

## SOILS AS AN IMPORTANT SINK FOR MERCURY IN THE AMAZON

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**Abstract.** This work focuses on the behaviour of mercury in lateritic soil profiles found in the Serra do Navio and Tartarugalzinho areas of the State of Amapá in Northern Brazil. The Hg contents are high in the upper horizons of the soil profiles ( $100\text{--}300\ \mu\text{g kg}^{-1}$ ), and decrease to less than  $100\ \mu\text{g kg}^{-1}$  at depths of 200 or 300 cm. The higher levels of Hg are associated with higher Fe concentrations, particularly in the ferruginous accumulations as mottles and nodules. For each horizon of the soil profile, balance calculations were used to distinguish the amount of mercury naturally accumulated from rocks through lateritic pedogenesis (lithogenic mercury) from the anthropogenic mercury introduced in the profile through atmospheric contamination. The results show that the anthropogenic contribution is significant in the upper horizons (up to 95% of the total Hg), and decreases downward in the soil profile. Mercury burdens were calculated for soil profiles in both upslope ( $272\ 880$  and  $217\ 440\ \mu\text{g m}^{-2}$  for the first 70 cm) and downslope positions ( $118\ 800$  and  $182\ 160\ \mu\text{g m}^{-2}$  for the first 70 cm). The loss of Hg in downslope profiles seems to be related to the natural evolution of iron duricrust into latossols, which has been brought about by climatic changes toward increasing humidity in the Amazon since the Tertiary.

**Keywords:** Amazon soils, latossols, mercury

### 1. Introduction

Mercury contamination from gold mining is considered one of the worst environmental problems affecting the Amazon ecosystems (Lacerda and Salomons, 1998). Amongst the evidences of such contamination are the high levels of mercury found in fish and fauna, and in the hair, urine and blood of Native Americans, prospectors and riverside populations in close proximity to the mining sites (Barbosa *et al.*, 1995; Bidone *et al.*, 1997a; Bidone *et al.*, 1997b; Boischio and Henshel, 1996; Hacon *et al.*, 1997a; Guimarães *et al.*, 1998). However, these ecotoxicological signals are also detected in remote areas not directly affected by mining activities (Lacerda *et al.*, 1991; Forsberg *et al.*, 1994), and this suggests that there are other sources of mercury contamination. Many studies have been carried out in the Amazon in order to establish how much mercury results from mining and other anthropogenic activities, and how much arises naturally.

According to Pfeiffer and Lacerda (1988), mercury pollution from gold mining



in the Amazon amounted approximately to 128 tons per year in the early 80's. Of this, 45% was released into rivers and 55% was released into the atmosphere. In 1997 the annual input of mercury into the atmosphere as a result of gold mining in Brazil was estimated as 78 tons (Lacerda and Marins, 1997). This was over 67% of total mercury emissions in Brazil that year and nearly all of it was released in the Amazon region.

Veiga *et al.* (1994) have argued that deforestation through forest fires is a major source of mercury contamination of aquatic systems. The burning of trees, they say, releases 90 tons of mercury directly into the atmosphere every year. This figure was considered an overestimation by Lacerda (1995) who recalculated the contribution of biomass burning to approximately 17 tons per year from 1978 to 1988, and only 8.7 tons per year from 1990 to 1991. This author thus concluded that gold mining, rather than deforestation, is the principle contributor to mercury pollution in the Amazon.

More recently, Roulet and his colleagues (Roulet and Lucotte, 1995; Roulet *et al.*, 1996; Roulet *et al.*, 1997) have claimed that neither gold mining nor the burning of forest biomass could account for the high mercury concentrations throughout the Amazon's ecosystems. Gold mining, they say, could have only contaminated aquatic ecosystems on a small scale, close to the mining sites, and the contribution of forest fires was limited to 6 to 9 tons of mercury per year. According to these authors, a large proportion of mercury found in the Amazon's ecosystems originates from the degradation of ferrallitic soils. Mercury has accumulated in association with iron oxyhydroxides, largely as a result of natural pedogenetic processes. The pedological evolution of these soils toward podzolization (Lucas *et al.*, 1996), whether occurring naturally or accelerated by deforestation, has resulted in the dissolution of iron oxyhydroxides and the release of associated mercury. Once in solution, mercury would have rapidly escaped into the aquatic ecosystems.

The importance of soils as sinks for mercury had been previously pointed out by many authors, such as Nriagu (1990) and Mason *et al.* (1994). The main contribution of Roulet and his colleagues was to show that ferrallitic soils are particularly efficient traps for mercury, and that the degradation of those soils presents a threat to the environment. However, their calculation that anthropogenic sources account for only 3% of the overall content of mercury in Amazonian soils would seem to be an underestimation. They have taken into account only the 1500 to 3000 tons of mercury released into the Amazon environment during the last gold rush (from 1979 to 1994). From this total, 35 to 85 tons per year would have been emitted to the atmosphere and ultimately incorporated to soils. Their estimates did not take into account the legacy of mercury pollution from the silver mines of the Spanish colonial era (from 1570 to 1900). According to Nriagu (1993), approximately 196 000 tons of Hg were released into the environment during this period, what is an average emission into the atmosphere of 594 tons per year. This atmospheric mercury could have been transported over thousands of kilometers (Hacon *et al.*, 1995), contaminating the Amazon basin. This figure has never been included

in the models of the mercury global cycle (Lacerda, 1997), but has undoubtedly contributed to the mercury currently found in terrestrial reservoirs.

Other sources of atmospheric mercury, whether natural (wind-borne soil particles, volcanoes, etc) or anthropogenic (industrial activities) may be responsible for the pollution of ecosystems (Nriagu, 1989; Nriagu 1990). However, in the Amazon these atmospheric sources seem to be considerably less important than regional contamination from the burning of amalgamated gold.

In conclusion, both anthropogenic and natural processes – though they operate in different time scales – are responsible for mercury contamination of the Amazon's ecosystems. Through gold mining – an entirely anthropogenic source – mercury enters the ecosystems either via water or the atmosphere. Atmospheric mercury moves from one region to another, is deposited, and subsequently incorporated into soils. In South America this has been going on for the past five hundred years, particularly intensively in the Amazon during the last gold rush of the 80's.

Forest fires are another significant means by which mercury is mobilized. Fires can occur naturally but, in recent years, it is often the human colonization of the Amazon which has been responsible for them. Mercury mobilized in this way derives from two sources. Some of it has accumulated over hundreds of years in vegetal tissues, as a result of biomass metabolism; some has become concentrated on the surface of leaves following atmospheric deposition.

The mercury content of soils derives from two sources: one is lithogenic and consists of mercury naturally accumulated by ferrallitic pedogenesis; the other is atmospheric, largely of anthropogenic origin, resulting from the deposition of mercury used for amalgamation. As a result, lateritic soils are both repositories of lithogenic mercury and filters that retain atmospheric mercury. Human interference, resulting in intensification of erosion and degradation of these soils, adds considerably to the contamination of aquatic systems.

This article aims to contribute to the discussion outlined above. It will contrast the behaviour of mercury in soil profiles in areas directly affected by gold extraction (Tartarugalzinho, in Amapá State), with that of mercury in areas situated some distance from the sources of contamination (Serra do Navio, in Amapá State). In the Serra do Navio region, the effect of lateral evolution of the pedologic cover on the behaviour of mercury will be assessed through the study of profiles in a range of different topographic locations.

## 2. Materials and Methods

### 2.1. THE STUDY AREAS AND SOIL SAMPLING

The study areas are located in the Amapá State, to the north-east of the Brazilian Amazon, approximately at the latitude of 00°00' (Figure 1). The climate is warm and humid, with mean temperatures between 30 and 35 °C and annual mean rainfall exceeding 3000 mm.

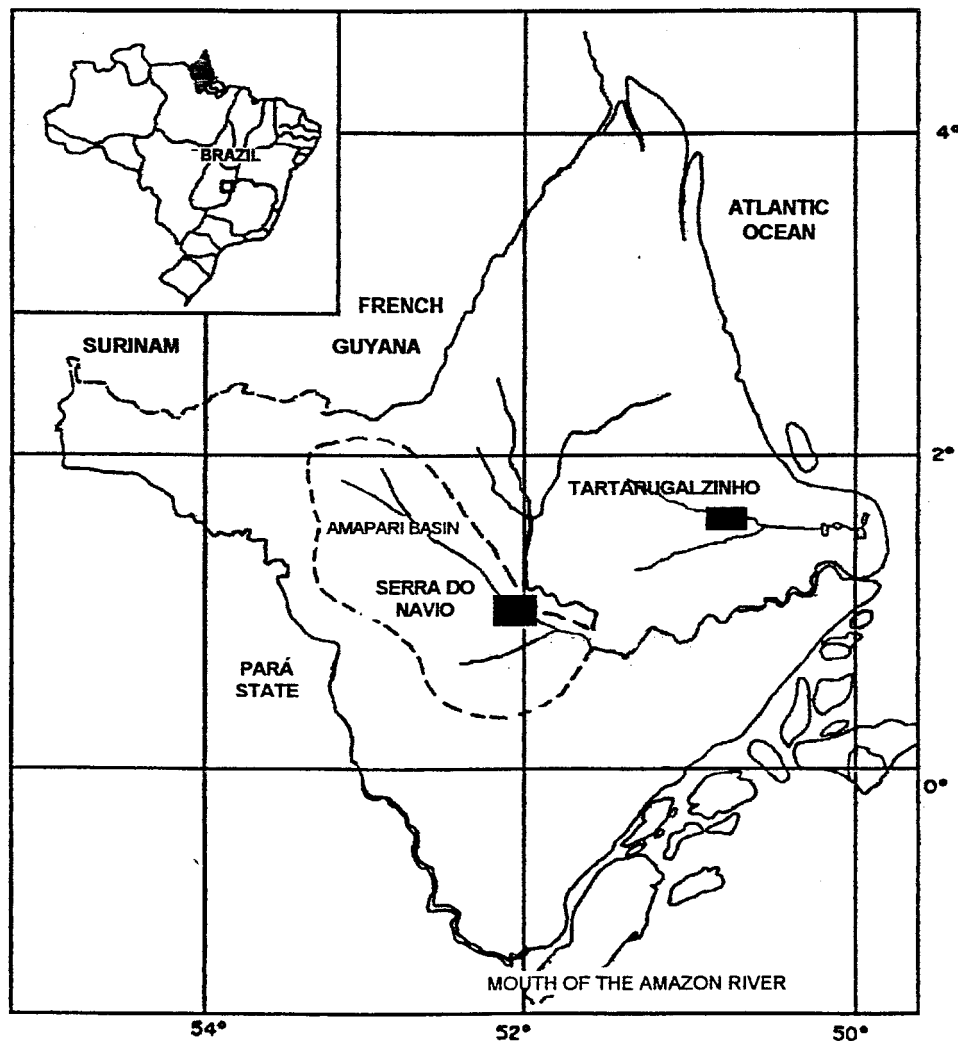


Figure 1. Amapá State. General location map of Serra do Navio and Tartarugalzinho areas.

The area chosen as control has no known goldmining sites nearby. It is a small catchment (Pedra Preta Creek) in the River Amapari valley, near to the village of Serra do Navio, in the central part of the State. On each margin of the catchment, two soil profiles on upslope and downslope positions were sampled (Figure 2). On the northern margin, profiles A and B are separated by about 300 m and are situated at altitudes of 100 m and at 30 m, respectively. Profiles C and D are on the southern margin, separated by 250 m, at altitudes of 10 and 30 m, respectively.

The sampled soils are typical *in situ* latosols with almost no accumulation of organic matter (%C < 5%) in the upper horizons. The upslope profiles (A and D) consist of a homogeneous argillaceous cover overlying a ferruginous nodular

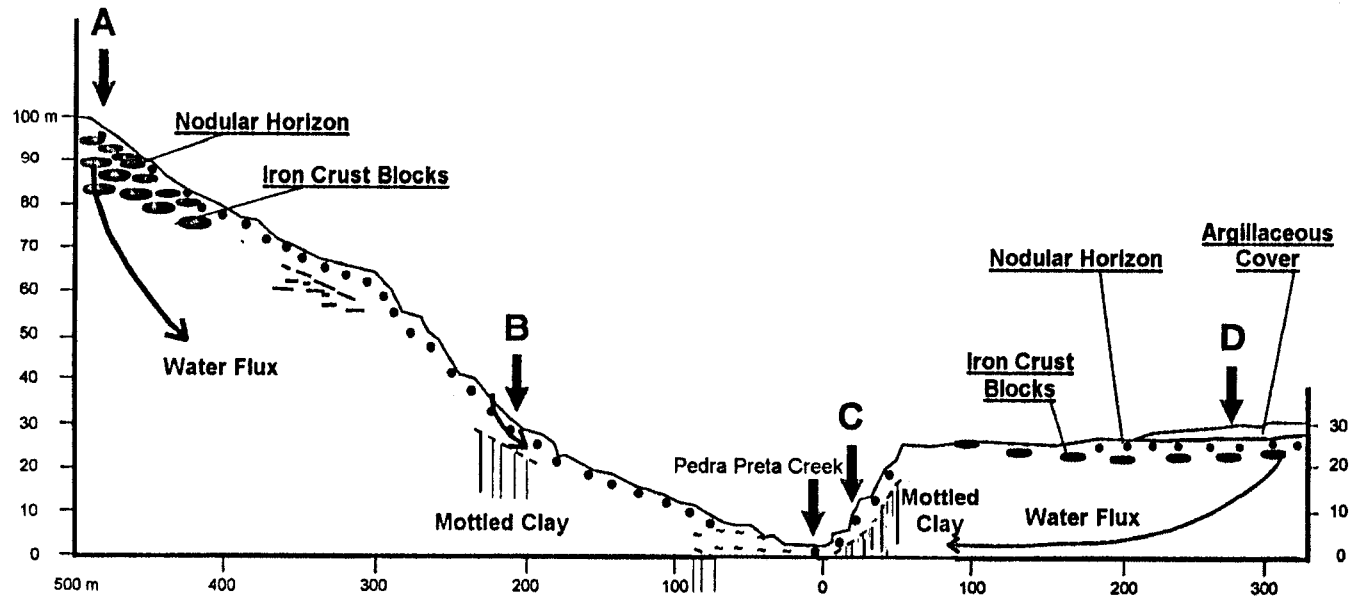


Figure 2. Location of soil profiles in the Serra do Navio area.

horizon that grades downward to an iron duricrust. The latter prevented sampling beyond a depth of 100 cm. The downslope profiles (B and C) could be sampled down to 450 and 370 cm, respectively. From top to bottom they consist of a homogeneous argillaceous cover, a ferruginous nodular horizon, a transition zone and a mottled clay horizon. In the nodular horizons, where possible, nodules and matrix were sampled separately.

The area directly affected by gold mining is situated near the village of Tararugalzinho. This is on the eastern border of the state of Amapá, about 200 km away from Serra do Navio. A soil profile near the mining operations was sampled down to 550 cm. From top to bottom it comprises a homogeneous argillaceous cover, a mottled clay horizon which includes a quartz stone-line, and a saprolite horizon derived from granitic rocks.

## 2.2. ANALYTICAL TECHNIQUES

Samples of about 1 kg were stored in plastic bags and immediately frozen. After drying at 40 °C, the >2 mm fraction was removed by sieving. Chemical and mineralogical analyses were then carried out on pulverized samples. Mercury concentrations were measured after acid digestion (Malm *et al.*, 1990) by cold-vapor atomic absorption spectrophotometry. Iron was determined by instrumental neutron activation analysis. Mineralogical composition was estimated by X-ray diffraction using an instrument with Cu-K $\alpha$  radiation at a speed of 0.020° 2 $\theta$  s<sup>-1</sup>. Undisturbed samples were measured for bulk density.

## 3. Results

### 3.1. ACCUMULATION OF MERCURY IN SOILS

Mineralogical and chemical data for mercury (Hg) and iron (Fe) are displayed in Tables I and II. The soils are mostly made up of quartz, kaolinite and goethite. Hematite and traces of mica are also sometimes present. The iron contents can be wholly attributed to oxyhydroxides (goethite and hematite), which are the dominant Fe-bearing minerals in these soils, by a long way.

Mercury behaviour is characteristic in soil profiles of the Serra do Navio (Table I). In the homogeneous argillaceous cover and in the nodular horizons, the mercury content is generally high, reaching more than 300  $\mu\text{g kg}^{-1}$ . Going deeper, a slight decrease of Hg is observed in the nodular horizons. In these horizons, hematite is more abundant, and both Hg and Fe are higher in nodules than in the surrounding matrix. In the mottled clay horizons of profiles B and C, mercury decreases to less than 100  $\mu\text{g kg}^{-1}$ , which corresponds to the background level for Amazonian soils (Pffeifer *et al.*, 1993; Lacerda, 1995).

In order to investigate whether Hg found in soil profiles was exclusively a result of pedogenesis or at least partially derived from external sources, geochemical

TABLE I  
Mineralogical composition and Hg and Fe contents in Serra do Navio soils (n = nodules; m = matrix)

Profiles	Description	Quartz	Goethite	Hematite	Kaolinite	Mica	Hg ( $\mu\text{g kg}^{-1}$ )	Fe ( $\text{g kg}^{-1}$ )
A								
0–20 cm	Argillaceous Cover	+++	+	–	++	–	197±1	82±3
60–70 cm (m)	Nodular Horizon						210±1	189±2
60–70 cm (n)	Nodular Horizon	++	++	+	+++	–	270±1	343±15
110–120 cm (m)	Nodular Horizon	(+)	++	+	++	+	224±1	233±3
110–120 cm (n)	Nodular Horizon	(+)	+++	+	++	(+)	316±1	320±4
B								
0–20 cm	Argillaceous Cover	+++	++	+	+	(+)	260±1	230±8
40–50 cm (m)	Nodular Horizon	+++	++	+	++	–	312±1	175±1
40–50 cm (n)	Nodular Horizon	(+)	+++	++	++	(+)	373±1	345±2
140–150 cm (m)	Nodular Horizon	+++	++	+	++	–	287±1	144±2
140–150 cm (n)	Nodular Horizon	(+)	+++	++	++	–	843±4	279±4
200–250 cm	Transition Zone	+++	++	+	+	(+)	306±9	180±5
250–300 cm	Transition Zone						325±1	155±2
300–350 cm	Mottled Clay Horizon	(+)	++	–	+++	–	79±1	115±2
350–400 cm	Mottled Clay Horizon	+++	++	(+)	+	(+)	78±1	207±3
400–450 cm	Mottled Clay Horizon	++	++	++	+++	–	81±1	250±8

(+) Trace.

+ Present.

++ Moderate.

+++ Abundant.

TABLE I  
Mineralogical composition and Hg and Fe contents in Serra do Navio soils (n = nodules; m = matrix)

Profiles	Description	Quartz	Goethite	Hematite	Kaolinite	Mica	Hg ( $\mu\text{g kg}^{-1}$ )	Fe ( $\text{g kg}^{-1}$ )
<b>C</b>								
0–20 cm	Argillaceous Cover	+++	+	–	+	(+)	203±1	71±1
50–60 cm (n)	Nodular Horizon	+++	++	+	++	(+)	203±1	364±1
80–90 cm (m)	Nodular Horizon	+++	++	–	–	(+)	139±1	111±.5
80–90 cm (n)	Nodular Horizon	+++	++	+	–	–	152±1	272±.5
120–130 cm	Transition Zone	+	+	–	–	(+)	139±1	89±.5
170–180 cm	Mottled Clay Horizon	+	+	–	–	–	94±4	39±1
240–250 cm	Mottled Clay Horizon	+	+	(+)	–	–	79±1	
360–370 cm	Mottled Clay Horizon	+	+	–	–	–	29±1	189±2
<b>D</b>								
0–5 cm	Argillaceous Cover	+++	++	–	++	–	373±1	80±1
30–35 cm	Nodular Horizon	+++	++	–	+++	–	311±1	81±.5
65–70 cm	Nodular Horizon	(+)	++	+	+++	(+)	634±2	142± 1

(+) Trace.

+ Present.

++ Moderate.

+++ Abundant.



TABLE II  
Mineralogical composition and Hg and Fe contents in Tartarugalzinho soils

	Description	Quartz	Goethite	Hematite	Kaolinite	Mica	Hg ( $\mu\text{g kg}^{-1}$ )	Fe ( $\text{g kg}^{-1}$ )
0–100 cm	Argillaceous Cover	+++	(+)	–	(+)	–	173±6	33±.5
100–200 cm	Mottled Clay Horizon	+++	(+)	+	(+)	(+)	212±9	124±3
200–300 cm	Mottled Clay Horizon	+++	(+)	–	+	(+)	134±1	50±.5
300–380 cm	Stone Line	+++	(+)	(+)	(+)	(+)	100±1	26±.3
380–400 cm	Mottled Clay Horizon	+++	+	+	+	++	100±1	136±1
400–450 cm	Saprolite	+++	–	–	+	+	68±5	10±.1
450–550 cm	Saprolite	++	–	–	+	+++	39±1	19±2

(+) Trace.

+ Present.

++ Moderate.

+++ Abundant.

balance calculations (Krauskopf and Bird, 1995) were performed for profiles B and C. For each profile, the lowermost and least weathered sampled horizon (mottled clay horizon), was taken as the parent material. The calculation was made using Fe as a control because this element does not change appreciably during lateritic pedogenesis, that is, it presents an essentially residual behaviour. Given this assumption, gains and losses of mercury during weathering could be calculated for each horizon. A gain means that Hg has been introduced in the soil; a loss means that Hg has been leached out from the soil.

$Hg_i$  and  $Fe_i$  are the Hg and Fe contents in the  $i$ -horizon. The first step is to calculate  $Hg_o/Fe_o$ , which is the Hg/Fe ratio in the parent material. The residual accumulation of Hg in the  $i$ -horizon ( $Hg'_i$ ) can be calculated as follows:

$$Hg'_i = Fe_i \times (Hg_o/Fe_o) .$$

Gain or loss of mercury in the  $i$ -horizon (positive and negative  $Hg''_i$ , respectively) is obtained by subtracting  $Hg'_i$  from  $Hg_i$ :

$$Hg''_i = Hg_i - Hg'_i .$$

$Hg'_i$  and  $Hg''_i$  can be expressed as percentages of the original amounts:

$$Hg'_i \% = (Hg'_i/Hg_i) \times 100$$

$$Hg''_i \% = (Hg''_i/Hg_i) \times 100 .$$

Such calculations reveal gains of mercury in all horizons in both profiles. In profile B (Table III) gains of Hg decrease with depth:  $Hg''_i$  varies from 70 to 90% in the upper horizons and in the transition zone, decreasing to 53% in the uppermost levels of the mottled clay horizon and to 14% in the intermediate levels of the mottled clay horizon. In profile C (Table IV) important gains of mercury are observed in all horizons, except in the mottled clay at a depth of 240–250 cm, where the lack of data for Fe meant that the calculation could not be carried out.

In the Tartarugalzinho profile, mercury contents of more than  $100 \mu\text{g kg}^{-1}$  are present above the stone-line, decreasing downward to  $30 \mu\text{g kg}^{-1}$  in the saprolite horizon (Table II). In this profile, iron does not accumulate as nodules but, instead, is concentrated in the hematite-rich reddish mottles, abundant in the mottled clay horizon. The highest content of mercury in this profile ( $212 \mu\text{g kg}^{-1}$ ) was found in one of these mottles. Calculations (Table V) show gains of mercury between 9 and 79% across all horizons, except for the 380–400 m mottled clay horizon, where there is an accentuated loss of mercury.

The first conclusion that can be drawn from these data is that mercury content in soil profiles tends to decrease downward, just as it normally does in temperate or cold regions (Adriano, 1986; Fergusson, 1990; Inácio *et al.*, 1998). However, in contrast with the shallow accumulations observed in such climates, at Serra do Navio and Tartarugalzinho Hg contents in soils are quite high, exceeding  $300 \mu\text{g kg}^{-1}$ ,

TABLE III  
Balance calculations for profile B, Serra do Navio (n = nodules; m = matrix)

	Description	Hg <sub>i</sub> (μg kg <sup>-1</sup> )	Fe <sub>i</sub> (g kg <sup>-1</sup> )	Hg' <sub>i</sub> (μg kg <sup>-1</sup> )	Hg'' <sub>i</sub> (μg kg <sup>-1</sup> )	Hg' <sub>i</sub> (%)	Hg'' <sub>i</sub> (%)
0–20 cm	Argillaceous Cover	260	230	75	+185	29	71
40–50 cm (m)	Nodular Horizon	312	175	57	+255	18	82
40–50 cm (n)	Nodular Horizon	373	345	112	+261	30	70
140–150 cm (m)	Nodular Horizon	287	144	47	+240	16	84
140–150 cm (n)	Nodular Horizon	843	279	90	+753	11	89
200–250 cm	Transition Zone	300	180	58	+242	19	81
250–300 cm	Transition Zone	325	155	50	+275	15	85
300–350 cm	Mottled Clay	79	115	37	+42	47	53
350–400 cm	Mottled Clay	78	207	67	+11	86	14
		Hg <sub>o</sub>	Fe <sub>o</sub>	Hg' <sub>o</sub>	Hg'' <sub>o</sub>	Hg' <sub>o</sub>	Hg'' <sub>o</sub>
400–450 cm	Mottled Clay	81	250	81	0	100	0

$$\text{Hg}'_i = \text{Fe}_i \times (\text{Hg}_o/\text{Fe}_o).$$

$$\text{Hg}''_i = \text{Hg}_i - \text{Hg}'_i.$$

$$\text{Hg}'_i \% = (\text{Hg}'_i/\text{Hg}_i) \times 100.$$

$$\text{Hg}''_i \% = (\text{Hg}''_i/\text{Hg}_i) \times 100.$$

TABLE IV  
Balance calculations for profile C, Serra do Navio (n = nodules; m = matrix)

Description		Hg <sub>i</sub> (μg kg <sup>-1</sup> )	Fe <sub>i</sub> (g kg <sup>-1</sup> )	Hg' <sub>i</sub> (μg kg <sup>-1</sup> )	Hg'' <sub>i</sub> (μg kg <sup>-1</sup> )	Hg' <sub>i</sub> (%)	Hg'' <sub>i</sub> (%)
0–20 cm	Argillaceous Cover	203	71	11	+192	5	95
50–60 cm (n)	Nodular Horizon	203	364	56	+147	28	72
80–90 cm (m)	Nodular Horizon	139	111	17	+122	12	88
80–90 cm (n)	Nodular Horizon	152	272	42	+110	28	72
120–130 cm	Transition Zone	139	89	14	+125	10	90
170–180 cm	Mottled Clay Horizon	94	39	6	+88	6	94
240–250 cm	Mottled Clay Horizon	79					
		Hg <sub>o</sub>	Fe <sub>o</sub>	Hg' <sub>o</sub>	Hg'' <sub>o</sub>	Hg' <sub>o</sub>	Hg'' <sub>o</sub>
360–370 cm	Mottled Clay Horizon	29	189	29	0	100	0

$$\text{Hg}'_i = \text{Fe}_i \times (\text{Hg}_o / \text{Fe}_o).$$

$$\text{Hg}''_i = \text{Hg}_i - \text{Hg}'_i.$$

$$\text{Hg}'_i \% = (\text{Hg}'_i / \text{Hg}_i) \times 100.$$

$$\text{Hg}''_i \% = (\text{Hg}''_i / \text{Hg}_i) \times 100.$$

TABLE V  
Balance calculations for Tartarugalzinho profile

	Description	Hg <sub>i</sub> (μg kg <sup>-1</sup> )	Fe <sub>i</sub> (g kg <sup>-1</sup> )	Hg' <sub>i</sub> (μg kg <sup>-1</sup> )	Hg'' <sub>i</sub> (μg kg <sup>-1</sup> )	Hg' <sub>i</sub> (%)	Hg'' <sub>i</sub> (%)
0–100 cm	Argillaceous Cover	173	33	52	+121	30	70
100–200 cm	Mottled Clay Horizon	212	124	192	+20	91	9
200–300 cm	Mottled Clay Horizon	134	50	80	+54	60	40
300–380 cm	Stone line	100	26	41	+59	41	59
380–400 cm	Mottled Clay Horizon	100	136	215	-115	100	
400–450 cm	Saprolite	68	10	16	+52	23	77
		Hg <sub>o</sub>	Fe <sub>o</sub>	Hg' <sub>o</sub>	Hg'' <sub>o</sub>	Hg' <sub>o</sub>	Hg'' <sub>o</sub>
450–550 cm	Saprolite	30	19	30	0	100	0

$$\text{Hg}'_i = \text{Fe}_i \times (\text{Hg}_o/\text{Fe}_o).$$

$$\text{Hg}''_i = \text{Hg}_i - \text{Hg}'_i.$$

$$\text{Hg}'_i \% = (\text{Hg}'_i/\text{Hg}_i) \times 100.$$

$$\text{Hg}''_i \% = (\text{Hg}''_i/\text{Hg}_i) \times 100.$$

even at depths superior to 200 cm. These data point to an intense translocation of mercury down the profile, which has never previously observed. Existing studies of Hg content in Amazonian soils have indicated high levels, but were restricted to the topsoil (Pffeifer *et al.*, 1993; Rodrigues Filho and Maddock, 1997) or to the first 60 or 70 cm (Roulet and Lucotte, 1995; Roulet *et al.*, 1996; Roulet *et al.*, 1997).

In the soil profiles studied, high concentrations of Hg are related to high concentrations of Fe, especially in the mottles and in ferruginous nodules. High levels of mercury associated with decaying plant matter – typical of soils from temperate and cold regions – are not found in the soils studied.

Higher concentrations of Hg in the surface horizons of soils have usually been attributed to widespread atmospheric distribution and deposition (Malm *et al.*, 1991; Steinnes, 1995). Calculations for Hg in the profiles studied make it possible to use a quantitative approach to this hypothesis. The calculated gains of Hg ( $Hg_i''$ ) can be interpreted as derived from external sources through atmospheric transportation (mainly anthropogenic mercury). The remainder ( $Hg_i' = 100\% - Hg_i''$ ) is Hg derived from rock weathering, accumulated together with Fe which is naturally concentrated during lateritic pedogenesis (lithogenic mercury). Comparing lithogenic and anthropogenic contributions to the overall mercury in the upper horizons of profiles B and C (Tables III and IV), it is evident that the latter prevails, and at much greater levels than the average value of 3% estimated by Roulet *et al.* (1997). Anthropogenic mercury decreases with depth as a result of vertical redistribution of Hg due to leaching by rain water.

Another important conclusion can be drawn from contrasting Hg content in surface soils of regions both with and without mining activities. At Tartarugalzinho and Serra do Navio the Hg contents are similar, which points to the importance of long-range atmospheric deposition. So this phenomenon, which is generally uniform on a regional scale, can be seen as the main factor responsible for Hg contamination of Amazonian soils.

### 3.2. RELEASE OF MERCURY FROM SOILS

In the Serra do Navio region, the structure of soil profiles changes as one moves from upslope to downslope positions. In upslope profiles there is an iron duricrust which grades laterally into a nodular horizon in midslope and downslope profiles. This evolution is characterized by loss of iron and, probably, loss of mercury as well. In order to investigate this hypothesis, Hg burdens were calculated for soils in both topographic positions (Table VI).

The Hg burdens for the 0–20 cm soils of Serra do Navio, between 36 000 and 73 200  $\mu\text{g m}^{-2}$ , are comparable to those found by Roulet *et al.* (1997) in the 0–20 cm ferrallitic soils in the Tapajós valley (10 000 to 30 000  $\mu\text{g m}^{-2}$ ) and in French Guyana (60 000 to 70 000  $\mu\text{g m}^{-2}$ ). According to these authors, these burdens are 10 times greater than those observed in the upper layers of temperate and nordic soils.

TABLE VI  
Hg burdens in Serra do Navio soils

Profiles	0–20 cm ( $\mu\text{g m}^{-2}$ )	0–70 cm ( $\mu\text{g m}^{-2}$ )
A (upslope)	49 200	217 440
B (downslope)	44 880	182 160
D (upslope)	73 200	272 880
C (downslope)	36 000	118 800

Considering the two profiles on the southern margin, high on the slope (profile D) the cumulative burdens in 0–20 and 0–70 cm layers are respectively 272 880 and 73 200  $\mu\text{g m}^{-2}$ . The latter results are comparable to those obtained by Fadini and Jardim (1998) for the 0–100 cm soils in the Negro valley. Downslope (profile C) the cumulative burdens in the 0–20 and 0–70 cm layers decrease to less than half of the upslope cumulative burdens. The same tendency, albeit a reduced one, is observed in profiles A and B on the northern margin.

The loss of mercury in downslope profiles can be related to the natural evolution of iron duricrusts into latossols as a consequence of climatic changes toward increasing humidity in the Amazon (Nahon *et al.*, 1989; Tardy, 1991). In such conditions duricrusts are no longer stable, and are dismantled by the dissolution of hematite and goethite under the prevailing hydromorphic conditions. The nodular horizons in the downslope profiles (B and C) are the relicts of such a process. In this situation, both Fe and Hg are released into aquatic systems.

In the Amazon region the natural degradation of iron duricrusts into latosols, as a consequence of increasing humidity, is a slow process. It has been occurring over millions of years, ever since the Tertiary. However, it can be drastically intensified by human intervention, such as that witnessed at Mount Lofty, Australia (Fritsch and Fitzpatrick, 1994). In that region, the replacement of eucalyptus forests by pasture led to hydromorphism, eluviation and salinization of soils. This has resulted in a total degradation and erosion of the lateritic cover in less than one hundred years.

#### 4. Final Remarks

The high Hg contents of surface soils in the Amazon – generally higher than 100  $\mu\text{g kg}^{-1}$  and therefore well above the normal contents for uncontaminated soils (Adriano, 1986) – represent an anomalous regional background which is the result of multiple, long-term atmospheric contamination foci. Extremely high Hg contents in soils, above 1000  $\mu\text{g kg}^{-1}$ , are only found in specific locations, such as

where the gold-dealers burn amalgam (Rodrigues F<sup>o</sup> and Maddock, 1997; Hacon *et al.*, 1997b). This pattern of Hg pollution – generally high levels with well-defined, intense anomalies – is quite different from that found in sediments, where contamination from the pollutant source occurs more gradually.

Amazonian lateritic soils are important reservoirs for mercury, delaying its transference to aquatic ecosystems. Mercury accumulation in soils are in part derived from bedrock through ferrallitic pedogenesis, and in part from regional and long-range atmospheric deposition. Balance calculations meant it was possible to distinguish the amounts of mercury related to each one of these processes, and show that most mercury in the upper horizons of soil profiles stems from atmospheric sources, mainly of anthropogenic origin.

Investigations into the dynamic behaviour of waters that are currently percolating the soil profiles studied (Fostier *et al.*, 1997; Fostier *et al.*, 1999) corroborate this model. These authors observe that in the Serra do Navio area Hg content is 3.5–23.4 ng L<sup>-1</sup> in rainwater and 16.5–82.7 ng L<sup>-1</sup> in throughfall. This difference is attributed to the washing out of dry deposition. Once in the soil, Hg is distributed through the profile, being adsorbed, for the most part, on the iron oxyhydroxides. As a consequence, stream waters contain only 1.2–6.1 ng L<sup>-1</sup> Hg. These results confirm that these soils are efficient filters for mercury.

As Roulet *et al.* (1996) observed in the Tapajós valley, deforestation and agricultural practices can affect the pedological equilibrium, giving rise to intense leaching and erosion of soils. The acceleration of these processes and the consequent release of mercury into aquatic ecosystems presents a serious threat to the environment.

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