20	C1	11.64	0.98	2.0	32.0	231 ± 22
Allic Aquic Paleudult	20–40	C2	1.65	0.17	1.9	33.2	200 ± 3
Allic Aquic Paleudult	40–80	C3	0.43	0.05	2.5	33.0	193 ± 2
Allic Superic Plinthaquox	0–10	A11	2.06	0.22	2.3	25.4	204 ± 19
Allic Superic Plinthaquox	10–45	A/B	0.87	0.16	2.7	27.7	165 ± 46
Allic Superic Plinthaquox	45–80	B21pl	0.50	0.15	4.7	36.3	320 ± 20
Allic Superic Plinthaquox	80–95 +	B22pl	0.30	0.12	10.2	33.1	198 ± 7
Ultic Haplorthox	0–25	A11	2.90	0.24	5.1	17.3	132 ± 56
Ultic Haplorthox	25–45	A/B	0.46	0.05	7.0	32.3	147 ± 45
Ultic Haplorthox	45–85	B21pl	0.45	0.08	8.5	38.2	304 ± 14
Ultic Haplorthox	85–160	B22pl	0.39	0.13	7.6	37.5	234 ± 7
Petroferric Pauleudult	0–5	A11	2.04	0.29	2.7	24.9	132 ± 5
Petroferric Pauleudult	5–30	A/B	1.38	0.17	3.8	31.1	134 ± 1
Petroferric Pauleudult	30–75	B21cn	0.92	0.19	6.7	38.2	163 ± 10
Petroferric Pauleudult	75–95 +	B21cn	0.89	0.15	9.6	33.6	300 ± 74
Petroferric Pauleudult	0–5	A11	8.70	0.55	1.8	16.7	119 ± 8
Petroferric Pauleudult	5–55	B21	0.67	0.14	2.6	37.6	113 ± 2
Petroferric Pauleudult	55–100 +	B22	0.44	0.12	4.0	29.0	106 ± 3
Aeric Arenic Tropaquod	0–60	A11	1.66	0.19	21.6	18.2	156 ± 63
Aeric Arenic Tropaquod	60–80	C1	0.69	0.15	21	23.7	178 ± 63
Aeric Arenic Tropaquod	80–130	C2	0.30	0.16	21.2	23.4	210 ± 50
Aeric Arenic Tropaquod	130–300 +	C3	0.26	0.15	26.9	25.6	81 ± 14

mild when compared with field conditions, especially considering that the extractor used (Ultrapure water) shows little Hg leaching capacity when compared with the ubiquitous acid black waters in the basin. This result shows that soil in the Negro River Basin has a large stock of mercury that can be slowly released by the action of geochemical transformation processes.

Comparing the mercury burden in the soil throughout the Negro River Basin to that estimated from the anthropogenic sources yields an interesting picture, similar to the ones observed by Roulet et al. (1999) and Fostier et al. (2000) for other Amazonian regions. The Negro River Basin has an area of $\sim 690 \times 10^3$ km^2, with a median concentration of mercury in the top 1 m layer of 164 $\mu g\ kg^{-1}$. This represents a pool of mercury of nearly 126×10^3 tons in this top layer of surface soil. The mercury input over the entire Brazilian Amazon during the present gold rush (last 30 years) has been estimated as 2000–3000 tons (Malm, 1998). This figure represents, in the worst scenario, only 2.4% of this natural pool of mercury. The hypothesis that mercury contamination over the entire Amazon is mainly due to mining activities is further weakened by the fact that the Negro River Basin represents only 11% of the total area of the Brazilian Amazon, and assuming an even distribution of this total mining mercury input over the entire area, the basin would receive 330 tons only, which corresponds to 0.26% of the mercury found.

However, the possibility of this mercury being accumulated due to total atmospheric deposition has to be ruled out. Considering a total atmospheric deposition of mercury of approximately 20.3 $\mu g\ m^{-2}\ year^{-1}$ at the present time, the Negro River Basin would receive a yearly an

Table 6
Total mercury concentrations in different types of soils around the world

Place	Hg_{total} ($\mu g\ kg^{-1}$)	Reference
Global average	50–100	WEC, 1989
Polluted area around a clinical thermometer plant in China	200	Wang, 1994
Mean in China	38	Wu et al., 1991
Limestone region in China,	190	Wu et al., 1991
Regions with intensive use of sewage in agriculture in China	180	Wu et al., 1991
Paddy soils in China,	120	Wu et al., 1991
Median in the Guianese Forest	182	Roulet and Lucotte, 1995
Background values in territories without anomalies	45 ± 3	Anoshin et al., 1996
Average value of 1267 soils in USA	58	Anoshin et al., 1996
Mineral horizon of Tapajós River Basin, Brazil	50–210	Roulet et al., 1998a
Agriculture soils in Europe	~100	Bringmark, 1997
Agriculture soils in USA	~110	Bringmark, 1997
South-eastern and northern Europe and Central USA	<60	Bringmark, 1997
Poland	150	Bringmark, 1997
Forest soils in continental Europe	50–150	Bringmark, 1997
Forest podzolin in Sweden, mor layer	250	Lindqvist et al., 1991
Forest podzolin in Sweden, elluvial layer	15	Lindqvist et al., 1991
Forest podzolin in Sweden, illuvial layer	30	Lindqvist et al., 1991
Forest podsol in Sweden, O, E, B and C horizons	7	Lindqvist et al., 1991
Negro River Basin, Amazon, Brazil	172	This work

input of 14 tons. Assuming the hypothetical total immobilization and trapping of this amount of metal in the top 1 m of soil, it would take nearly 90 centuries to build up to the mercury stock

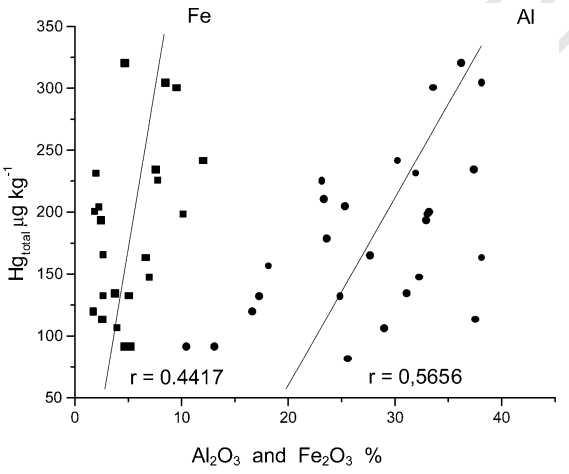

Fig. 2. Correlation of total mercury with aluminum and iron in soils collected over the Negro River Basin.

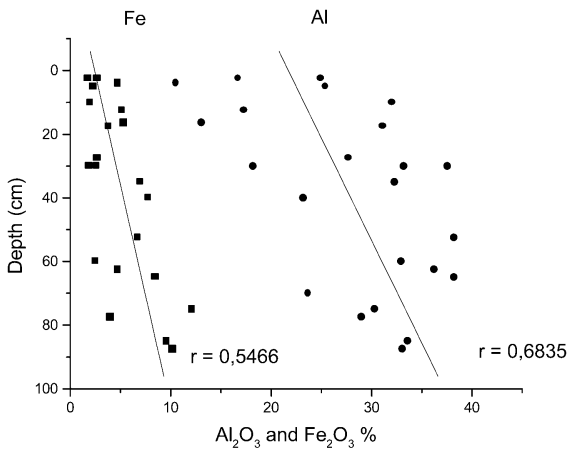

Fig. 3. Correlation of aluminum and iron with depth in soils collected over the Negro River Basin.

found experimentally. When the 4.1 tons of mercury washout are taken into consideration, then it would take 127 centuries to reach this figure.

However, as pointed out by Nriagu (1994), mining activities in the Spanish America between 1580 and 1900, may have contributed ~196 000 tonnes of mercury spread in South and Central America. Even assuming (very unlikely) that all this mercury was equally deposited over the Brazilian Amazon, this would account only for 20 000 tonnes, which represents just 10% of the total concentration found in the top 1 m layer.

Bulk deposition observed in this work can be considered high (Table 3), not due to the high Hg concentration in the rain water, but mainly as a result of large inputs of precipitation (~2000 mm $year^{-1}$). This fact, associated to a relatively low level of gaseous mercury in the atmosphere (Table 4), shows the intense dynamic of Hg cycling in the region, involving re-emission and atmospheric deposition steps.

4. Summary and conclusions

The importance of this work is to produce the first comprehensive inventory of mercury in different environmental reservoirs over the northern Amazon. The results indicate high levels of mercury in river and lake waters, with no seasonal variations. Soils presented high contents of Hg, in agreement with results obtained to others region in the Brazilian Amazon (Roulet et al., 1998a,b, 1999; Fostier et al., 2000). However, it is important to point out that Roulet et al. (1998a,b, 1999) worked with soils from the Tapajós basin, a region with historical gold mining activities. In accordance with the findings of Roulet et al. (1998a), the Hg concentrations in these soils are not related to the carbon concentration. Total Fe and Al contents show good correlation with the Hg burden. For the Negro River Basin, it is feasible to assume the soil as the major source of mercury to all other reservoirs through pedogeochemical transformation processes (Roulet et al., 1998a). Mercury bioaccumulation due to the enrichment of water bodies from the continuous metal release from soils has already being translated to public health problems due to the consumption of fish with very high levels of mercury (Lindqvist et al., 1991; Barbosa et al., 1997, 1998; Barbosa and Dorea, 1998; Silva-Forsberg et al., 1999).

Taking into consideration the levels of mercury detected in the reservoirs investigated in this work, any further input of mercury in this region (e.g. garimpos) has to be critically considered due to the already high natural concentrations. Also, further actions towards mitigating public health aspects related to mercury contamination in the riparian population need to be taken on a short-term basis.

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