

Mercury emissions from forest burning in southern Amazon

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Received 17 December 2009; revised 7 April 2010; accepted 19 April 2010; published 15 May 2010.

[1] Several recent studies have indicated that forest fires are likely to re-emit important quantities of atmospherically deposited mercury (Hg) to the atmosphere. Although the Amazon forest accounts for approximately 25% of the world's total rainforest, few data are available about these emissions. The emissions of mercury from prescribed fires of two 4-ha plots of Amazon forest were investigated. Hg concentration and Hg burden were determined for vegetation, litter and soil before and after the fires. The data show that only Hg present in the aboveground vegetation and in the O-horizon was volatilized; no significant soil emission was observed. Before the fire, the Hg stored in the vegetation (logs, branches, leaves and litter) ranged from 3.7 to 4.0 g ha⁻¹ while 1.8 g ha⁻¹ was found in the O-horizon. The mass balance calculations of the present work indicate an average Hg emission of 3.5 g ha⁻¹ due to forest fires, with 1.6 ha⁻¹ originating from O-horizon and 1.9 from above ground vegetation. On the base of the average annual deforestation rate of the Brazilian Amazon between 2000 and 2008, an annual Hg emission of 6.7 Mg yr⁻¹ was estimated. **Citation:** Michelazzo, P. A. M., A. H. Fostier, G. Magarelli, J. C. Santos, and J. A. d. Carvalho Jr. (2010), Mercury emissions from forest burning in southern Amazon, *Geophys. Res. Lett.*, 37, L09809, doi:10.1029/2009GL042220.

1. Introduction

[2] Once released into the atmosphere, elemental mercury (Hg⁰) is oxidized to Hg(II), which is rapidly deposited and can be converted to methyl-mercury (MeHg), one of the most toxic Hg species that bio-accumulates in aquatic and terrestrial food chains. Mercury enters the atmosphere from natural and anthropogenic sources, and by re-emission of anthropogenic and natural mercury previously deposited onto terrestrial and water surfaces. In forest ecosystems mercury inputs are mainly associated with dry and wet atmospheric depositions [Grigal, 2003]. Forest canopies are effective in trapping atmospheric Hg because they present a high adsorption surface area for interception. Mercury compounds that accumulate on foliage are then transferred to the soil by precipitation and by litterfall deposition and its subsequent decomposition [Rea et al., 2002]. Understanding the relative importance of forested systems within regional or global

cycles is essential to developing a global Hg budget [Turetsky et al., 2006].

[3] Estimates of total mercury emissions ranged from 6060 to 6600 Mg y⁻¹, with one-third attributed to anthropogenic activities, one-third to natural emissions from land and oceans and the rest to re-emission of anthropogenic mercury [Swain et al., 2007]. However, emission of mercury from biomass burning is not mentioned in the study. On the other hand, global estimates of mercury emission related to biomass burning vary by an order of magnitude, including 380–1330 Mg y⁻¹ [Brunke et al., 2001], 250–430 Mg y⁻¹ [Sigler et al., 2003], 104–853 Mg y⁻¹ [Friedli et al., 2003], and 209–656 Mg y⁻¹ [Ebinghaus et al., 2007], showing the need for better estimation of this source of mercury emission.

[4] Mercury release during forest fires relates to: 1) volatilization of large amounts of mercury in biomass that is almost completely emitted into the atmosphere [Friedli et al., 2003]; and 2) thermal desorption of mercury from soil which is strongly dependent on temperature, Hg speciation and bonding type [do Valle et al., 2005]. During a forest fire, soil temperature increase mainly depends on fire severity, which can be influenced by variations of the fuel load, fuel and soil moisture, topography, weather and fire dynamics [DeBano, 2000]. Forest burning is therefore responsible for the re-emission of at least part of the accumulated Hg in biomass and soil and could sharply accelerate emission and deposition cycles between terrestrial ecosystems and the atmosphere. Although soil constitutes the largest reservoir of Hg in ecosystems, the soil contribution to Hg emission during forest fires still remains uncertain and needs to be better studied [Friedli et al., 2003; Engle et al., 2006; Turetsky et al., 2006]. The published data on Hg emission from forest fires in the Amazon region show a variation of one order of magnitude: 88 Mg yr⁻¹ [Veiga et al., 1994], 8.7 Mg yr⁻¹ [Lacerda, 1995], and from 6 to 9 Mg yr⁻¹ [Roulet et al., 1999]. This discrepancy can be mainly attributed to the lack of inventory of Hg content in the biomass and soils of Amazon forests.

[5] The purpose of this study is 1) to quantify the stock of mercury in the two main forest reservoirs (plants and soil), 2) to quantify mercury release during a planned “slash and burn” fire in an area located in the Amazon arc of deforestation, and 3) to estimate a budget for mercury emissions from Amazon forest fires. This work is part of a large program that studies biomass fire consumption and forest fire environmental impacts conducted in Northern Mato Grosso, Brazil [Carvalho et al., 2001; Rabelo et al., 2004; Christian et al., 2007; Neto et al., 2009].

2. Experiment

[6] The study was carried out in 2004 and 2005 at the Caiabi farm, near the city of Alta Floresta, state of Mato

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Table 1. Mercury Concentration (ng g^{-1}) in Vegetation From the Studied Area^a

Common Name	Latin Name	Hg (ng g^{-1}) Sampling Year	
		2004	2005
Leaves			
Açaí	<i>Euterpe oleracea</i> Mart.	35.1	57.9
Amescla	<i>Pogonophora schomburgkiana</i>	45.0	36.5
Cacauí	<i>Theobroma cacao</i> L.	60.0	26.1
Canela	<i>Aniba canelilla</i>	37.0	-
Embaúba Branca	<i>Cecropia</i> spp	49.0	30.7
Embaúba Vermelha	<i>Cecropia purpurascens</i> C. C. Berg	54.2	28.1
Ingá Amarela	<i>Ingá nobilis</i> Willd.	58.1	-
Laranjinha	<i>Guatteria citriodora</i> Ducke	-	36.8
Leiteira	<i>Brosimum parinarioides</i> Ducke	37.4	-
Marfim	<i>Calycophyllum acreanum</i> Ducke	-	19.3
Pata de Vaca	<i>Bauhinia rufa</i> (Bong.) Steud.	-	31.9
Quina	<i>Coutarea hexandra</i>	43.4	30.7
Tachi	<i>Triplaris surinamensis</i>	41.3	27.7
Tucum	<i>Bactris maraja</i>	-	33.4
Mean and standard deviation for leaves		46 ± 9	33 ± 10
Branches and Logs		<DL ^b	<DL
Litter		60 ± 10	111 ± 23

^aAll individual values correspond to the mean of three analytical replicates with precision <10%.

^bDL, detection limit (15 ng g^{-1}).

Grosso, Brazil. The vegetation in a $200 \times 200 \text{ m}^2$ area was felled during June. Fires were lit at the end of the dry season (Aug–Sep). Plant samples were collected just after felling of each plot. A forest inventory was performed in the central 1-ha area prior to the felling and vegetation samples were collected from the 10 dominant tree species found in this area. For each of the 10 dominant tree species, one composite sample of logs, one of twigs and one of leaves were collected from five trees of each species found in the area; 20 leaves/tree and 10 twigs/tree were randomly collected at different heights; for logs, one sample of sawdust was collected for each tree. Litter was also collected at five points uniformly distributed in the 1-ha area, but in this case the five samples were individually analyzed. Soil samples were collected at the same five points used for litter, before and after the burning. In 2004, soil sampling depths were 0–10, 10–20, 20–40, 40–60, 60–80 and 80–100 cm, and sampling was performed with a core sampler. In 2004 the O-horizon (layer of decomposing organic material which lies between litter and mineral soil) was collected together with litter. In 2005 the O-horizon was separately sampled and soil samples were collected at 0–2, 2–5, 5–10, 10–20 cm depths. Ash was also sampled at the same points after burning by scraping the soil surface (approximately 1 cm) and was actually a mixture of vegetal ash, charcoal and superficial soil. Fuel inventory and biomass consumption are described by Christian *et al.* [2007] and Neto *et al.* [2009].

[7] Vegetation and soil samples were dried in a laminar flux hood for 24 h. Vegetation samples were digested at 75°C with a mixture of concentrated HNO_3 and 30% v/v H_2O_2 , according to a method adapted from Rodushkin *et al.* [1999]; for soil samples, the US EPA method 3050B was used. For all samples, total Hg was determined by cold vapor atomic absorption spectrometry, using a Buck Scientific model 4000A Mercury Analyzer. Analytical accuracy was checked for every 6 samples using NIST- Standard Reference Materials® (SRM): SRM-1515 apple leaves and SRM-2709 San Joaquim soil. Error was less than 5% for soil and less than

11% for plants. Triplicate analyses showed an analytical precision <10% for vegetation and soils.

3. Results and Discussion

3.1. Mercury Concentrations

[8] Among the top 10 tree species in the area in 2004, seven were found in the area of 2005, showing similarity between the two areas (Table 1). The average Hg concentrations in leaves were not significantly different (Mann-Whitney U-test, $p > 0.05$). Although the mean concentration in the litter samples collected in 2005 was significantly higher ($p < 0.05$) than in the samples collected in 2004, no sound explanation was found for this difference. The present data for Hg concentrations in leaves and litter were of the same order as those found in various remote areas of the Amazon region [Roulet *et al.*, 1999; Mèlières *et al.*, 2003; Silva *et al.*, 2009]. The concentration of Hg in wood was always lower than the detection limit of 15 ng g^{-1} ; nevertheless, for calculation purposes, a concentration of 7.5 ng g^{-1} was considered for branches and logs, together with an uncertainty of 50%. For the Amazon forest, only Roulet *et al.* [1999] reported Hg concentrations in stems, twigs and branches, varying from 9 to 40 ng g^{-1} (mean = 27 ng g^{-1}). In deciduous hardwood and coniferous forests from the Northern Hemisphere, various studies have reported very low concentrations, on the order of 1 to 13 ng g^{-1} [Grigal, 2003].

[9] Hg concentrations in soil (0–100 cm) collected in 2004 varied between 67 and 134 ng g^{-1} and no significant difference was found when comparing mean concentrations before and after burning. It was then considered that Hg loss could account only in the upper soil layer (0–2 cm) and could be too small to be detected in 0–10 cm samples; the sampling strategy was therefore modified in 2005. The Hg concentrations found in the unburned soil collected in 2005 (Table 2) were of the same order of magnitude as the average ($61.9 \pm 50.6 \text{ ng g}^{-1}$) reported by Lacerda *et al.* [2004] for forest soils also collected in the Alta Floresta region. Other studies on

Table 2. Hg Concentration, Hg Burden, pH and Organic Matter Concentration in Soil and Ash, Before and After the 2005 Fire^a

	Hg Concentration (ng g ⁻¹)		Hg Burden (g ha ⁻¹)		pH		OM (%)	
	Before Fire	After Fire	Before Fire	After Fire	Before Fire	After Fire	Before Fire	After Fire
O-horizon (~1 cm)	96.5 ± 16.5	-	1.8 ± 0.3	-	6.6 ± 0.7	-	49.6 ± 15.9	-
Ash (~1 cm)	-	69.7 ± 13.0	-	5.6 ± 1.0	-	7.7 ± 0.5	-	22.0 ± 6.4
0–2 cm	94.1 ± 18.4	83.0 ± 14.6	15.3 ± 3.0	13.5 ± 2.4	6.2 ± 0.6	7.0 ± 1.0	19.8 ± 3.2	17.0 ± 2.0
2–5 cm	91.9 ± 24.6	89.2 ± 21.1	27.2 ± 7.3	26.4 ± 6.3	5.5 ± 0.6	5.3 ± 0.4	19.8 ± 3.8	18.2 ± 2.1
5–10 cm	59.0 ± 22.1	66.0 ± 37.1	29.5 ± 11.0	32.6 ± 18.6	4.8 ± 1.1	5.1 ± 0.7	20.9 ± 4.5	18.5 ± 1.2
10–20 cm	69.3 ± 32.0	67.5 ± 28.6	68.4 ± 31.6	66.7 ± 28.3	4.9 ± 0.8	5.4 ± 0.5	22.0 ± 5.1	18.9 ± 1.1

^aOM, organic matter. Concentration is mean ± standard deviation.

forest soils of the Amazon region show highly variable Hg concentrations (40 to 300 ng g⁻¹) [Roulet *et al.*, 1998; Fostier *et al.*, 2000; Magarelli and Fostier, 2005; Almeida *et al.*, 2005]. After burning, Hg concentration in ash remained at a relatively high value, comparable to the concentrations found at 5–20 cm. Although average Hg concentration in the 0–2 cm layer was lower after burning, the difference was not statistically significant ($p > 0.05$) when compared with the concentration before burning. Thermal desorption of mercury from soil is strongly dependent on temperature, Hg speciation and bonding type [e.g., *do Valle et al.*, 2005]. Although temperatures can reach 850 °C at the soil-litter interface, depending on fire severity, temperatures quickly decrease with depth and, at 5 cm in mineral soil, rarely exceed 150 °C [DeBano, 2000]. Some parameters as pH and organic matter content (OM) can be used as indicators of fire severity [Certini, 2005]. According to Arocena and Opio [2003], significant increases of pH occur only at high temperatures (>450–500 °C), in coincidence with complete combustion of fuel and consequent release of basic substances. In the 0–2 cm layer, pH values before and after burning were not significantly different (Table 2), indicating that the temperature in this layer probably stayed below 450 °C. In the O-horizon and at 0–2 cm depth, OM contents were 49.6 ± 15.9% and 19.8 ± 3.2%, respectively, the latter value being in agreement with (19.1 ± 5.9%) reported by Lacerda *et al.* [2004] for 0–2 cm forest soils of the same region. After the fire, the OM in ash was still 22.0 ± 6.4%, and did not change significantly in the 0–2 cm layer. As mentioned above, ash was actually a mixture of vegetal ash, charcoal and superficial soil, includ-

ing the burned O-horizon (which was no longer visible) (Figure 1). Considering that burned O-horizon and vegetal ash was almost completely mineralized, remaining OM can be attributed to soil sampled together with ash. According to Giovannini *et al.* [1988], substantial consumption of OM begins in the 200–250 °C range and is completed around 460 °C. The lack of change in OM content in the 0–2 cm soil layer suggests that the temperature had already sharply decreased in the first centimeter of soil (<200 °C). The dominant forms of Hg in soils are Hg(II) and Hg⁰, both in solution or adsorbed on soil particles. In most terrestrial systems Hg(II) is predominantly complexed with organic matter but it can also be bound to various ligands such as OH⁻ and Cl⁻ and associated to clay minerals [Grigal, 2003]. Some studies have shown that the desorption temperature of Hg bound to humic acid is 250–430 °C and <250 °C for Hg associated with mineral phase and that Hg⁰ and Hg²⁺ could be emitted below 150 °C [*do Valle et al.*, 2005]. In view of this data, it appeared that fire severity was therefore too low to promote significant desorption of Hg, likely mainly complexed with soil organic matter.

3.2. Mercury Burden and Mercury Emissions

[10] The calculated total dry biomass on the site before the fire was 306 and 258 Mg ha⁻¹ for 2004 and 2005, respectively (Table 3); an uncertainty of 30% has been estimated for the biomass loading and fuel consumption values [Christian *et al.*, 2007]. These values are in agreement with those published for other parts of the Amazon forest [e.g., Brown *et al.*, 1995; Carvalho *et al.*, 1998, 2001]. Mercury burdens in dry

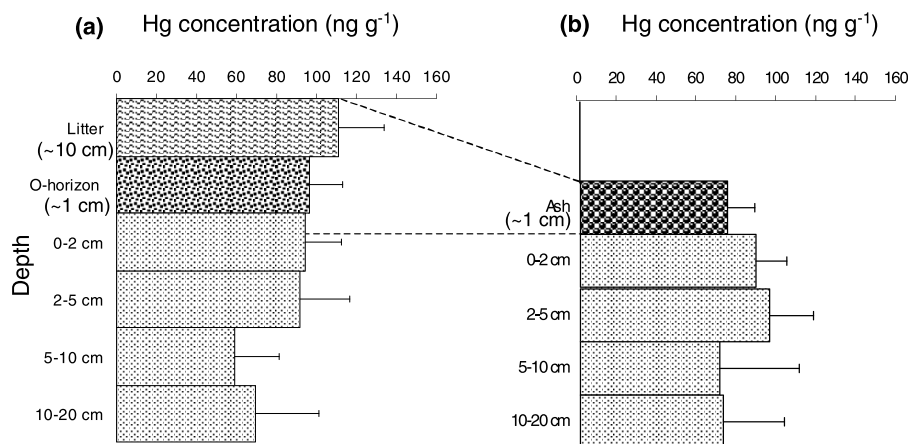


Figure 1. Hg concentration in litter, ash and soil (a) before and (b) after the fire of the 2005 experiment (error bars represent the standard deviation calculated from 5 samples).

Table 3. Total Biomass, Hg Burden and Hg Emission Factor for Biomass Burning

Biomass Category	Biomass Distribution (%)	Dry Biomass (Mg ha ⁻¹)	Hg Concentration (ng g ⁻¹)	MB ^a (g ha ⁻¹)	CF ^a (%)	RF ^a (%)	Hg EF ^a (g ha ⁻¹)
Logs, DBH > 30 cm	40.9	103.7	7.5	0.78	4.82	83	0.03
Logs, 10cm < DBH < 30 cm	19.1	48.4	7.5	0.36	16.9	83	0.05
Branches, D > 10 cm	23.5	59.6	7.5	0.45	16.9	83	0.06
Branches, D < 10 cm	6.5	16.5	7.5	0.12	88.6	83	0.09
Leaves	5.2	13.2	33	0.43	88.6	83	0.32
Litter	4.8	16.3	111	1.81	88.6	83	1.33
Total (2005)	100	257.6		4.0 ± 0.9 ^c			1.9 ± 1.5 ^c
Total (2004) ^b	100	306.2		3.7 ± 0.8 ^c			1.8 ± 1.5 ^c

^aMB, Hg burden in the dry biomass; CF, fraction of biomass combusted; RF, fraction of Hg released to the atmosphere from the complete combustion of biomass; Hg EF, Hg emission factor.

^bFor 2004 values, details of calculation are not shown.

^cFinal uncertainty was calculated by propagation of uncertainty, considering the uncertainty associated with each data mentioned in the text.

biomass were very similar in 2004 and in 2005 (Table 3). For the Amazon region, *Roulet et al.* [1999] calculated that 1.18 g ha⁻¹ was stocked in litter and 5.21 g ha⁻¹ in fresh vegetation (logs, branches and leaves). The higher value of Hg burden was mainly due to the relatively high Hg concentration they determined in wood (17 ng g⁻¹).

[11] The Hg emission factor from biomass combustion (HgEF, g ha⁻¹) was calculated according to the equation HgEF = MB * CF * RF, where MB is the Hg burden in the dry biomass, CF is the fraction of biomass combusted, and RF is the fraction of Hg released to the atmosphere from the complete combustion of biomass. CF for each size class was measured in the 2004 experiment [*Christian et al.*, 2007], and the same values were applied for the 2005 calculations. RF was obtained in a complementary study performed in laboratory by *Michelazzo* [2007], where eleven vegetation samples from different tree species and litter were burned, giving an RF of 83 ± 7%, which is in the same range of the 90% considered by *Veiga et al.* [1994]. The mean HgEF (1.9 g ha⁻¹) (Table 3) is comparable to the 2.3 g ha⁻¹ found by *Roulet et al.* [1999], for aboveground vegetation.

[12] The Hg burden in the O-horizon was significantly lower than in the first two centimeters of the soil (Table 2) due to the much higher density of this layer (0.814 g cm⁻³) when compared to the O-horizon (0.186 g cm⁻³). As mentioned above, O-horizon was no more visible after burning and RF = 90% was applied to calculate the O-horizon emission (1.6 g ha⁻¹). After the fire, the Hg burden in ash was 5.6 ± 1.0 g ha⁻¹. The ash layer density (0.810 g cm⁻³) confirmed that superficial soil (<1 cm) was sampled together with ash. In a complementary study performed in laboratory, *Michelazzo* [2007] found that Hg concentration in ash originating from litter and vegetation burning was always lower than 7 ng g⁻¹. *Friedli et al.* [2001], for similar conditions, reported values lower than 5 ng g⁻¹. In the light of these data, it seems reasonable to consider that Hg remaining in ash samples mainly originated from superficial soil collected together with ash. Summing up emission from biomass combustion (1.9 g ha⁻¹) and from O-horizon (1.6 g ha⁻¹) resulted in an overall emission of 3.5 g ha⁻¹. For Amazon aboveground vegetation, *Veiga et al.* [1994], *Lacerda* [1995] and *Roulet et al.* [1999] estimated Hg release around 13.5, 4.0 and 2.3 g ha⁻¹, respectively. In the Northern hemisphere, where more studies have been performed, total Hg emissions from different fires range from 1.5 to 5.3 g ha⁻¹ [*Engle et al.*, 2006]. *Wiedinmyer*

and *Friedli* [2007] reported 6.4 ± 1.1 g ha⁻¹ for fires in the United States.

4. Final Considerations

[13] According to National Institute for Space Research (2009, http://www.obt.inpe.br/prodes/sisprodes2000_2008.htm), the average annual deforestation rate of the Brazilian Amazon between 2000 and 2008 was around 1.9 × 10⁶ ha yr⁻¹. By applying the average Hg emission of 3.5 g ha⁻¹ to this area, an annual Hg re-emission of 6.7 Mg yr⁻¹ due to forest fires was estimated for this region. In performing such extrapolation, it was assumed that all fires in the Amazon region would result in the same EF, which has limitations because of the number of parameters that affect Hg emission, as discussed in the text (e.g., biomass loading, Hg concentrations in soil and vegetation, fire severity, etc.). Nevertheless, it was shown that biomass loading and Hg concentration in vegetation in the studied plots were in the same order of those in many Amazon regions; soil parameters and fires severity would therefore introduce the largest uncertainty on this extrapolation, pointing out the need for further studies on Hg emissions in other Amazon areas with different fire characteristics. For the United States, *Wiedinmyer and Friedli* [2007] estimated that an uncertainty in the Hg emission factors due to limited data leads to an uncertainty in the emission on the order of ±50%. This is a small portion of the global estimates of mercury emission to the atmosphere related to biomass burning (1 to 6%) when considering emissions of 104–853 Mg y⁻¹, estimated by *Friedli et al.* [2003]. Nevertheless, at a local/regional scale, the impacts of the annual mobilization of nearly 7 Mg of Hg needs to be further studied.

[14] **Acknowledgments.** Support of this research from the Fundação de Amparo à Pesquisa do Estado de São Paulo, FAPESP, Brazil (projects 04/01122-3, 03/07822-4 and 02/08964-4) is acknowledged. The burns were conducted under authorization of the Conselho Nacional de Desenvolvimento Científico e Tecnológico, CNPq (process CMC-004/04) and the Instituto Brasileiro do Meio Ambiente e dos Recursos Naturais Renováveis, IBAMA, Brazil. The authors also thank the owners of the Caiabi farm for permission to carry out the experiments and Carol H. Collins for English revision.

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