



Mercury sink in Amazon rainforest: soil geochemical data from the Tapajos National Forest, Brazil

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Abstract

Atmospheric mercury sink in Amazonian rainforest was tested in this work. Forest soil was analyzed for 11 soil profiles at depths of 0–40 cm in the northern portion of the Tapajos National Forest, Brazilian Amazon. Major oxides and Hg contents in soil and rock were determined by XRF and by Zeeman AAS, respectively. A mercury content of $146 \mu\text{g kg}^{-1}$ was found in the bedrock, whereas the mercury content in soil averaged $240 \mu\text{g kg}^{-1}$. The soil pH varied from 4.4 to 5.2 and the organic matter contents varied from 1.4 to 7.5%. Enrichment factors of Hg for soil were calculated in relation to the parental bedrock assuming aluminum as immobile element. The lower soil layers presented mercury enrichment of 29–98% in comparison to the bedrock. The upper soil layers showed mercury enrichment of 4–24% in comparison to lower soil layers. Mercury in soil was highly correlated with Al, Fe, and Ti; however, no significant correlation was observed between Hg and organic matter contents. All results pointed to mercury retention in forest soils as a result of the combined action of rock weathering processes and continuous input of metal from the atmosphere, estimated to be between 31 and 46% of the total Hg in the upper soil layers. These results are consistent with the hypothesis that the Amazon rainforest is a sink for atmospheric Hg and they also highlight the role of rainforest as a barrier for a number of substances associated with suspended atmospheric particulate.

Keywords Mercury · Soil geochemistry · Tapajos National Forest · Amazon · Brazil

Introduction

The geochemical pathways of mercury in Amazon have attracted the attention of researchers for many decades as an important research subject and theme of controversies. A number of studies performed in Amazon have focused on mercury distribution in rocks, sediments, soils, and water, as well as on mercury accumulation in fishes and human tissues. Several authors have discussed different aspects of biogeochemical cycling of mercury and highlighted negative effects of human exposure to mercury (Mason et al. 1994; Bidone et al. 1997; Malm 1998; Fostier et al. 2015).

High mercury concentration ($> 100 \mu\text{g kg}^{-1}$) in Amazonian soils has often been reported and the values far exceed Hg contents usually found in soils from other regions of the world (Roulet and Lucotte 1995; Bastos and Lacerda

2004). High mercury concentrations in soils from Amazon were interpreted either as a result of weathering of parental bedrocks (Brabo 2010; Roulet et al. 1998) or originated from atmospheric deposition influenced by metal emission due to artisanal gold mining in certain areas of the Amazon region. Several studies carried out in the Tapajos River and Madeira River basins (Amazon), where the artisanal gold miners made intensive use of mercury for amalgamation of gold, favor the latter explanation (Bastos and Lacerda 2004; Bidone et al. 1997). Atmospheric deposition of mercury has mainly been explained as a consequence of mining activity, so lesser attention has been placed on the role of other Hg natural and anthropogenic sources, which may be responsible for atmospheric deposition of the metal.

More recently, it was accepted that high mercury concentration in the environment may occur in Amazon at places away from the gold mining districts (Mascarenhas et al. 2004). In fact, Fadini and Jardim (2001) found high mercury concentration in air, water, and soil from the Negro River basin where gold mining or other industrial activity never existed. The authors estimated around $20.3 \mu\text{g m}^{-3} \text{ year}^{-1}$ of mercury deposition associated with natural rain precipitation

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in the Negro River basin. They also argued that the process of atmospheric mercury deposition, associated with gold mining emissions in Amazon, is not consistent with the volume of mercury stored in Amazonian soils.

Brabo (2010) investigated several soil profiles developed on different rock types from the Tapajós River basin and concluded that Hg enrichment in soil is due to pedogenic processes associated with rock alteration. In spite of the relative Hg enrichment towards the top observed in some local soil profiles by the author, the enrichment was explained in terms of Hg retention at iron hydroxide-oxide-rich soil horizons in the course of rock weathering. In agreement, mercury retention in soil has also been explained as a function of immobilization by Fe-rich lateritic horizons by Roulet and Lucotte (1995) in French Guiana and by De Oliveira et al. (2001) in the Amapá State, Brazil. The latter also considered Hg retention in soils as an important factor, which can influence Hg sink in Amazon.

It is worth mentioning that most rocks (granites, basalts, and sedimentary rocks) may contain significant amounts of mercury (averaging at $80 \mu\text{g kg}^{-1}$ according to Smith and Huyck 1999). The sedimentary rocks of the Upper Cretaceous Alter-do-Chão Formation, which are widespread in the central part of the Amazon Basin (Vasquez et al. 2008; CPRM 2006), are not an exception. This geologic unit includes a variety of rocks such as clayey sandstone, mudstone, conglomerate, and iron-rich beds, and stands as an ancient almost undisturbed natural mercury reservoir. Consistently, a large extension of soil profiles derived from these rocks can be seen in the central part of the Amazon region (IBGE 2008).

It is known that the global troposphere may contain 1.6 ng m^{-3} Hg originated from natural as well as anthropogenic sources. According to Mason et al. (1994), the present atmospheric Hg level far exceeds the mercury content of pre-industrial atmosphere of 0.5 ng m^{-3} . Mason (2008) indicates terrestrial Hg input of $1.850 \text{ Mg year}^{-1}$ and Hg input of $2.680 \text{ Mg year}^{-1}$ from the ocean. The author attributed only 5% of current terrestrial input to geogenic sources.

According to Lindqvist and Rodhe (1985), 80% or even more of the atmospheric mercury may occur as elemental Hg^0 , whilst the rest may be oxidized to Hg^{2+} in which form it is likely to be transported and deposited. Mercury (Hg^{2+}) is transported in the atmosphere probably as adsorbed phase onto suspended particulate matter, as suggested by Pacyna (1996). Hence, it is reasonable to consider that both the weathering of parental bedrocks and the mercury deposition from atmosphere may account for the current Hg content in soil.

The chemistry of atmospheric-suspended particulate matter in the Amazon region was approached by a few studies recently. Gonçalves et al. (2016) collected atmospheric particulate matter (APM) outside the Alenquer city, Pará State,

during the rainy and dry seasons of 2014. The authors indicated Na, Cl, K, NO_3^- , and SO_4^{2-} as the major chemical species in APM for both seasons and noted an increase in Al, Fe, and Mn concentrations in the dry season in comparison to the rainy season. Based on the chemical composition of particulates and on the air masses retrotrajectories in the sampling periods, they suggested the Atlantic sea aerosol, the continental resuspended soil, biogenic emissions, and the biomass burning as the probable sources of atmospheric particulate matter in the area. Figueiredo et al. (2015) reported elevated concentrations of Ca, K, Al, Na, and Fe in the fine fraction (particle size $< 2.5 \mu\text{m}$) of particulate matter collected in the northern part of the Tapajós National Forest in the dry season of 2012.

Deposition of retained particles on forest soil occurs by both precipitation and throughfall. Fostier et al. (2000) found mercury concentrations between 3.5 and 23.4 ng L^{-1} in rainwater and 16.5 – 82.7 ng L^{-1} in throughfall in a catchment in Serra do Navio (Amapá State, eastern Amazon). In addition, the chemical elements absorbed on the foliage, as previously described by Schroeder and Munthes (1998), will be added to topsoil by litterfall decay. Teixeira (2008) obtained Hg concentrations ranging from 60 to 215 ng g^{-1} in five tree species from the Atlantic Forest of Rio de Janeiro State, Brazil. A mercury annual mean concentration of $238 \pm 52 \text{ ng g}^{-1}$ in litterfall, which corresponds to the Hg flow of $184 \pm 98 \mu\text{g m}^{-2} \text{ year}^{-1}$ and litterfall production of 7.6 t ha^{-1} , was indicated by the author. More recently, Teixeira et al. (2017) estimated at $34.6 \pm 1.2 \mu\text{g m}^2 \text{ year}^{-1}$ the accumulation of Hg via litterfall flux in the Atlantic Forest. In the Negro River basin, Amazon, Magarelli (2006) obtained a mean annual deposition of mercury via litterfall of $43 \pm 14 \mu\text{g m}^{-2}$ with values ranging from 26 to $62 \mu\text{g m}^{-2}$. Fostier et al. (2015) indicated an average flux of $49 \pm 14 \mu\text{g m}^{-2} \text{ year}^{-1}$ for mercury deposition via litterfall in the Amazonian rainforest.

The present study focused on the occurrence of mercury in soil and bedrock, and it was carried out in the northern portion of the Tapajós National Forest, west Pará State, Amazon, Brazil. This study was conducted according to the hypothesis that the forest (approximately 15 – 45 km wide and 50 m high) acts as a barrier for the air masses, which then facilitate the deposition of suspended particulate matter originated from local and remote sources. The study aimed at assessing the processes that may be responsible for the high mercury contents reported for Amazonian soils.

Study area, sampling, and analytical methods

Soil and rock samples were collected in the northern portion of the Tapajós National Forest. The Tapajós National Forest (TNF) is a large (6000 km^2) environmental preservation area

bordered by the Tapajós River at west and by the Federal Road BR163 at east (Fig. 1). The predominant wind directions are from E and SE (Arbage et al. 2011; Gonçalves et al. 2016). To east and close to the border of the TNF, there are croplands and farms which cover a very large deforested area commonly affected by biomass burning at certain periods of the year. The sampling sites are reached by driving on the road BR163 from Santarem city towards south. The sites are located along secondary roads which cross BR163 at kilometers 67, 72, and 83 where soil profiles were dug and sampled. A total of 44 soil samples were collected from 11 soil profiles at 0–5, 5–10, 10–20, and 30–40 cm depth. One rock sample was collected from the sedimentary rock (clayey sandstone) outcropping in the study area. Sampling sites are indicated in Fig. 1.

In the study area, the soils developed from sedimentary rocks of the Alter-do-Chão Formation. This geological formation includes clayey sandstone interbedded with mudstone, arkosean conglomerate, and iron-rich horizons among other lithotypes of Upper Cretaceous age (Vasquez et al. 2008). Yellow and red clayey Oxisols as well as sandy soils are described in the Tapajós National Forest (IBGE 2008). Based on field observations, clay texture Oxisols dominated the sampled area and only site 5 showed sandy texture soils. The clayey Oxisols occur in a flat landscape in which Horizon A and B can be easily characterized in depth and soil horizons are well correlated laterally. Soil samples were collected from Horizon A and upper part of Horizon B (0–40 cm depth).

Changes in temperature, humidity, and precipitation are induced by alternate dry (August–December) and rainy (January–July) seasons.

The bulk mineralogy of soil samples was approached by X-ray diffraction analyses as reported by Figueiredo

et al. (2017). This previous study confirmed the presence of quartz and kaolinite as the single clay-mineral phase in soil accompanied by goethite and anatase.

Soil organic matter (OM) concentrations and pH values were determined for a subgroup of 28 soil samples. Soil pH was measured in water, whereas the OM concentration was determined by colorimetric method, according to EMBRAPA (1997).

Soil samples were dried at ambient temperature and only the grain size fractions less than 2 mm were stored for chemical analysis. The chemical composition of all samples was determined by X-ray fluorescence using pressed pellets. The quality of the results was checked by simultaneous analysis of the certificate reference materials SO-2, GSS-5, and GSS-6.

The mercury concentrations were determined by Zeeman atomic absorption spectrometry using the Pyro-915 + analyzer with an estimated lowest detection limit of 3 ng g^{-1} . Duplicate samples were also analyzed to check the quality of chemical analysis.

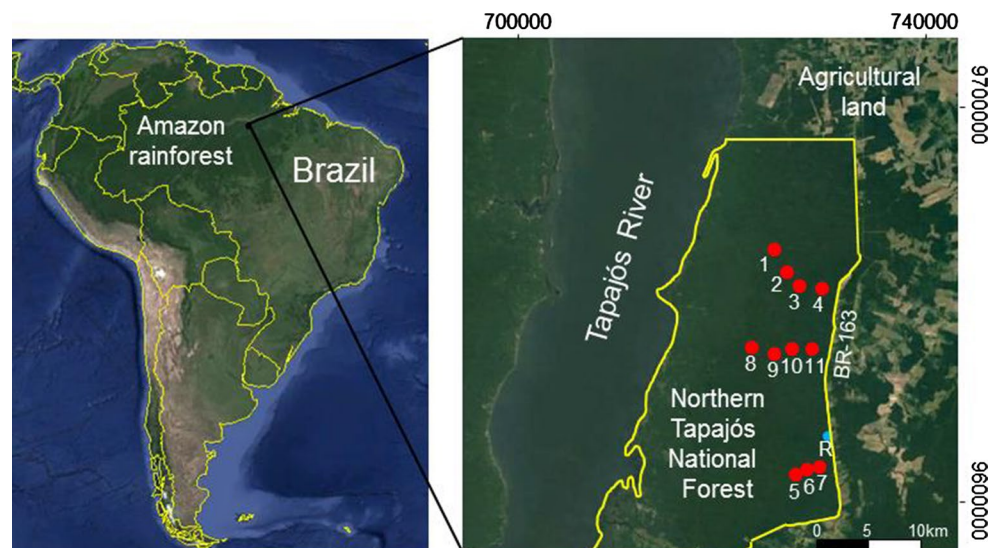
Enrichment factors were calculated using the chemical concentrations of elements in soil and rock samples and assuming that aluminum was immobile during soil formation. In this study, the following expression was used for calculation of enrichment factors:

$$EF = [C_s/Al_s]/[C_r/Al_r],$$

where C_s and C_r are element concentrations in soil and rock samples, and Al_s and Al_r are aluminum concentrations in soil and rock samples, respectively.

The Pearson correlation coefficient was calculated for all measured chemical parameters. In addition, mass balance was assessed to estimate the relative proportions between Hg-derived from the weathering of parental rock and Hg-derived from total atmospheric deposition in the

Fig. 1 Location map of the study area. Numbered dots refer to soil sampling sites (1–11); R dot refers to rock sampled site (R). BR-163 is the main access road. Modified from Landsat-7 ETM+ image (Dec. 12, 2015)



study area using the approaches suggested by De Oliveira et al. (2001) and Guedron et al. (2006).

Results

Mercury and major oxides concentrations in topsoil (0–5 cm depth) and bedrock samples are presented in Table 1. The soil samples exhibit a quite homogeneous chemical composition which is characteristic of yellow clayey Oxisol.

Major oxides contents were in the range of 37–46% SiO₂, 27–30% Al₂O₃, and 5–6% Fe₂O₃.

One soil profile (Site#5, 0–40 cm) showed a very distinct chemical composition (47–75% SiO₂, 16–32% Al₂O₃, and 2–6% Fe₂O₃). This sample was included neither in the calculation of average values shown in Table 1 nor in the determination of soil enrichment factor due to the fact that it represents a different soil type (sandy texture), less suitable to metal retention, as demonstrated by the lower Hg content (83 µg g⁻¹), in comparison to other soil samples. It is worth noticing that mineralogical and chemical similarities between the soils and the rock samples strongly suggest a genetic relationship between local clayey Oxisols and clayey sedimentary rock layers. However, this geochemical affiliation was not found for soil samples collected at site 5, which should be derived from a silica-rich layer of Alter-do-Chão Formation.

The chemical composition of the parental bedrock (46% SiO₂, 31% Al₂O₃, and 7% Fe₂O₃) was the expected chemical composition for clayey sandstone outcropping

in the area. Mercury concentration in the rock was 146 ng g⁻¹, whereas the mean Hg concentration in the soil was 244 ng g⁻¹ Hg for samples at 0–20 cm depth and 238 ng g⁻¹ Hg for samples at 20–40 cm depth. These values are consistent with mercury concentrations reported for soils from elsewhere in Amazon (Roulet and Lucotte 1995; Bastos and Lacerda 2004; Fadini and Jardim 2001).

Enrichment factors (EF) were calculated for the soil samples, as shown in Fig. 2. The EF curves show remarkably similar patterns for most soil profiles. Note that EF = 1 corresponds to the sampled rock modeled as parental bedrock. In respect to this baseline (EF = 1), samples from the bottom of soil profiles present a relatively large enrichment of mercury in the range of 29–98%, whereas samples from the top of the soil profiles showed enrichment in the range of 4–24% relative to Hg contents of samples from the bottom. This pattern is quite consistent for most soil profiles and may be the result of the combination of weathering of Hg-bearing parental bedrock with Hg input from the atmosphere. The latter was facilitated by specific functions of the forest as natural barrier for particulate carrying air masses, as well as by throughfall and litterfall processes associated with precipitation events.

No significant correlation was found between the concentrations of Hg and organic matter content in soil and this is in agreement with the previous studies (Roulet and Lucotte 1995). Mercury content highly correlates with Al, Fe, and Ti concentrations in soil (correlation coefficient values are 0.84, 0.83, and 0.83, respectively), which

Table 1 Mercury and major oxides contents in soil and bedrock from the Tapajós National Forest, Brazil

Soil samples (0–5 cm)	1	2	3	4	6	7	8	9	10	11	Average soil	Rock
pH	–	4.5	4.6	4.4	–	–	4.7	4.6	4.6	4.4	4.5	
%												
SiO ₂	38.95	38.64	40.79	36.90	46.37	42.90	38.77	39.67	39.64	39.64	40.23	46.21
TiO ₂	1.65	1.64	1.56	1.37	1.2	1.39	1.6	1.64	1.62	1.62	1.53	1.39
Al ₂ O ₃	27.66	28.92	29.79	27.28	26.96	29.57	29.44	29.77	29.83	29.83	28.9	31.24
Fe ₂ O ₃	5.34	6.11	5.72	5.26	5.25	5.3	5.75	6.25	5.94	5.94	5.69	6.68
MnO	0.014	0.005	0.004	0.005	0.003	0.005	0.005	0.004	0.004	0.004	0.005	0.003
MgO	dL	dL	dL	dL	0.02	dL	dL	dL	dL	dL	dL	0.08
CaO	0.38	0.05	0.05	0.08	0.06	0.06	0.06	0.01	0.05	0.05	0.08	0.05
Na ₂ O	0.08	0.07	0.07	0.07	0.08	0.08	0.08	nd	0.07	0.07	0.07	0.06
K ₂ O	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.01
P ₂ O ₅	0.10	0.03	0.04	0.05	0.04	0.03	0.05	0.04	0.03	0.03	0.05	0.01
OM	–	5.7	4.6	7.5	–	–	6.2	5.0	5.2	5.6	5.7	
LOI (105 °C)	4	2.83	2.64	3.73	2.65	2.37	2.78	2.57	2.68	2.68	2.89	1.42
LOI (1000°)	21.8	21.6	19.2	25.1	17.28	18.2	21.3	19.8	20	20	20.4	12.7
(µg/kg)												
Hg	205.8	264.7	288.9	291.4	190.4	203.5	292.5	257.4	223.5	236.2	245.4	145.8

OM organic matter content, LOI loss on ignition, dL below detection limit, – not determined

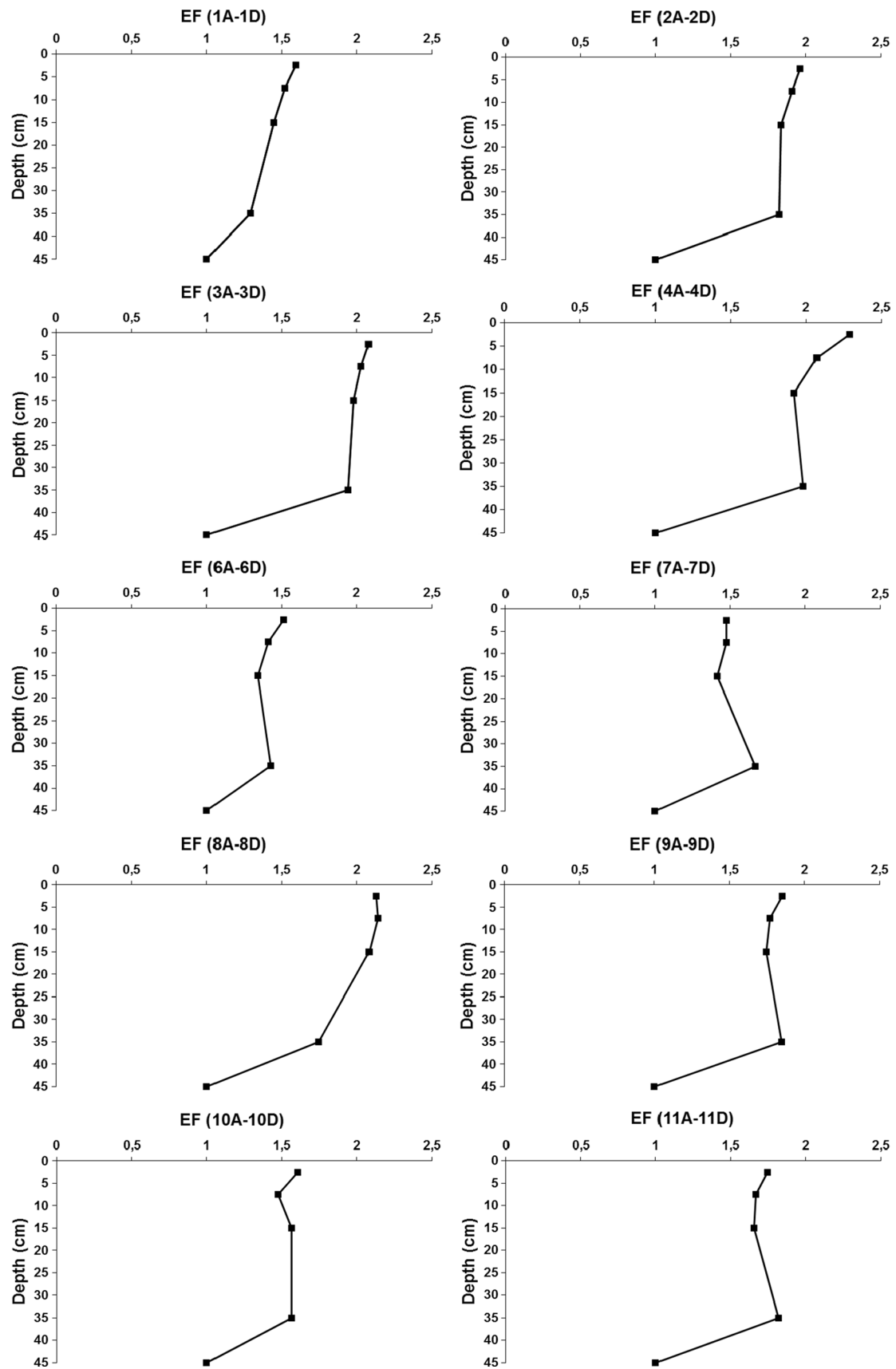


Fig. 2 Enrichment factor (EF) for different soil depths. Numbers and letters between parentheses refer to soil profiles sampled at different sites

has also been observed elsewhere in Amazon (Roulet and Lucotte 1995; De Oliveira et al. 2001).

The atmospheric contribution to mercury content in soil was assessed following the mass balance approaches as explained by De Oliveira et al. (2001) and Guedron et al. (2006). In the present study, the Fe, Ti, and Hg mean concentrations in the top soil (0–20 cm) and in the parental bedrock were used for mass balance calculation. The lithogenic mercury contribution accounts for 54–69% of the total Hg content in topsoil. Hence, the atmospheric Hg input was estimated at 31–46% of the total Hg concentration in topsoil. Atmospheric Hg may be associated with metal addition to the topsoil layer via wet deposition, throughfall and litterfall decay, disregarding the probable interference of Hg emission from forest soil as well as infiltration of a mercury soluble phase down the soil profile.

Concluding remarks

The consistent variability in the relative enrichment factors with depth for mercury in the studied undisturbed Amazon rainforest soil may result from both the combined actions of pedogenesis and the continuous and long-term metal deposition from the atmosphere. The Hg budget of the widely distributed sedimentary rocks of the Alter-do-Chão Formation may have significantly contributed to Hg enrichment in Amazonian soils through mercury retention in Oxisols. The role played by the rainforest, which facilitates atmospheric mercury deposition in the Amazon region, might have also been important. High Hg concentration in Amazonian soils is not restricted to certain mining areas where the metal is intensively used but can be also observed in other places where the rain forest acts as natural barriers for the Hg-bearing particulate matter carried by circulating air masses of local and remote origin.

Mason et al. (1994) estimated the current Hg global concentration in the troposphere at 1.6 ng m^{-3} and, more specifically, Fadini and Jardim (2001) indicated a mean Hg concentration of 1.3 ng m^{-3} in the atmosphere of the Negro River basin, northern Amazon.

The mode of occurrence of the atmospheric mercury was clarified by Lindqvist and Rodhe (1985) who suggested that Hg atmospheric deposition may occur as expenses of oxidized form Hg^{2+} adsorbed onto suspended particulate matter. The particulate matter may continuously accumulate on the trees from where it is washed (throughfall) facilitating the chemical compounds to reach the soil during the frequent rain events. In addition, the metal is also incorporated to the soil via wet deposition and by litterfall-binding metal processes, since part of the atmospheric mercury may be absorbed by the forest foliage. Fostier et al. (2015) estimated an annual removing of ca. 8% of the total atmospheric

mercury deposition in the Amazonian rainforest due to litterfall processes. In the Atlantic Forest (Brazilian Southeast), Teixeira (2008) estimated an annual mercury deposition of 8 t via litterfall and 28 t via throughfall. According to Schroeder and Munthes (1998), atmospheric Hg deposition via litterfall may be even more significant than deposition via throughfall.

Contribution of biomass burning to the suspended particulate matter in Amazon is well documented (Artaxo et al. 1988; Martin et al. 2010). One possible effect of biomass burning would be to increase the concentration of ozone (O_3) in the Amazon troposphere. This process may be important to oxidize atmospheric Hg^0 to Hg^{2+} or HgO , hence facilitating adsorption onto suspended particles in the air. In addition to the ozone, other oxidative agents associated with suspended atmospheric particulate matter may influence different areas in Amazon.

Considering that the current input of Cl^- to the Amazon forest may be significant, due to the transportation of aerosol masses from the Atlantic Ocean (Gonçalves et al. 2016), the soluble HgCl_2 is likely to form, facilitating infiltration of mercury down the soil profile and the metal retention onto Fe-rich soil horizons. This would occur at a favorable soil pH ($\text{pH} > 4$ according to Barrow and Cox 1992). Taking into account the probable infiltration of Hg down in the acidic soil profile, the persistency of high metal concentrations at the topsoil may indicate that the process of atmospheric deposition is a continuous phenomenon.

Besides mercury deposition, Figueiredo et al. (2017) indicated that other metals such as K, Sr, Na, and Mn displayed similar enrichment patterns towards the top of the forest soil profiles. Phosphorus and sulfur contents, which are highly correlated with soil organic matter concentrations, also show enrichment in the topsoil in the study area. All these results led to the conclusion that the atmospheric input of mercury and other metals to the soil is a general and significant phenomenon associated with the circulation of particulate matter in Amazon.

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