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Altered nature of terrestrial organic matter transferred to aquatic systems following deforestation in the Amazon

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ABSTRACT

Slash-and-burn agriculture practiced by several thousand small-scale farmers in the Tapajós region of the Brazilian Amazon has contributed to accelerated deforestation over the past decades. The present study aims to quantify and qualify changes in the transfer of terrestrial organic matter (TOM) to aquatic environments following deforestation. Lignin biomarkers have been analyzed from sediment cores collected in three floodplain lakes and in suspended particulate matter sampled during both wet and dry seasons. These analyses are interpreted with regard to lignin biomarker signatures of surface and deeper horizons of common soils, and of dominant plant species from forested and deforested environments. Dating the sediment cores with ^{210}Pb allows reconstructing the successive deforestation cycles since the onset of European colonization two centuries ago. Further, satellite images coupled to a GIS approach is used to correlate the evolution of sedimentary TOM and anthropogenic land-use from 1986 to 2009. Over this period, sedimentation rates have sharply increased, and the nature of the sedimentary TOM has been shifted from being linked to primeval forest soils to degraded soils following deforestation for subsistence cropping and/or pasture lands. The intensity of changes in the nature of sedimentary TOM appears inversely related to the connectivity of flood lakes to the river. In the least connected flood lake, weathering of pasture soils in the watershed dominates TOM inputs particularly during the dry season. Massive deforestation in the Amazon thus triggers major changes in the nature of TOM transferred and sedimented in aquatic systems.

1. Introduction

The Tapajós River region (State of Pará, Brazil) is a major settlement area where subsistence agriculture is one of the principal causes of deforestation (Le Tourneau and Bursztyn, 2010). Several studies conducted in the region have examined the impacts of this perturbation on local population health (Lebel et al., 1997; Passos and Mergler, 2008), ecosystem dynamics (Philipps, 1997; Roulet et al., 1999), and soil functions (Béliveau et al., 2009). Besides, intense soil podzolization and arenization on slopes, aggravated by deforestation and subsequent land uses leave the uppermost soil horizons vulnerable to erosion (Farella et al., 2001; Roulet et al., 1998). This triggers major changes in the transfer of terrestrial organic matter (TOM) to aquatic environments, and as such profoundly modifies the nature of the sediments (Farella et al., 2001, 2006; Roulet et al., 2000). In that sense, several studies conducted in the boreal environment have advocated the need of relating TOM weathering to watershed characteristics (Moingt et al., 2014; Ouellet et al., 2009; Teisserenc et al., 2010).

The landscape of the Tapajós region has been profoundly modified by slash-and-burn agricultural practices over the last fifty years (Béliveau et al., 2009; Metzger, 2003). The ashes of the burnt vegetation are used as natural fertilizer allowing crops to grow in the nutrient poor and acidic soil of the region (Fearnside, 1991). However, these practices only enhance soil fertility for one or two years, forcing the farmers to return their land to fallow for several years or to use it as pasture (Béliveau et al., 2009; Farella, 2005; Fearnside, 1991), and to repeat the slash-and-burn process on another piece of land (Metzger, 2003).

The present study was conducted as part of the Poor Land Use, Poor Health interdisciplinary research project, which studied the health risks of vulnerable communities of the Amazon deriving from environmental degradation. This study aims at qualifying and quantifying the changes in TOM transfers after deforestation from terrestrial to aquatic environments in three watersheds of the Tapajós region. As such, we relate the characteristics of a watershed (physical geography, vegetation cover and soil nature) to TOM found in the aquatic environment

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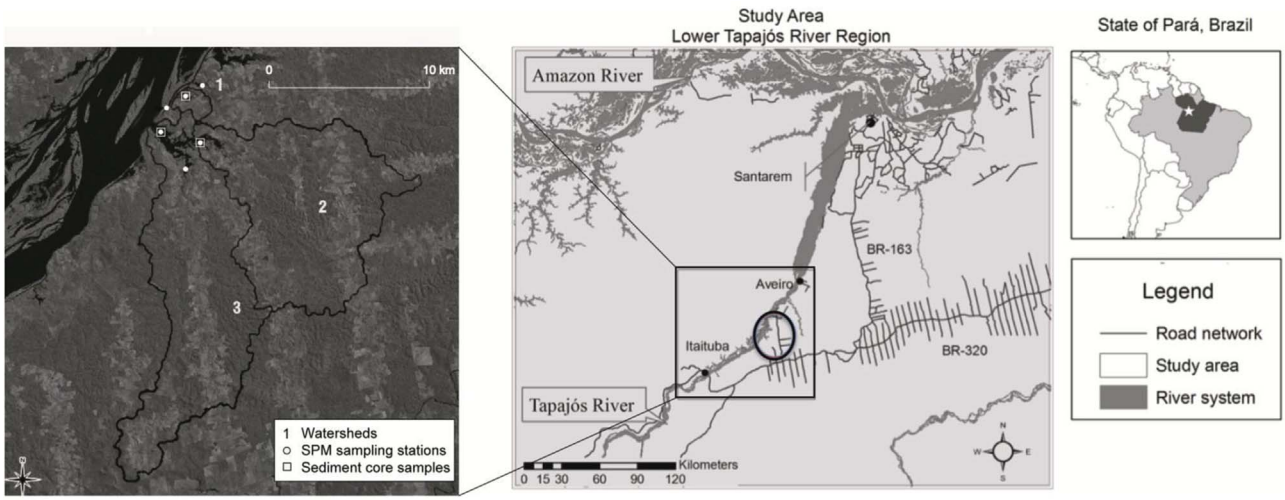

Fig. 1. Tapajos River region, watersheds delimitation (1 = Bom Intento, 2 = Demanda and 3 = Araipa), SPM and sediment core sampling sites selected for this study (modified from Oestreicher et al. (2016)).

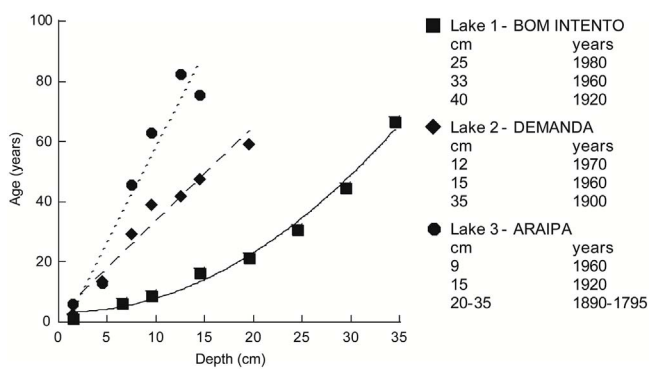

Fig. 2. ^{210}Pb sediment age prediction based on constant rates of supply model in the three lakes of the study.

(suspended particulate matter (SPM) and recent sediment). We use an approach combining Geographic Information Systems (GIS), ^{210}Pb core dating and lignin biomarkers. While lignin biomarkers have been widely used to evaluate the composition and source of TOM in aquatic environments (Gordon and Goñi, 2003; Hedges et al., 1986; Onstad et al., 2000; Prahl et al., 1994), and are considered as reliable tracers of TOM (Bélanger et al., 2015; Teisserenc et al., 2010), little is known about lignin transformations during its transit from the vegetal cover to the soil, water bodies and sediments in tropical environments (Bélanger et al., 2015; Feng et al., 2016).

2. Materials and methods

2.1. Study region

The sampling sites are located in the lower Tapajós region (Fig. 1). Historical data indicate that human settlement in the region occurred in successive episodes (Casa Civil, 2006; Oestreicher et al., 2016). The first settlement wave dates back from the 17th to the beginning of the 19th century, during the Portuguese expeditions that were conducted with the aim of taking possession of the land and establishing villages. The cities of Aveiro and Itaituba, in between which the studied lakes are situated (Fig. 1) were founded at this time (1781 AD and 1812 AD respectively). The region experienced a second major settlement wave coinciding with the rubber boom in the 1920s. Automobile industry magnate Henry Ford acquired more than 10,000 km^2 of land between Aveiro and Itaituba to exploit rubber. In the 1970's, the implementation of an integration and settlement policy at the regional level by the Brazilian government brought thousands of new families in the region. Colonists have practiced since then slash and burn subsistence agriculture, exposing cropland, fallow and pasture soils to heavy rains, which radically changed the nature of recent sediments (Farella et al., 2001; Roulet et al., 2000). One last major wave of immigration occurred in the 1980's with the gold rush (Cohenca, 2005) and along with the opening of secondary roads, such as the BR-163 connecting with the *Trans*-Amazonian highway (Fig. 1).

The region experiences alternating wet and dry seasons, and heavy precipitation during the wet season (1800–2200 mm of rain between December and April) (Interministerial, 2006) resulting in a several meter rise in river water levels, flooding the adjacent flatlands (De Oliveira Campos et al., 2001; Salati, 1986) and forming flood lakes. This hydrological system is characterized by an input of water from two separate sources in the wet season, one being the swollen river, and the other being the runoff from immediate surrounding watersheds. As it is difficult to quantify and distinguish water and SPM inputs coming from the river and from the watershed (Bonnet et al., 2008), the studied watersheds were chosen according to a gradient of interconnectivity between the flood lakes and the river (Fig. 2, Table 1a). In particular, Lake 1 (locally called Lake Bom Intento) is primarily fed by the small Watershed 1, being only connected to the Tapajós River in the wet season through a narrow channel. Lake 2 (locally called Lake Demanda)

Table 1
a and b: Location and morphological characteristics of the studied lakes and evolution of combined land use in the three studied watersheds. Percentages of land use exclude water surfaces in the watersheds. DA/LA = drainage area/lake area.

Table 1a	Watershed 1	Watershed 2	Watershed 3
Localization (lat./long.)	03.98S - 55.56W	04.01S - 55.55W	04.06S - 55.54W
Lake area (km^2)	0.27	2.99	2.99
Drainage area (km^2)	3.33	102.15	97.85
DA/LA	12.33	34.16	32.73
Mean slope (%)	10.28	8.14	9.11

Table 1b	Land uses for the three combined watersheds		
Year	1986	2001	2009
Forest (%)	80.6	64.8	48.9
Cropland, pasture (%)	3.7	8.6	16.4
Fallow forest (%)	12.1	21.4	26
Bare soil (%)	2.5	4.1	7.3

is linked to the river through the flood lake 3 and is influenced by its larger Watershed 2. Lake 3 (known as Lake Araipa) is in direct contact with the Tapajós River in the wet season, but also fed with by its larger Watershed 3. The three watersheds are strongly affected by slash-and-burn agriculture, with a heterogeneous and mosaic-like landscape.

2.2. Watershed analysis

The watersheds were analyzed from a spatiotemporal perspective. Landsat satellite imaging data collected during 1986, 2001 and 2009 and GIS was used to create a continuous time series (Rozon et al., 2015). The choice of years was guided by the quality and clarity of images available, and by the intervals between the years of study. Fairly regular intervals of at least five years between the selected years were selected to follow noticeable land use changes. Classification of land use was done using GRASS software based on spatial and spectral analyses (Rozon et al., 2015), for the three selected years. This classification was validated with over 200 control sites in the field. Four land uses, namely forest, fallow forest, cropland and bare soil and pasture, were selected in the present study. Watershed topography was established by distinguishing three classes of slope: (1) from 0% to 7.99%, (2) from 8% to 24.99% and (3) > 25%.

2.3. Sampling

Sediment cores were collected using a Wildco® hand core sediment sampler, 5 cm in diameter and 120 cm in length, with an extension of 3–6 m, depending on the depth of each sampled lake, lined with PVC tubing. The cores were collected from the central part of the flood lakes where water is present year-round even during the dry season. The sediment cores were then sub-sampled in 20 ml glass vials every centimetre using a Teflon® spatula within 12 h following extraction. Vials were pre-combusted at 500 °C for 3 h to avoid carbon contamination of the vessel and capped with Teflon® liners. In order to avoid cross contamination between samples, only the center of each slice was kept. Water content of each sample was determined by weighing sediment before and after freeze drying. The SPM was collected in both wet (February 2012) and dry (September 2012) seasons in every other day during a sampling period of four continuous weeks at six sites (Fig. 1). At each station, 50–100 L of water from the water column were collected with an electric pump and series filtered using 210 μm and 64 μm cartridge filters. SPM was then collected by tangential flow filtration using a Pellicon Millipore cartridge (previously sterilized via autoclave) and a Durapore 0.45 μm membrane, and concentrated at a volume of less than 1 L (Ouellet et al., 2009). The SPM used in the study therefore presented a size comprised between 45 and 64 μm. All samples were kept frozen (−20 °C) until being freeze-dried in the Montreal laboratories prior to analysis.

2.4. Geochemical analyses

Core and SPM samples were treated with vapor acidification in order to remove traces of inorganic carbon they may contain. Organic carbon (OC) analyses were conducted using a Carlo Erba (NA-1500) elemental analyzer with a relative precision of ± 5%. The OC contents were determined in all SPM samples, all samples in the top 15 cm of each core and samples at 5 cm intervals in the deeper layers of each core. Lignin biomarkers were extracted from the same sediment layers and on all SPM collected. Samples were oxidized with an alkaline cupric solution as described in Moingt et al. (2014). Briefly, the samples were weighed to contain between 2 and 5 mg of OC and mixed with 330 ± 0.1 mg of CuO and around 3.2 mL of 2N NaOH in a reaction bomb purged with N_2. Oxidation took place in a Hewlett Packard 5890A™ gas chromatograph oven modified by Prime Focus Inc. (Seattle, WA). Reaction bombs were heated to 150 °C for 150 min. After cooling to room temperature, 50 μL of internal standards (cinnamic acid and ethyl-vanillin) were added to the reaction bomb and the supernatant was decanted and acidified to pH 1 with HCl (2N). Then, a liquid-liquid extraction was conducted on the organic phase and dried by rotary evaporation. Finally, the extract was resuspended in pyridine and derivatized with N,O-bis(trimethylsilyl)trifluoroacetamide (BSTFA) and trimethylchlorosilane (TCMS; 99:1). A 2 μL extraction fraction was injected in splitless mode on a GC/MS (Varian 3800/Saturn, 2000™) fitted with a fused capillary column (DB-1 from J & W, 60 m, 320 μm). Helium was used as carrier gas whereas injector and detector were both held at 300 °C. The initial column oven temperature was set at 100 °C with a temperature gradient of 4 °C/min to 320 °C followed by a holding time of 10 min. In each sample injection series, SAG-05, a reference sediment sample, was included to evaluate analysis reproducibility trough time (Moingt et al., 2014).

2.5. Lignin indicators

The lignin derived-phenols are classified in four main families: vanillyls (V), p-hydroxyls (P), syringyls (S) and cinnamyls (C) (Hedges and Ertel, 1982). Based on these families, indicators have been used to evaluate sources and/or degradation state of the TOM in the aquatic systems (Hedges and Mann, 1979; Louchouarn et al., 1999; Prahl et al., 1994). Lambda is the sum of all eight lignin compounds, normalized to OC content (mg/100 mg of OC), while Sigma8 is the same sum but normalized to dry sample weight (mg/10 g of dsw). The latter two indicators thus express the amount of TOM with regards to OC or total sample weight (Houel et al., 2006). Vanillyls are ubiquitous in vascular plants, while cinnamyls are mostly present in non-woody plant species (herbaceous plants, leaves, needles) and syringyls are mostly abundant in angiosperms and absent in gymnosperms (Hedges and Mann, 1979). S/V ratio is usually used as a proxy to determine the relative contribution of angiosperm species to TOM content (Hedges and Mann, 1979; Tesi et al., 2008), but can also be used as an indicator of degradation since syringyls are more prone to degradation than vanillyls (Houel et al., 2006; Moingt et al., 2014; Opsahl and Benner, 1995). The C/V ratio is used as a proxy to decipher woody from non-woody plant parts since vanillyls are ubiquitous in vascular plants whereas cinnamyls are abundant in non-woody parts and almost non present in woody parts (Hedges and Mann, 1979). P/(V + S) and acid/aldehyde ratio of vanillyls ((Ad/Al)v) are both used as indicators of TOM degradation since degradation induces the loss of OCH_3 group in syringyls and vanillyls whereas *p*-hydroxyphenols cannot be affected, and that aldehyde functional group can be oxidized to carboxylic acid groups during degradation (Dittmar and Lara, 2001; Goñi et al., 1993; Hedges et al., 1988; Moingt et al., 2016; Opsahl and Benner, 1995).

2.6. ^{210}Pb dating

Radiometric measurements of ^{210}Pb activity were carried out using the method described by Moingt et al. (2014) in the three flood lake cores. Chronology was established using downcore unsupported ^{210}Pb and a Constant Rate of Supply (CRS) model (Ghaleb, 2009). Bonotto and García-Tenorio (2014) demonstrated that a CRS model provides more accurate results in Brazilian hydrographic basins when compared to other geochronological models. Cesium radioisotope (^{137}Cs) measurements were also realized in order to corroborate sediment age predictions via the CRS model (Ali et al., 2008).

2.7. Statistical analysis

Analyses of variation (ANOVAs) with two classification criteria were conducted on the SPM samples to ascertain the differences between sampling sites and seasons for all lignin biomarkers, as well as for OC contents. Pearson bivariate correlation tests were used to check the differences in lignin indicators and OC in relation to depth in the sediment cores. Covariance analyses between indicators within a

sediment core were also conducted. All statistical analyses were conducted using JMP 5.1 software (SAS).

3. Results

3.1. Watershed characteristics

Contingency analyses revealed that each year was different from the others, regardless of the watershed considered (Prob. > X^2, $p < 0.0001$). While obviously no topography change in either of the watersheds was observed between 1986 and 2009 the three combined watersheds experienced significant deforestation during that period with a drastic average decrease in forest area from 81% to 49% (Table 1b). In parallel, the average areas for cropland/pasture and fallow forest increased from 4% to 16% and 12%–26% respectively. Watersheds 2 and 3 had regular distributions of different land uses according to the slope classes (Bélanger, 2012). On the other hand, Watershed 1 distinguished itself by uneven land use distribution according to slope classes with most primary forest fragments found on gentle slopes (0–8% incline) and the majority of cropland, pasture, and fallow forest located on the steeper slopes (8–25% incline) (Bélanger, 2012).

3.2. Core dating

^{210}Pb dating revealed differences in CRS sediment rates in each lake even when they are adjacent to each other. Fig. 2 shows the results of ^{210}Pb dating in the three cores. Lake 1 has an average sedimentation rate of 0.78 cm/year with values ranging from 0.23 to 1.23 cm/year, the higher values corresponding to the surface layers. Lake 2 has an average sedimentation rate of 0.47 cm/year with values comprised between 0.19 and 1.10 cm/year. Finally, Lake 3 has an average sedimentation rate of 0.21 cm/year with values ranging from 0.09 to 0.44 cm/year.

3.3. TOM analyses in sediment cores

Lambda and Sigma8 indicators are frequently used to estimate the relative contribution of TOM to lake sediments (Houel et al., 2006; Moingt et al., 2014; Teisserenc et al., 2011). In Lake 1 sediment core, OC, Lambda and Sigma8 are considerably higher at the surface than in older sediment with values comprised between 1.8 and 14.4%, 0.75 and 8.85 mg/100 mg of OC and 4.73 and 127.12 mg/10 g of sample respectively (Fig. 3). In the three profiles, the same discontinuity is observed between 25 cm and 20 cm depth, which corresponds to the 1980–1990 period. The quantities of OC increase close to the surface ($p = 0.046$) and are correlated to both Lambda and Sigma8 fluctuations ($r^2 = 0.89$ and 0.93 respectively). This indicates that in Lake 1, increases in OC to the sediment correspond mainly consist of additional TOM inputs.

In Lake 2 sediment core, OC, Lambda and Sigma8 are also generally higher in the surface layers (Fig. 4). Values range from 2.4 to 9.7%, from 0.27 to 5.32 mg/100 mg of OC and from 0.71 to 51.73 mg/10 g of sample respectively. A discontinuity is also observed in this sediment core at 12 cm depth, which corresponds to around 1970 (Fig. 4). In this sediment core, OC values show a high variability throughout the entire core and are less stable than Lambda and Sigma8, suggesting a noticeable contribution of autochthonous organic matter. OC and Lambda profiles appear weakly correlated over the entire sediment core ($r^2 = 0.42$). If one only considers the surface layers above the discontinuity, OC and Lambda profiles become quite similar ($r^2 = 0.79$) suggesting that the TOM inputs from the watershed to the lake noticeably increased since 1970. OC and Sigma8 also show the same trends along the profiles after the 1970s ($r^2 = 0.68$).

In Lake 3 sediment core, OC, Lambda and Sigma8 are ranging from 1.8 to 8.6%, 0.56–23.15 mg/100 mg of OC and 1.03–137.26 mg/10 g of sample respectively (Fig. 5). In that core, high Lambda and Sigma8 values are observed in both surface and deeper sediment layers whereas OC values tend to decrease from the bottom to the top of the sediment core. The 20 to 15 cm depth interval corresponds to a period between 1850 and 1930. After this period, all three OC, Lambda and Sigma8 indicators concomitantly increase ($p = 0.0001$; 0.064; 0.0045).

Lambda and Sigma8 values measured in the sediment cores of the three lakes are in agreement with values reported in the literature for lignin biomarkers in sediment cores from tropical ecosystems (Farella et al., 2001; Pempkowiak et al., 2006; Sun et al., 2016). Measured S/V have average values of 1.04 ± 0.24, 0.78 ± 0.32 and 0.99 ± 0.20 whereas measured C/V have average values of 0.47 ± 0.18, 0.41 ± 0.42 and 0.23 ± 0.08 for lakes 1, 2 and 3 respectively (Fig. 6a, b and 6c). In Lake 1, both C/V and S/V significantly increase in the centimeters closest to the surface ($p = 0.001$; 0.0054), with the most marked increase occurring since the 1980s for both indicators ($p = 0.0041$; 0.0001) (Fig. 6a). In Lake 2, the discontinuity takes place early in the 20th century, with a significant change in TOM composition observable for both C/V and S/V at 30 and 35 cm depth respectively ($p = 0.0326$; 0.0004) (Fig. 6b). In Lake 3, S/V and C/V start to increase in the 1890's ($p = 0.0024$) and in the 1920s ($p = 0.0394$) respectively (Fig. 6c).

In Lake 1, P/(V + S) and (Ad/Al)v values range from 0.13 to 0.37 and from 0.43 to 0.78 respectively. No significant difference in P/(V + S) values could be observed in relation to depth (Fig. 7a). In Lake 2, P/(V + S) and (Ad/Al)v values are comprised between 0.06 and 0.76 and between 0.35 and 4.42, respectively (Fig. 7b). This two indicators show significant differences between the surface sediment (0–12 cm) and the rest of the sediment core (P/(V/S): $p < 0.0001$ and (Ad/Al)v: $p = 0.0298$). Since the 1970s, a strong correlation is observed between

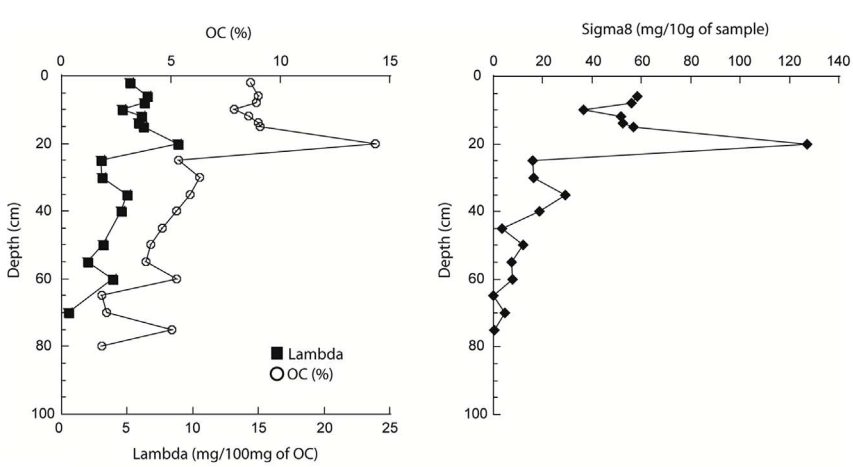

Fig. 3. Evolution of Lambda and organic content with depth in Lake 1 (Bom Intento) sediment core.

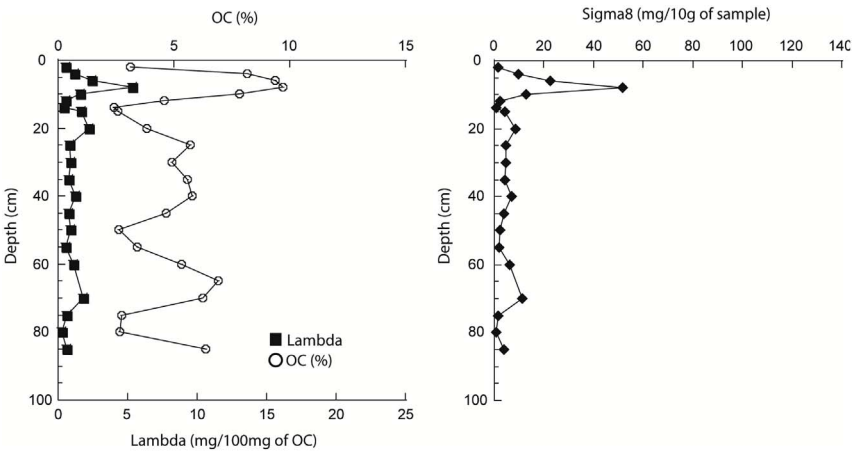

Fig. 4. Evolution of Lambda and organic content with depth in Lake 2 (Demanda) sediment core.

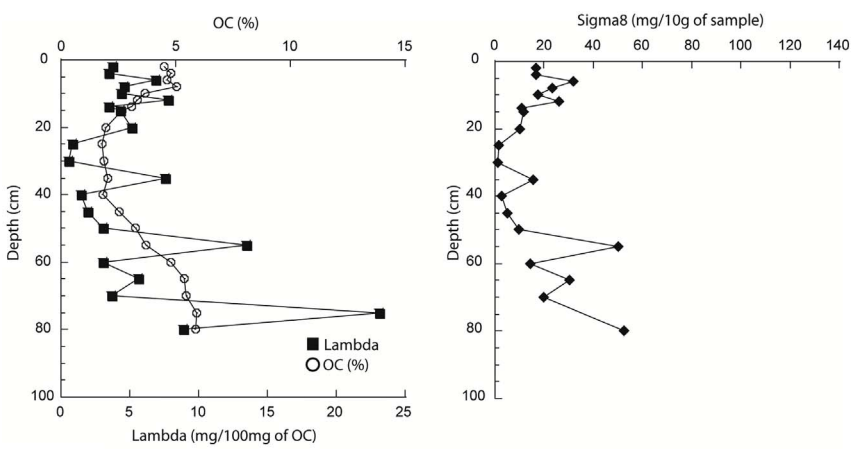

Fig. 5. Evolution of Lambda and organic content with depth in Lake 3 (Araipa) sediment core.

Sigma8 and P/(V + S) (r^2 = 0.86). Finally, in Lake 3, P/(V + S) and (Ad/Al)v values range from 0.11 to 0.54 and from 0.34 to 1.01 respectively (Fig. 7c). We observe the same phenomenon in Lake 3, except that the change in quantity and freshness occurred closer to the beginning of the 20th century ((Ad/Al)v: p = 0.0384).

3.4. Quantity and quality of suspended particulate matter

During the wet season, the SPM is characterized by Lambda and Sigma8 values significantly higher than in the dry season (p = 0.0008 and 0.0019) but comparable to those measured in the uppermost soil horizons of adjacent watersheds (Bélanger et al., 2015). During the wet season, the SPM Lambda values are fairly similar to those found in the uppermost soil horizon with an average of 2.87 ± 2.10 mg/100 mg of OC compared to an average value of 3.06 ± 1.41 mg/100 mg of OC (Bélanger et al., 2015). During the dry season, however, Lambda values are much lower with values closer to those measured in deeper soil horizons with average values of 0.31 ± 0.13 mg/100 mg of OC and 1.07 ± 1.30 respectively (Bélanger et al., 2015).

Fig. 8a illustrates S/V vs. C/V in SPM during the dry and wet

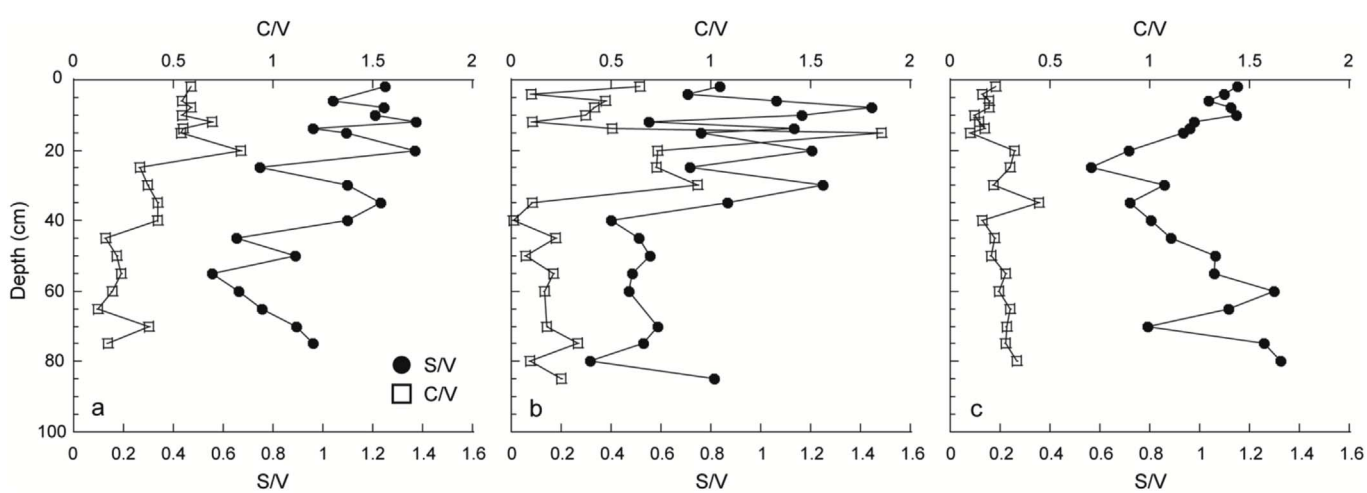

Fig. 6. Evolution of S/V and C/V ratios with depth in the three sediment cores of the study (a = Lake 1 (Bom Intento), b = Lake 2 (Demanda) and c = Lake 3 (Araipa)).

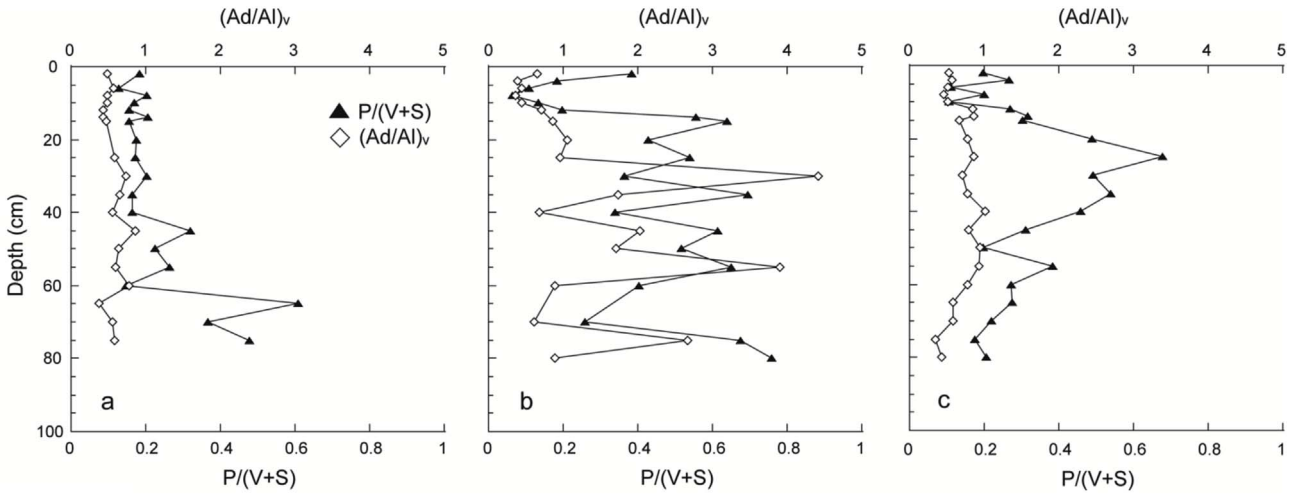

Fig. 7. Evolution of P/(V + S) and (Ad/Al)v ratios with depth in the three sediment cores of the study (a = Lake 1 (Bom Intento), b = Lake 2 (Demanda) and c = Lake 3 (Araipa)).

seasons. High C/V and S/V values are generally observed during the dry season and appear to correspond to the surface horizons of pasture soils. On the other hand, the SPM found in the aquatic system during the wet season shows mixed influences of forested and pasture soils from different horizons.

During the wet season, the (Ad/Al)v indicator in SPM (Fig. 8b) appears correlated to woody environments (SPM: 0.83 ± 0.26, forest soils: 0.74 ± 0.22) (Bélanger et al., 2015). This indicator is more similar to that of pasture during the dry season (SPM: 0.62 ± 0.10, pasture: 0.54 ± 0.23) (Bélanger et al., 2015). Moreover, a clear distinction between the wet and the dry season can be noticed when considering the P/(V + S) indicator with the higher values being measured during the dry season (Fig. 8b).

4. Discussion

4.1. History of colonization and its overall impacts on TOM transfers from terrestrial to aquatic environments

Soil lignin phenols may advantageously be used in studies of TOM transfers from watersheds to aquatic systems. Indeed, their signatures integrate various degradation processes that occur on pure vegetation sources in terrestrial environments (Bélanger et al., 2015; Farella et al., 2001; Houel et al., 2006; Moingt et al., 2016; Teisserenc, 2009). Sediment profiles of lignin biomarkers reported in this study exhibit important discontinuities through time, which do not necessarily occur simultaneously in the three lakes. The sediment core taken in Lake 3, going back till 1795, presents a marked discontinuity in Lambda and Sigma8 indicators, testimony of the first important deforestation activities that occurred in the region between 1795 and 1890 (Fig. 2). A clear discontinuity in the sediment profiles of lakes 2 and 3 is noticeable for a short period at the start of the 20th century (Fig. 6b and c). C/V and S/V values in sedimentary TOM then correspond to a marked influence of herbaceous vegetation and pasture soil after clear cutting the local primary forest for rubber tree monoculture. In the present study, significant changes in TOM profiles are clearly noticeable in Lake 2 core with a sharp OC, Lambda, Sigma8, C/V and S/V discontinuities at 12 cm depth and upwards (Figs. 4 and 6b). Sediment record of Lake 1 shows major changes in both the amount and composition of TOM starting in the 1980s, probably due to the establishment of several subsistence farming families in Watershed 1 at that time. In 1986, Watershed 1 was already partially deforested, and fallows and pastures were already observed in the landscape. Over the following years, deforestation in Watershed 1 increased, directly impacting the nature of TOM in most recent sediments (Figs. 6a and 8a).

4.2. Impact of land uses over the last decades on TOM transfers from terrestrial to aquatic environments

In a study describing lignin biomarkers of dominant plants and common soils of forested and deforested sites of the studied region, Bélanger et al. (2015) showed that Lambda and Sigma8 indicators in surface soil horizons (0–5 cm) are significantly higher than those of deeper horizons (20–55 cm). Bélanger et al. (2015) also emphasized significant differences between soil horizons for the degradation indicator P/(V + S). As such, P/(V + S) values in recent sediments of

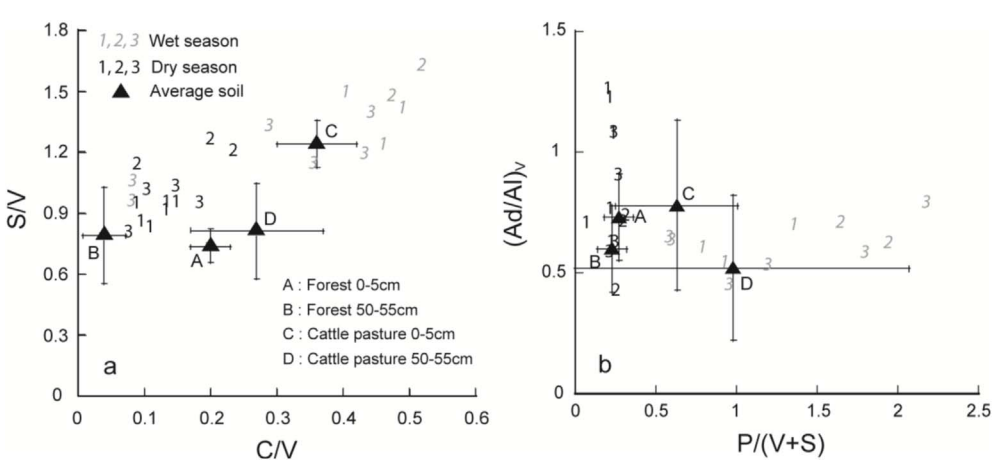

Fig. 8. S/V vs. C/V values and (Ad/Al)v vs. P/(V + S) values in SPM from the three watersheds (1, 2 and 3 markers corresponding to lakes 1, 2 and 3, respectively) during wet (blue) and dry (red) seasons. Mean soil values (Bélanger et al., 2015) are represented by solid black triangle with the associated standard deviation. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

lakes 1, 2 and 3 correspond to the average values reported by the previous authors for the uppermost soil horizon (0.24 ± 0.10) rather than those for deeper horizon (0.95 ± 1.02) (Fig. 7). Thus, both Lambda and P/(V + S) indicators suggest that TOM in recent sediments of any of the three studied lakes is dominated by weathered soil particles of uppermost soil horizon. These results corroborate those of Roulet et al. (2000) and Farella et al. (2001), who argued that deforestation along the Tapajós River contributes to massive sedimentation of particles from organic soil horizons in the aquatic system.

As far as the degradation index (Ad/Al)v is concerned, the observed values in recent sediments of the three studied lakes fall in between those found in soils and in fresh vegetation of the region by Bélanger et al. (2015) (Fig. 7). During the transfer from terrestrial to aquatic environments, biodegradation processes induce compositional changes of TOM either in the water column or in sediments (Dittmar and Lara, 2001; Opsahl and Benner, 1995; Orem et al., 1997; Tareq et al., 2004). However, recent studies have shown that TOM stability is likely due to biological and physicochemical interactions with the surrounding environment rather than to its intrinsic characteristics and thus can be variable (Schmidt et al., 2011; Thevenot et al., 2010). Since the (Ad/Al)v observed in the watersheds and the corresponding surface sediment cores are similar, it seems that in our case TOM degradation occurring in the water column is negligible and that TOM accumulating in recent sediments is composed of a mixture of TOM originating from both soils and fresh vegetation from the surrounding watersheds.

Besides, the ranges of C/V values measured in recent sediments of Lake 1 (0.5–0.9) and Lake 2 (0.2–0.6) are centered on the average value (0.31 ± 0.19) reported for cattle pasture soils by Bélanger et al. (2015) (Fig. 6a and b). Higher C/V values could rather indicate higher fractions of non-woody organic matter compared to woody organic matter (Hedges and Mann, 1979). Indeed, higher C/V values in sediments of lakes 1 and 2 may represent the direct influences of fresh cattle pasture grass, palm trees, vegetal material from the subsistence agricultural areas and/or macrophytes, which have C/V values around 1 (Bélanger et al., 2015; Bernardes et al., 2004; Hedges et al., 1986; Rezende et al., 2010; Zocatelli et al., 2011). The lower C/V values measured in Lake 3 (Fig. 6c) could represent a preferential degradation process of cinnamyls over vanillyls (Hedges et al., 1988; Opsahl and Benner, 1995). However, this explanation is not supported by the fact that as at the same time lignin degradation indicators should increase, which is not the case. If we assume that the watershed is the only input of TOM to the lake, a higher contribution of fresh woody material could explain a lignin signature with low C/V ratios and relatively unchanged degradation indicators. However, since Watershed 3 exhibits many common characteristics with Watershed 2 (e.g. slope distribution, land use, DA/LA) it is unlikely that the nature of the TOM inputs coming from the watershed be radically different. Thereby, another source of TOM to the sediments of Lake 3 has to be considered.

A S/V versus C/V plot is commonly used to identify TOM sources and estimate the relative contribution of gymnosperm/angiosperm and woody/non-woody parts to TOM (Hedges and Mann, 1979; Houel et al., 2006; Moingt et al., 2016). Since gymnosperms are uncommon in the Amazon, Bélanger et al. (2015) argued that both S/V and C/V can be used to discriminate forested and non-forested areas. Fig. 9 seems to confirm that the lignin signature of recent sediments from lakes 1 and 2 is due to TOM inputs from cattle pasture soils but mixed with inputs of fresh vegetal material (palm trees, cattle pasture grass and vegetal material from subsistence agriculture areas). On the other hand, Lake 3 recent sediments appear to be more influenced by TOM coming from fallowed forest and fragmented primary rainforest (Fig. 9).

On a longer term, the chronological analysis (1986, 2001 and 2009) of land use changes shows a drastic reduction in forest area from 1986 to 2009 (Table 1b). These land use changes correspond to noticeable changes in both quantity and quality of the TOM reaching lakes sediment. GIS analyses highlight the physiographical specificities of Watershed 1 as compared to watersheds 2 and 3. Watershed 1 has a small

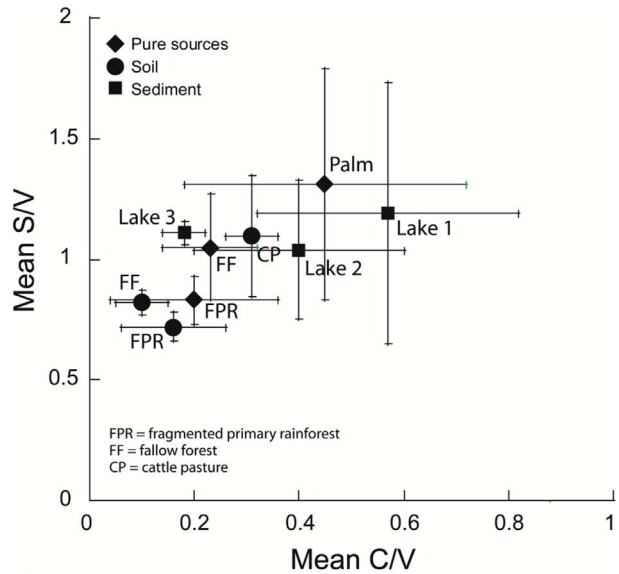

Fig. 9. Mean S/V vs. C/V values in pure sources (data from Bélanger et al., 2015; solid black diamonds), soils (data from Bélanger et al., 2015; solid black rhombs) and sediments (solid black squares) from the studied region.

drainage area coupled with a small lake surface area (DA/LA: 12.33, Table 1a). The sedimentation rate in Lake 1 is higher than the other watersheds (Fig. 2). A study conducted on TOM transfers from watersheds to lakes in boreal ecosystems determined that the average slope in the watershed plays a key role in lake sediment input (Teisserenc et al., 2010). These authors showed that moderate slopes between 4% and 11% promote TOM transfer between surface soil horizons and aquatic systems, while more gentle slopes encourage water retention, resulting in groundwater percolation through deeper soil horizons (Houel et al., 2006; Teisserenc et al., 2010). All three watersheds studied presenting moderate slopes on average (Bélanger, 2012), TOM in sediment cores effectively corresponded to soil particles eroded in the top 0–5 cm soil layers in the watersheds (Figs. 6 and 7). The average slope of Watershed 1 is slightly greater than those in watersheds 2 and 3 (Table 1a). In Watershed 1, however, cattle pasture and to less extent cropland dominate the moderate and steep slopes (8–25% incline; Bélanger (2012)). This could explain why the contribution of fresh vegetal material (both from cattle pasture and subsistence agriculture areas) was more important in Lake 1 in comparison to Lake 2 (section 4.1, Fig. 9).

4.3. Influence of the connectivity of the flood lake with the river on the TOM nature of its SPM and recent sediment

TOM lignin signatures in the SPM vary with the seasonal nature of particles weathered to the aquatic system. Lambda and Sigma8 values in the SPM (Table 2) are significantly higher during the wet season than the dry season (2.87 ± 2.10 mg/100 mg of OC versus 0.13 mg/100 mg 0.31 ± 0.13 mg/100 mg of OC and 82.18 ± 69.17 mg/10 g of sample versus 82.18 ± 69.17 mg/10 g of sample respectively). Although the Lambda values during the wet season are similar to the ones measured in surface soil horizons, the Sigma8 values are way higher rather suggesting a mixed contribution from all soil horizons and from fresh vegetal material (Bélanger et al., 2015). On the contrary, during the dry season the principal source of TOM to the SPM seems to be the surface soil horizons since the Sigma8 values are similar (Bélanger et al., 2015) whereas the lower Lambda values in the SPM could be explained by an important contribution of the primary production (Hélie and Hillaire-Marcel, 2006) to the organic pool thus diluting the Lambda indicator. These observations thus confirm that heavy rains during the wet season trigger TOM erosion from the loose organic horizons of forest and

Table 2
Lignin parameters for suspended particulate matter collected in the three studied watersheds during both wet and dry season.

	Wet season										
	Lambda	Sigma8	ΣS	ΣV	ΣC	ΣP	S/V	C/V	P/(V+S)	3.5-Bd/V	(Ac/Ad)v
Watershed 1	2.18	68.56	0.99	1.03	0.15	0.48	0.96	0.15	0.24	0.25	1.08
	9.44	294.10	4.18	4.81	0.45	0.59	0.87	0.09	0.07	0.25	0.71
	2.74	77.75	1.22	1.34	0.18	0.55	0.91	0.13	0.22	0.05	0.76
	2.52	73.18	1.17	1.24	0.11	0.50	0.94	0.09	0.21	0.22	1.22
	4.29	137.60	1.85	2.20	0.24	0.80	0.84	0.11	0.20	0.22	1.26
	2.51	86.13	1.15	1.20	0.16	0.52	0.96	0.13	0.22	0.19	0.63
Watershed 2	1.41	44.14	0.70	0.58	0.13	0.38	1.21	0.23	0.30	0.14	0.72
	1.35	40.06	0.70	0.55	0.11	0.39	1.27	0.20	0.31	0.13	0.74
	2.21	65.42	1.05	1.01	0.15	0.44	1.04	0.15	0.21	0.21	0.58
	1.98	54.54	0.89	0.93	0.17	0.48	0.96	0.18	0.26	0.26	0.91
Watershed 3	2.31	50.02	1.11	1.09	0.11	0.50	1.02	0.10	0.23	0.20	1.09
	2.38	37.90	1.02	1.26	0.09	0.56	0.81	0.07	0.25	0.19	0.63
	1.96	38.97	1.00	0.88	0.88	0.47	1.15	0.09	0.25	0.07	0.43
	Dry season										
	Lambda	Sigma8	ΣS	ΣV	ΣC	ΣP	S/V	C/V	P/(V+S)	3.5-Bd/V	(Ac/Ad)v
Watershed 1	0.48	14.03	0.22	0.18	0.08	0.37	1.25	0.46	0.94	0.56	0.55
	0.56	15.43	0.29	0.19	0.08	0.38	1.49	0.40	0.79	0.49	0.60
	0.28	7.66	0.14	0.10	0.05	0.32	1.41	0.49	1.36	0.60	0.70
Watershed 2	0.19	4.83	0.10	0.06	0.03	0.31	1.63	0.52	1.95	0.36	0.63
	0.26	5.81	0.13	0.09	0.04	0.37	1.48	0.47	1.65	0.38	0.70
	0.29	6.11	0.13	0.12	0.04	0.30	1.14	0.35	1.20	0.20	0.54
	0.33	6.35	0.17	0.13	0.04	0.29	1.33	0.29	0.96	0.70	0.45
Watershed 3	0.21	5.89	0.09	0.08	0.03	0.38	1.20	0.43	2.19	0.45	0.79
	0.21	5.32	0.10	0.07	0.03	0.32	1.40	0.44	1.81	0.34	0.59
	0.64	9.13	0.32	0.30	0.02	0.36	1.06	0.08	0.57	0.16	0.65
	0.65	9.39	0.31	0.32	0.03	0.36	0.96	0.08	0.58	0.27	0.64

cropland soils and facilitate the transfer of fresh vegetation material in nearby watersheds (Fig. 8a). Conversely during the dry season, little TOM of surface forest or cropland soils appears subject to direct erosion and further transfer to the aquatic environment (Sundborg and Rapp, 1986) while the weathering is probably insufficient to transfer material derived from the vegetation cover over a long distance. However, scarce and light rain events are still responsible for the weathering of surface pasture soils under the trampling action of animals (Bélanger et al., 2015). Pasture soils most probably contribute year-round to TOM weathering from terrestrial to aquatic systems, but appear masked by massive inputs of surface forest and cropland horizons (Fig. 8a and b). Bélanger et al. (2015) have also reported a distinctive P/(V + S) signature of palm trees (1.60 ± 0.51). It is then interesting to note that during the dry season, palm trees appear to influence the composition of TOM in the SPM (Fig. 8b). This observation seems to be directly linked to the fact that palm trees constitute a dominant feature of pastures in the Tapajós region (Gonçalves, 2010).

A factor that could explain the variable nature of TOM in SPM and sediments of a flood lake is its connectivity to the Tapajós River. Zocatelli et al. (2011) showed that when a sampling site in an Amazonian lake is connected to the river, its TOM appeared more degraded. Among the three studied lakes, Lake 1 is the least connected to the Tapajós River (Fig. 1). Consequently, TOM lignin signatures in both SPM and recent sediments appear quite similar to that of uppermost soil horizons in Watershed 1. On the other hand, Lake 3 is directly connected to the Tapajós River, which results in TOM lignin signatures in both SPM and recent sediments influenced both by direct weathering of uppermost soil horizons in Watershed 3 as well as TOM brought by the river to the lake. The latter is composed of TOM having resided a longer time in the water column and as such undergone degradation as indicated by lower cinnamyl and syringyl values, thus lower C/V and S/V values (Fig. 9) (Hedges et al., 1988). It should be noted that these lower values, as well as the Sigma8, (Ad/Al)v ones, correspond to the ones measured in SPM samples collected in the Tapajós River in the same region by Farella et al. (2001). Thus, lignin signatures observed in Lake 3 sediment core could represent a mixture of SPM from the Tapajós River with fresh woody material coming from the watershed (both having similar lignin signature). But considering the dominant influence of the Tapajós River on Lake 3 system, it is more likely that massive SPM inputs from the river overshadow the local inputs from Lake 3 watershed.

The important contribution of the Tapajós River to the sedimentary TOM pool of Lake 3 could also explain the fact that lignin indicators such as S/V and C/V are less variable in this lake than in the two others (Fig. 9). Indeed, the sporadic TOM inputs from the watershed should be diluted by the signal coming from the river. Although Watershed 2 exhibits many common characteristics with Watershed 3, notably slope distribution, land use, drainage area and lake area, C/V values in sediments of Lake 2 are generally higher than those of Lake 3 (0.2–0.6 vs.0.1 to 0.25) and more variable. This observation suggests that Lake 2 is indeed less influenced by TOM inputs from the river, being only marginally connected to it through flood Lake 3 (Fig. 1).

5. Conclusion

Successive waves of colonization in the Amazon over the past decades have profoundly altered the primeval ecosystem through massive and ubiquitous deforestation. Multiple land uses such as subsistence cropping and pasturelands resulted in enhanced weathering of soils formerly protected by the rainforest cover. As anthropogenic activities resulted in a highly heterogeneous landscape, a watershed approach was needed to follow the drastic changes affecting terrestrial organic matter transfers from terrestrial to aquatic environments. To do so, the study of lignin biomarkers in recent sediments proved to represent a powerful tool to quantitatively and qualitatively estimate TOM fluxes through time. Coupling these results to historical records of land uses revealed by satellite images, we could demonstrate that the impact of colonization on TOM dynamics was highly variable from one watershed

to another. When a floodplain system is weakly connected to the main river, the local erosion of degraded soils along with subsistence cropping or pastureland heavily contributes to the rapid accumulation of sediment in the lake, more so when the watershed slopes are moderately steep. This phenomenon appears particularly active during the dry season. On the opposite, in a floodplain system well connected to the river, the overall impact of regional deforestation and further land degradation modulates the sedimentary rate and nature in the lake. Linking deforestation to the alteration of the aquatic system integrity constitutes a major argument for promoting alternative sustainable land uses in the Amazon. Solutions though must be adapted in function of the local geography such as the degree of connectivity between a floodplain lake and the main river.

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References

Ali, A.A., Ghaleb, B., Garneau, M., Asnong, H., Loisel, J., 2008. Recent peat accumulation rates in minerotrophic peatlands of the Bay James region, Eastern Canada, inferred by < sup > 210 Pb and < sup > 137 Cs radiometric techniques. Appl. Radiat. Isotopes 66, 1350–1358.

Bélanger, E., 2012. Utilisation des biomarqueurs de la lignine comme traceurs de l'impact de l'occupation du territoire sur les transferts de matière organique terrigène entre les milieux terrestres et aquatiques en région tropicale. Master. Université du Québec à Montréal, pp. 109.

Bélanger, E., Lucotte, M., Grégoire, B., Moingt, M., Paquet, S., Davidson, R., Mertens, F., Passos, C.J.S., Romaña, C.A., 2015. Lignin signatures of vegetation and soils in tropical environments. Adv. Environ. Res. 4, 247–262.

Béliveau, A., Lucotte, M., Davidson, R., Do Canto Lopes, L.O., Paquet, S., 2009. Early Hg mobility in cultivated tropical soils one year after slash-and-burn of the primary forest, in the Brazilian Amazon. Sci. Total Environ. 407, 4480–4489.

Bernardes, M.C., Martinelli, L.A., Krusche, A.V., Gudeman, J., Moreira, M., Victoria, R.L., Ometto, J.P.H.B., Ballester, M.V.R., Aufdenkampe, A.K., Richey, J.E., Hedges, J.I., 2004. Riverine organic matter composition as a function of land use changes, southwest Amazon. Ecol. Appl. 14, 263–279.

Bonnet, M.P., Barroux, G., Martinez, J.M., Seyler, F., Moreira-Turcq, P., Cochonneau, G., Melack, J.M., Boaventura, G., Maurice-Bourgoin, L., León, J.G., Roux, E., Calmant, S., Kosuth, P., Guyot, J.L., Seyler, P., 2008. Floodplain hydrology in an Amazon floodplain lake (Lago Grande de Curuaí). J. Hydrology 349, 18–30.

Bonotto, D., García-Tenorio, R., 2014. A comparative evaluation of the CF: CS and CRS models in < sup > 210 Pb chronological studies applied to hydrographic basins in Brazil. Appl. Radiat. Isotopes 92, 58–72.

Casa Civil da Presidência da Republica, 2006. Plano de Desenvolvimento Sustentavel para a Area de Influência da Rodovia BR-163 Cuiaba - Santarem. Brasil, Brasilia, pp. 44.

Cohenca, D., 2005. A expansào da fronteira agricola e sua relaçào com o desmatamento detectado em imagens Landsat TM e ETM+ na regiào Norte da BR-163, Para entre os anos de 1999 a 2004. Santarém, Brasil, pp. 23.

De Oliveira Campos, I., Mercier, F., Maheu, C., Cochonneau, G., Kosuth, P., Blitzkow, D., Cazenave, A., 2001. Temporal variations of river basin waters from Topex/Poseidon satellite altimetry. Application to the Amazon basin. Comptes Rendus de l'Académie des Sci. - Ser. IIA - Earth Planet. Sci. 333, 633–643.

Dittmar, T., Lara, R.J., 2001. Molecular evidence for lignin degradation in sulfate-reducing mangrove sediments (Amazônia, Brazil). Geochimica Cosmochimica Acta 65, 1417–1428.

Farella, N., 2005. Les fermes des régions frontières d'Amazonie brésilienne: relations entre les origines familiales, les pratiques agricoles, les impacts sur les sols et le déboisement. Institut des Sciences de l'Environnement, pp. 185 Ph.D. UQAM.

Farella, N., Lucotte, M., Davidson, R., Daigle, S., 2006. Mercury release from deforested soils triggered by base cation enrichment. Sci. Total Environ. 368, 19–29.

Farella, N., Lucotte, M., Louchouarn, P., Roulet, M., 2001. Deforestation modifying terrestrial organic transport in the rio Tapajós, brazilian Amazon. Org. Geochem. 32, 1443–1458.

Fearnside, P.M., 1991. Développement agricole et déforestation en Amazonie Brésilienne. Cah. Des. Sci. Hum. 27, 235–253.

Feng, X., Feakins, S.J., Liu, Z., Ponton, C., Wang, R.Z., Karkabi, E., Galy, V., Berelson, W.M., Nottingham, A.T., Meir, P., West, A.J., 2016. Source to sink: evolution of lignin composition in the Madre de Dios River system with connection to the Amazon basin and offshore. J. Geophys. Res. Biogeosciences 121, 1316–1338.

Ghaleb, B., 2009. Overview of the Methods for the Measurement and Interpretation of Short-lived Radioisotopes and Their Limits. IOP Conference Series: Earth and Environmental Science. 5. IOP Publishing, pp. 012007.

Gonçalves, D., 2010. Phytosociological Structure of Forest Succession Ecosystems with Prevalence of F Attalea Maripa (Aubl.) Mart. And Astrocaryum Aculeatum G. Mey in Riverine Communities of Tapajos River. Ciencias Florestais. Master. Universidad federal rural da amazonia, Belem.

Goñi, M.A., Nelson, B., Blanchette, R.A., Hedges, J.I., 1993. Fungal degradation of wood lignins: geochemical perspectives from CuO-derived phenolic dimers and monomers. Geochimica Cosmochimica Acta 57, 3985–4002.

Gordon, E.S., Goñi, M.A., 2003. Sources and distribution of terrigenous organic matter delivered by the Atchafalaya River to sediments in the northern Gulf of Mexico. Geochimica Cosmochimica Acta 67, 2359–2375.

Hedges, J., Clark, W., Quay, P., Richey, J., Devol, A., Santos, U., 1986. Compositions and fluxes of particulate organic material in the Amazon river. Limnol. Oceanogr. 31, 717–738.

Hedges, J.I., Blanchette, R.A., Weliky, K., Devol, A.H., 1988. Effects of fungal degradation on the CuO oxidation products of lignin: a controlled laboratory study. Geochimica Cosmochimica Acta 52, 2717–2726.

Hedges, J.I., Ertel, J.R., 1982. Characterization of lignin by gas capillary chromatography of cupric oxide oxidation products. Anal. Chem. 54, 174–178.

Hedges, J.I., Mann, D.C., 1979. The characterization of plant tissues by their lignin oxidation products. Geochimica Cosmochimica Acta 43, 1803–1807.

Hélie, J.-F., Hillaire-Marcel, C., 2006. Sources of particulate and dissolved organic carbon in the St Lawrence River: isotopic approach. Hydrol. Process. 20, 1945–1959.

Houel, S., Louchouarn, P., Lucotte, M., Canuel, R., Ghaleb, B., 2006. Translocation of soil organic matter following reservoir impoundment in boreal systems: implications for in situ productivity. Limnol. Oceanogr. 51, 1497–1513.

Interministerial, G.D.T., 2006. Plano de Desenvolvimento Sustentável para a Área de Influência da BR-163. Coordenação da Casa Civil da Presidência da República, Brasília, Brasil, pp. 44.

Le Tourneau, F.-M., Bursztyn, M., 2010. Assentamentos rurais na Amazônia: contradições entre a política agrária e a política ambiental. Ambiente Sociedade 13, 111–130.

Lebel, J., Roulet, M., Mergler, D., Lucotte, M., Larribe, F., 1997. Fish diet and mercury exposure in a riparian Amazonian population. Water, Air Soil Pollut. 97, 31–44.

Louchouarn, P., Lucotte, M., Farella, N., 1999. Historical and geographical variations of sources and transport of terrigenous organic matter within a large-scale coastal environment. Org. Geochem. 30, 675–699.

Metzger, J.P., 2003. Effects of slash-and-burn fallow periods on landscape structure. Environ. Conserv. 325–333.

Moingt, M., Lucotte, M., Paquet, S., 2016. Lignin biomarkers signatures of common plants and soils of Eastern Canada. Biogeochemistry 129, 133–148.

Moingt, M., Lucotte, M., Paquet, S., Ghaleb, B., 2014. Deciphering the impact of land-uses on terrestrial organic matter and mercury inputs to large boreal lakes of central Québec using lignin biomarkers. Appl. Geochem. 41, 34–48.

Oestreicher, J.S., Lucotte, M., Moingt, M., Bélanger, É., Rozon, C., Davidson, R., Mertens, F., Romaña, C.A., 2016. Environmental and anthropogenic factors influencing mercury dynamics during the past century in floodplain lakes of the Tapajós River, brazilian Amazon. Archives Environ. Contam. Toxicol. 1–20.

Onstad, G.D., Canfield, D.E., Quay, P.D., Hedges, J.I., 2000. Sources of particulate organic matter in rivers from the continental USA: lignin phenol and stable carbon isotope compositions. Geochimica Cosmochimica Acta 64, 3539–3546.

Opsahl, S., Benner, R., 1995. Early diagenesis of vascular plant tissues: lignin and cutin decomposition and biogeochemical implications. Geochimica Cosmochimica Acta 59, 4889–4904.

Orem, W.H., Colman, S.M., Lerch, H.E., 1997. Lignin phenols in sediments of Lake Baikal, Siberia: application to paleoenvironmental studies. Org. Geochem. 27, 153–172.

Ouellet, J.-F., Lucotte, M., Teisserenc, R., Paquet, S., Canuel, R., 2009. Lignin biomarkers as tracers of mercury sources in lakes water column. Biogeochemistry 94, 123–140.

Passos, C.J.S., Mergler, D., 2008. Human mercury exposure and adverse health effects in the Amazon: a review. Cad. Saúde Pública 24, s503–s520.

Pempkowiak, J., Tylmann, W., Staniszewski, A., Golebiewski, R., 2006. Lignin depolymerization products as biomarkers of the organic matter sedimentary record in 210Pb-137Cs-dated lake sediments. Org. Geochem. 37, 1452–1464.

Philipps, O.L., 1997. The changing ecology of tropical forests. Biodivers. Conservation 6, 291–311.

Prahl, F.G., Ertel, J.R., Goni, M.A., Sparrow, M.A., Eversmeyer, B., 1994. Terrestrial organic carbon contributions to sediments on the Washington margin. Geochimica Cosmochimica Acta 58, 3035–3048.

Rezende, C.E., Pfeiffer, W.C., Martinelli, L.A., Tsamakis, E., Hedges, J.I., Keil, R.G., 2010. Lignin phenols used to infer organic matter sources to Sepetiba Bay – RJ, Brasil. Estuarine, Coast. Shelf Sci. 87, 479–486.

Roulet, M., Lucotte, M., Canuel, R., Farella, N., Courcelles, M., Guimarães, J.R.D., Mergler, D., Amorim, M., 2000. Increase in mercury contamination recorded in lacustrine sediments following deforestation in the central Amazon1. Chem. Geol. 165, 243–266.

Roulet, M., Lucotte, M., Farella, N., Serique, G., Coelho, H., Sousa Passos, C.J., De Jesus Da Silva, E., Scavone De Andrade, P., Mergler, D., Guimarães, J.R.D., Amorim, M., 1999. Effects of recent human colonization on the presence of mercury in amazonian ecosystems. Water, Air, Soil Pollut. 112, 297–313.

Roulet, M., Lucotte, M., Saint-Aubin, A., Tran, S., Rhéault, I., Farella, N., De Jesus Da Silva, E., Dezencourt, J., Sousa Passos, C.J., Santos Soares, G., Guimarães, J.R.D., Mergler, D., Amorim, M., 1998. The geochemistry of mercury in central Amazonian soils developed on the Alter-do-Chão formation of the lower Tapajós River Valley, Pará state, Brazil1. Sci. Total Environ. 223, 1–24.

Rozon, C., Lucotte, M., Davidson, R., Paquet, S., Oestreicher, J.S., Mertens, F., Passos, C.J.S., Romana, C., 2015. Spatial and temporal evolution of family-farming land use in the Tapajós region of the Brazilian Amazon. Acta Amaz. 45, 203–214.

Salati, E., 1986. The Climatology and Hydrology of Amazonia. Pergamon Press, Oxford, Amazonia.

Schmidt, M.W.I., Torn, M.S., Abiven, S., Dittmar, T., Guggenberger, G., Janssens, I.A., Kleber, M., Kogel-Knabner, I., Lehmann, J., Manning, D.a.C., Nannipieri, P., Rasse, D.P., Weiner, S., Trumbore, S.E., 2011. Persistence of soil organic matter as an ecosystem property. Nature 478, 49–56.

Sun, S., Schefuß, E., Mulitza, S., Chiessi, C.M., Sawakuchi, A.O., Zabel, M., Baker, P., Hefter, J., Mollenhauer, G., 2016. Origin and processing of terrestrial organic carbon in the Amazon system: lignin phenols in river, shelf and fan sediments. Biogeosciences Discuss. 2016, 1–29.

Sundborg, A., Rapp, A., 1986. Erosion and sedimentation by water: problems and prospects. Ambio 15, 215–225.

Tareq, S.M., Tanaka, N., Ohta, K., 2004. Biomarker signature in tropical wetland: lignin phenol vegetation index (LPVI) and its implications for reconstructing the paleoenvironment. Sci. Total Environ. 324, 91–103.

Teisserenc, R., 2009. Dynamique de la matière organique terrigène et du mercure dans les lacs et réservoirs boréaux. Institut des Sciences de l'Environnement, pp. 182 Ph.D. UQAM, Montréal.

Teisserenc, R., Lucotte, M., Houel, S., 2011. Terrestrial organic matter biomarkers as tracers of Hg sources in lake sediments. Biogeochemistry 103, 235–244.

Teisserenc, R., Lucotte, M., Houel, S., Carreau, J., 2010. Integrated transfers of terrigenous organic matter to lakes at their watershed level: a combined biomarker and GIS analysis. Geochimica Cosmochimica Acta 74, 6375–6386.

Tesi, T., Langone, L., Goñi, M.A., Turchetto, M., Miserocchi, S., Boldrin, A., 2008. Source and composition of organic matter in the Bari canyon (Italy): dense water cascading versus particulate export from the upper ocean. Deep Sea Res. Part I Oceanogr. Res. Pap. 55, 813–831.

Thevenot, M., Dignac, M.-F., Rumpel, C., 2010. Fate of lignins in soils: a review. Soil Biol. Biochem. 42, 1200–1211.

Zocatelli, R., Cecanho, F., Amorim, M., Bernardes, M., Moreira-Turcq, P., Turcq, B., Sifeddine, A., Cordeiro, R.C., 2011. Uso dos fenóis da lignina no estudo da matéria orgânica na várzea do Lago Grande Curuái, Pará e no Lago do Caçó, Maranhão, Brasil. Acta Amaz. 41, 195–204.