Marked-up manuscript

# Spatial and seasonal contrasts of sedimentary organic matter in floodplain lakes of the central Amazon basin

# R. L. Sobrinho<sup>1,2,\*</sup>, M. C. Bernardes<sup>1</sup>, G. Abril<sup>1,3</sup>, J-H. Kim<sup>2,4</sup>, C. I. Zell<sup>2</sup>, J-M. Mortillaro<sup>5</sup>, T. Meziane<sup>5</sup>, P. Moreira-Turcq<sup>6</sup>, J. S. Sinninghe Damsté<sup>2,7</sup>

<sup>1</sup>Universidade Federal Fluminense, Department of Geochemistry, 24020-141, Niteroi, Rio de Janeiro, Brazil

<sup>2</sup>NIOZ Royal Netherlands Institute for Sea Research, Department of Marine Organic Biogeochemistry, PO Box 59, 1790 AB Den Burg, Texel, The Netherlands

<sup>3</sup>Laboratoire EPOC, Université Bordeaux 1, CNRS UMR-5805. Avenue des Facultés, 33405 Talence, France

<sup>4</sup>Department of Marine Science and Convergence Technology, Hanyang University ERICA campus, 55 Hanyangdaehak-ro, Sangnok-gu, Ansan-si, Gyeonggi-do 426-791, South Korea.

<sup>5</sup>UMR-CNRS-IRD-UPMC 7208, BOREA, Département Milieux et Peuplements Aquatiques, MNHN, CP 53, 61 rue Buffon, 75231 Paris Cedex 05, France <sup>6</sup>IRD (Institut de Recherche pour le Développement) GET (Géosciences Environnement

Toulouse), Casilla 18-1209, Lima 18, Peru

<sup>7</sup>Utrecht University, Faculty of Geosciences, P.O. Box 80.021, NL-3508 TA Utrecht, The Netherlands

\*Corresponding author:

Rodrigo de Lima Sobrinho

Phone: (+31) (0)222-369406

E-mail: <u>rsobrinho@nioz.nl</u>

#### Abstract

In this study, we investigated the seasonal and spatial pattern of sedimentary organic matter (SOM) in five floodplain lakes of the central Amazon basin (Cabaliana, Janauaca, Canaçari, Miratuba, and Curuai) which have different morphologies, hydrodynamics and vegetation coverages. Surface sediments were collected in four hydrological seasons: low water (LW), rising water (RW), high water (HW) and falling water (FW) in 2009 and 2010. We investigated commonly used bulk geochemical tracers such as the C:N ratio and the stable isotopic composition of organic carbon  $(\delta^{13}C_{org})$ . These results were compared with lignin-phenol parameters as an indicator of vascular plant detritus and branched glycerol dialkyl glycerol tetraethers (brGDGTs) to trace the input of soil organic matter (OM) from land to the aquatic settings. We also applied the crenarchaeol as an indicator of aquatic (rivers and lakes) OM. Our data showed that during the RW and FW seasons, the surface sediments were enriched in lignin and brGDGTs in comparison to other seasons. Our study also indicated that floodplain lake sediments primarily consisted of allochthonous, C<sub>3</sub> plant-derived OM. However, a downstream increase in C4 macrophyte-derived OM contribution was observed along the gradient of increasing open waters, i.e. from upstream to downstream. Accordingly, we attribute the temporal and spatial difference in SOM composition to the hydrological dynamics between the floodplain lakes and the surrounding flooded forests.

Keywords: Amazon floodplain lakes, sedimentary organic matter, lignin phenols, GDGTs

#### **1** Introduction

Inland waters play a significant role in the global carbon budget. Lakes and rivers are active systems where the transport, transformation and storage of organic carbon (OC) affect the carbon cycle on a landscape and global scale (e.g., Cole et al., 2007; Tranvik et al., 2009; Raymond et al., 2013). In this context, the wetlands are dynamic interfaces between the terrestrial and aquatic realms, which promote the redistribution of carbon sources and sinks. Thus, they must be taken into account for the carbon fluxes and storage in the continents and for climate change mitigation strategies (Battin et al., 2009). Floodplain lakes are temporary or permanent water bodies formed in the wetlands of the Amazon basin. They are among the most productive ecosystems in the world (Junk, 1997; Melack and Forsberg, 2001). The primary production is performed by the flooded forests, macrophytes, phytoplankton, and periphyton (Junk et al., 2010). Inputs of CO<sub>2</sub> from plant respiration and reactive OC produced in floodplain lakes are significant sources of CO<sub>2</sub> outgassed in the central Amazon basin (Abril et al., 2014). The periodic floods intensify the exchange of organic compounds, nutrients and minerals between rivers, lakes and flooded soils (Junk, 1997). Although only 10-20% of the organic matter (OM) produced in the water column reaches the sediment and is finally buried (Devol et al., 1984), the sediments in these lakes are important sinks of carbon (Moreira-Turcq et al., 2004). Most of the sedimentary organic matter (SOM) in freshwater systems is derived from terrestrial vascular plants (Goñi and Hedges, 1992; Moreira-Turcq et al., 2004; Mortillaro et al., 2011). In the Amazon basin, many studies have characterized the suspended particulate organic matter (SPOM) in the rivers and the floodplain lakes and concluded that the main source of OM to the aquatic system is the forests and the upstream Andean soils (e.g., Hedges et al., 1986; Quay et al., 1992; Victoria et al., 1992; Hedges et al., 1994; Moreira-Turcq et al., 2004; Aufdenkampe et al., 2007; Mortillaro et al., 2011; Moreira-Turcq et al., 2013; Zell et al., 2013b). However, little is known about the molecular composition of the SOM in the floodplain lakes in general, and in particular, the contribution of the multiple sources of OM (upland soils, flooded and non-flooded forests, aquatic macrophytes, and phytoplankton) remains uncertain (Mortillaro et al., 2011; Zocatelli et al., 2011; Moreira et al., 2014).

The seasonality and the spatiality of the wetlands in the Amazon basin strongly influence the dynamics and the quality of OM in the surface sediments of floodplain lakes. Most of the SOM is supposed to be transported to the floodplain lakes via Amazon River mainstem during the rising and high water seasons (Hedges et al., 1986; Victoria et al., 1992; Moreira-Turcq et al., 2004; Mortillaro et al., 2011; Moreira-Turcq et al., 2013). However, a significant increase in the vertical flux of OM was observed in lake Curuai during the falling water season, which is interpreted as the result of resuspended sediments when the lake becomes smaller and shallower (Moreira-Turcq et al., 2004). In terms of the spatiality, the downstream lakes present higher values of  $\delta^{13}C_{org}$  in comparison to the upstream lakes (Victoria et al., 1992). This variability may be explained by the differences in the interfaces between the rivers and the lakes along the upstream-downstream transect or in aquatic primary production (mainly aquatic plants), which is more widespread in the open water lakes downstream. A previous study of bulk parameters and fatty acids in the central Amazon basin (Mortillaro et al., 2011) was not conclusive about the sources of SOM in floodplain lakes. Hence, the present work applies multiple biomarkers, namely lignin phenols, branched glycerol dialkyl glycerol tetraethers (brGDGTs) and crenarchaeol (isoprenoid GDGT), in addition to the bulk parameters, to disentangle the sources of SOM in floodplain lakes of the central Amazon basin and the role of the spatiality and seasonality in determining the composition of the SOM.

Lignin is produced by vascular plants. It is composed of phenolic compounds and general considered as a recalcitrant organic molecule. As a consequence, the products of CuO degradation of lignin (Hedges and Ertel, 1982) have been widely applied as biomarkers to trace plant material to aquatic systems (Hedges et al., 1986; Bernardes et al., 2004; Aufdenkampe et al., 2007; Kuzyk et al., 2008). Previous works in the Amazon basin showed that lignin is an important component of fossil OC in floodplain lakes (Zocatelli et al., 2013) but also a relevant carbon source for the outgassing of CO<sub>2</sub> in the Amazon River (Ward et al., 2013). This apparently contradiction reflects the relevance of environmental conditions on the degradation of organic molecules (Schmidt et al. 2012) which must be considered in the application of these biomarkers. The GDGTs are membrane lipids mainly composed of acyclic or cyclic biphytane core lipids with two glycerol head groups (Hopmans et al., 2000). The head groups are easily degraded while the two biphytanyl core lipids are well preserved in sediments and soils (White et al., 1979; Harvey et al., 1986). The GDGTs are found in diverse environments worldwide but, the brGDGTs are mainly produced in the soil, by the bacteria domain (e.g., Weijers et al., 2006), and the crenarchaeol is predominant in the aquatic environments and produced by Thaumarchaeota (Sinninghe Damsté et al., 2002). Accordingly, the relative amount of brGDGTs to the crenarchaeol, so-called the Branched and Isoprenoid Tetraether (BIT) index, have been proposed to quantify the OC proportion originating from soils and aquatic environments (e.g., Hopmans et al., 2004; Herfort et al., 2006; Belicka and Harvey, 2009; Smith et al., 2010). Previously, this method was successfully applied in rivers and floodplain lakes of the Amazon basin (e.g., Kim et al., 2012; Zell et al., 2013a; Moreira et al., 2014). A comparison between lignin phenols and GDGTs as markers for terrestrial OC has been performed in marine and lacustrine systems (e.g., Smith et al., 2010). This comparison showed complementary information on the transport and sedimentation of terrestrial OC in aquatic systems. Finally, the combination of these two groups of biomarkers with the bulk parameters, analyzed in superficial sediments collected in five floodplain lakes of the central Amazon basin in four hydrological seasons, provides new insights into the link between the hydrology of the Amazon basin to the sources of SOM in floodplain lakes.

#### 2 Study area

The Amazon River is the world's largest river with a drainage basin area of  $6.1 \times 10^6$  km<sup>2</sup> covering about 40% of South America (Goulding et al., 2003). The mean annual discharge is 200 × 10<sup>3</sup> m<sup>3</sup> s<sup>-1</sup> at Óbidos, the most downstream gauging station in the Amazon River (Callede et al., 2010). Rivers within the Amazon drainage basin are traditionally classified according to water color, as well as physical and chemical parameters (Sioli, 1950): white water (e.g., Solimões, Madeira, and Amazon rivers), black water (e.g., Negro River), and clear water (e.g., Tapajós River). The total area of wetland is  $350 \times 10^3$  km<sup>2</sup> (Melack and Hess, 2011). 17% of the central Amazon basin is subjected to periodical floods. This creates large temporary wetlands, i.e. seasonally flooded forests, woodlands, and shrubs, which corresponds to 58% of the total flooded area during the high water season. Aquatic macrophytes, floating meadow, and marsh cover 5 to 8% of the wetlands, and open waters correspond to 12 and 14% in low and high water seasons, respectively (Hess et al., 2003).

Five floodplain lakes were investigated in this study: Cabaliana, Janauaca, Mirituba, Canaçari, and Curuai (Fig. 1A, Table 1). The lakes are located along the Solimões-Amazon river shoreline in a biogeographic gradient of upstream flooded

8

forests to downstream flooded woodlands and open water lakes (Bourgoin et al., 2007; Abril et al., 2014). Cabaliana is a round shape lake surrounded by flooded forests and two sub-regions (Fig. 1B). In the northern region, the Manacapuru River discharges black water while in the southern region, the white water brought by the Solimões River, mixes with black water. Janauaca has a peculiar morphology with a ravine shape surrounded by flooded forests (Fig. 1B). Solimões water comes through the channel in the north, and some clear water comes through the stream system in the south. Conductivity values in Lakes Cabalina and Janauaca are close to that of the Solimões river, evidencing that white waters predominate. Mirituba has a round shape and receives white water from the Madeira River and the Amazon River through a complex drainage system (Fig. 1C). It is a white water lake surrounded by flooded forests and woodlands, with no significant contribution of black water streams. Canacari has two well-defined sub-regions (Fig. 1C). In the northern region, the Urubu River discharges black water and in the southern region, the Amazon River discharges white water. It is surrounded by flooded forests and woodlands and the conductivity is close to that of the white whaters of the Amazon River. Curuai is the largest lake in the central Amazon basin, mainly surrounded by woodlands and open waters (Fig. 1D). It receives in majority white water from the Amazon River through channels connected to the main stem, .Small contributions of black water streams occur in the Curuai floodplain, but remain spatially restricted to their most Southeastern region.

# **3 Materials and methods**

#### 3.1 Sample collection

Surface (0-2 cm) sediment samples (n=57) were collected using a grab sampler of 100 cm<sup>3</sup> in lakes Cabaliana, Janauaca, Mirituba, Canaçari and Curuai in the central Amazon basin between Manaus and Santarém (Fig. 1). The four hydrological seasons

were targeted during different research cruises with a small vessel (Fig. 2): in June and July 2009, which covered the high water (HW) season; in October 2009 the low water (LW) season, in August 2010 the falling water (FW) season and in January 2011 the rising water (RW) season. In each floodplain lake, sediment samples were collected at three stations in each season. However, sometimes only two samples were collected when stations were not accessible during a specific season. Thus, it was obtained approximately 12 samples in each lake and 15 samples per season. The samples were collected in the three most distinct sites of each lake: near the connecting channel, in the middle of the lake and near the flooded forests. Most of the sampling sites were located in areas flooded by the Solimões-Amazon River water (white water). In Cabaliana and Canacari one station was located near the black water streams in order to represent the heterogeneity of the lakes.

Four wetland soils and three nonfloodable soils from well above the maximum inundations levels, known as "terra firme", were also collected during the LW season. In addition, four samples of C<sub>3</sub> (*Eichornia sp., Pistia stratiotes*) and C<sub>4</sub> (*Paspalum repens*) aquatic plants (macrophytes) were sampled during the HW season in the lakes Janauaca and Curuai. All samples were kept frozen (-20°C) on the ship and transported frozen to the Universidade Federal Fluminense laboratory (Brazil), where they were freeze-dried.

#### 3.2 Bulk geochemical parameters

For the samples collected during the HW and LW seasons, total carbon (TC), total nitrogen (TN), and  $\delta^{13}$ C of TC were determined at the Davis Stable Isotope Facility (Department of Plant Sciences, University of California at Davis, California, USA) using a Europe Hydra 20-20 mass spectrometer equipped with a continuous flow isotope ratio monitoring device. Other samples gathered during the FW and RW cruises were analyzed for TC, TN, and  $\delta^{13}$ C of TC using a Flash 2000 organic elemental

analyser interfaced with a Delta V advantage isotope ratio mass spectrometer at Royal Netherlands Institute for Sea Research (NIOZ, The Netherlands). The average precision was ±0.1 mg C g<sup>-1</sup> for TC and ±0.05 mg N g<sup>-1</sup> for TN. In addition, sixteen decarbonated sediment samples were analyzed for the total organic carbon (TOC) contents at NIOZ and at Universidade Federal Fluminense (UFF) using a Carlos Erba elemental analyser EA 1110. These analyses were determined in duplicate with a precision of 0.1 mg C g<sup>-1</sup>. TC (wt.%) correlated very well with TOC (wt.%) with an intercept no significant from 0 ( $R^2$ =0.96, p<0.001, n=16). This indicates that TC in floodplain lakes sediments investigated was predominantly TOC and the fraction of carbonates was minor. Therefore, TC was considered as TOC in this study. In order to assess contribution of inorganic nitrogen ( $NH^{+4} + NO^{-2} + NO^{-3}$ ) to TN, TN (wt.%) and TOC (wt.%) were correlated (R<sup>2</sup>=0.89; p<0.001; n=16). The interception of the correlation line on the TN axis (0.06) was interpreted as the percentage of inorganic nitrogen, suggesting that a contribution of mineral nitrogen present in fine-grained sediments accounted for ca. 0.06 wt.%. We thus subtracted 0.06 wt.% from the TN content and used this for the calculation of the C:N ratio. The  $\delta^{13}$ C values of TC are also considered as  $\delta^{13}$ C of TOC  $(\delta^{13}C_{org})$  in this study and reported in the standard delta notation relative to Vienna Pee Dee Belemnite (VPDB) standard. The analytical precision (as the standard deviation for repeated measurements of the internal standards) was  $\pm 0.06\%$  for  $\delta^{13}C_{org}$ .

#### 3.3 Lignin phenol analysis

The lignin phenols were extracted from approximately 500 mg lake sediment and soil samples and from 50 mg of macrophytes samples. The samples were freezedried and the extraction method applied was the alkaline CuO oxidation (Hedges and Ertel, 1982; Goni and Hedges, 1992) at the Universidade Federal Fluminense laboratory (Brazil). In brief, sediments or macrophytes were transferred to stainless steel reaction vials and digested with 300 mg CuO in 2N NaOH under N<sub>2</sub> in an oxygen-free atmosphere at 150°C for 150 min. The samples were acidified to pH 1-3 and subsequently 6 ml of ethyl acetate was added. After centrifuging at 2,500 rpm for 5 min, the supernatant was collected, dried over sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>), evaporated under a stream of N<sub>2</sub>, reconstituted in pyridine, and converted to trimethylsilyl derivatives using bis-(trimethylsilyl) trifluoroacetamide (BSTFA) at 60°C for 20 min. Oxidation products were analyzed using an HP Agilent 6890N Series gas chromatography (e.g. Zocatelli et al., 2013).

The recovery factor was calculated using the internal standard ethyl vanillin added after the CuO oxidation and prior to analysis (values above 60% were considered). The response factor was performed using a mixture of commercial standards in four different concentrations, which were periodically injected for calibration. To confirm the identification of each lignin phenol, eight selected samples were analyzed with an Agilent 7890A gas chromatograph (GC-FID) coupled to an Agilent 5975C VL MSD mass spectrometer using a selective ion monitoring (SIM) at NIOZ (The Netherlands).

Phenol concentrations were reported as the carbon-normalized sum of eight lignin-derived reaction products ( $\lambda 8 \text{ mg } g_{oc}^{-1}$ ), including vanillyl (V-series) phenols (vanillin, acetovanillone, and vanillic acid), syringyl (S-series) phenols (syringealdehyde, acetosyringone, and syringic acid), and cinnamyl (C-series) phenols (p-coumaric and ferulic acid). Ratios S:V and C:V were calculated to identify angiosperm tissue sources. The ratio of acidic to aldehyde vanillyl phenols ((Ad:Al)v) was used as an indicator of the lignin degradation state since acidic phenols are produced from aldehyde functional groups during the lignin degradation (Hedges and Ertel, 1982).

#### 3.4 GDGT analysis

All samples for the lipid analysis were processed at NIOZ (The Netherlands). The freeze-dried samples were extracted with a modified Bligh and Dyer technique (Bligh and Dyer, 1959; Pitcher et al., 2009). In brief, the samples were extracted three times with a mixture of methanol (MeOH):dichloromethane (DCM):phosphate buffer (8.7 g of K<sub>2</sub>HPO<sub>4</sub> in 1 L bidistilled water) 10:5:4 (v:v:v) in an ultrasonic bath (10 min.). Extracts and residues were separated each time by centrifugation at 2,500 rpm for 2 min. DCM and phosphate buffer were added to the extracts to give a new volume ratio 1:1:0.9 (v:v:v). This mixture was centrifuged at 2,500 rpm for 2 min. to obtain a good phase separation. The DCM phase was then collected in a round bottom flask. The MeOH-phosphate phase was washed twice with DCM and then discarded. The collected DCM fractions were reduced under rotary vacuum.

The Bligh and Dyer extracts were fractioned into core lipids and intact polar lipids (IPLs). The separation was carried out on activated silica with *n*-hexane:ethylacetate 1:1 (v:v) for core lipids and MeOH for IPLs (Pitcher et al., 2009). To each fraction, 0.1 µg C<sub>46</sub> GDGT internal standard was added (Huguet et al., 2006). Two third of the IPL fraction was hydrolyzed to cleave off polar head groups. The hydrolysis was carried out by refluxing (3 h) in 2 N HCl:MeOH 1:1 (v:v). The solution was adjusted to pH 5 with 2 N KOH-MeOH. This mixture was washed three times with DCM. The DCM fractions were collected, reduced by rotary evaporation, and dried over a Na<sub>2</sub>SO<sub>4</sub> column. Core lipids fractions were separated into polar (DCM:MeOH 1:1, v:v) and apolar (DCM) fraction over an activated Al<sub>2</sub>O<sub>3</sub> column.

The core lipids and IPL GDGTs were analyzed using high performance liquid chromatograph-atmospheric pressure positive ion chemical ionization-mass spectrometry (HPLC-APCI-MS, an Agilent 1100 series LC/MSD SL, Alltech Prevail Cyano column (150 × 2.1 mm × 3  $\mu$ m)) in a selected ion monitoring (SIM) mode according to Schouten et al. (2007). Quantification of the GDGTs was achieved by integrating the peak areas and using a C<sub>46</sub> GDGT internal standard according to Huguet et al. (2006).

#### 3.5 Long-Chain *n*-Alkanes carbon isotopes

Two sediment samples collected in the LW season, one from lake Janauaca and another from lake Curuai, were used to compare the differences in the  $\delta^{13}$ C values of plant-wax derived long-chain *n*-alkanes in the upstream and in the downstream lakes. The extraction of *n*-alkanes was performed with an Accelerated Solvent Extraction method (ASE). The extracts were fractionated in apolar and polar fractions using an activated aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) column with hexane and MeOH:DCM (1:1, v:v), respectively, as the eluents. n-alkanes in the apolar fractions were identified by a Thermo Finnigan Trace DSQ gas chromatograph-mass spectrometry (GC-MS) and quantified with an HP 6890 GC system. To quantify the concentration of the *n*-alkanes, an internal standard was added to the apolar extracts. To further clean up the apolar fraction, the extracts were passed over a silver nitrate (AgNO<sub>3</sub>) column using hexane as the eluent. The  $\delta^{13}$ C values of higher *n*-alkanes were determined using an isotope-ratiomonitoring mass spectrometer (IRM-GC-MS) Thermo Delta V Advantage and the results were obtained using the software Isodat 3.0. Isotope values were measured against calibrated external reference gas and performance of the instrument was monitored by daily injects of a mixture of a  $C_{20}$  and a  $C_{24}$  perdeuterated *n*-alkane with known isotopic compositions. The  $\delta^{13}$ C values for the *n*-alkanes are reported in the standard delta notation against the Vienna Pee Dee Belemnite (VPDB) standard. All samples were run four times with an average standard deviation of 0.3 ‰ for the C<sub>25</sub> nalkane, 0.3 ‰ for the C<sub>29</sub> *n*-alkane, and 0.2 ‰ for the C<sub>31</sub> *n*-alkane (Sinninghe-Damsté et al., 2011).

#### 3.6 Statistical analysis

To evaluate the differences in mean values between different groups, the nonparametric Mann-Whitney U-test was used, which does not meet the normality assumption of the one-way analysis variance (ANOVA). Groups that showed significant differences (p<0.05) were assigned with different letters. The statistical test was performed with the software package SIGMAPLOT 13.0.

#### 4 Results

#### 4.1 Bulk parameters

The TOC content was the lowest in the downstream lake Curuai (2.0±0.6 wt.%) and the highest in lake Cabaliana (3.3±0.8 wt.%) (Fig. 3a, Table 3). No significant seasonal variation was observed (p=0.145) (Fig. 4A). The C:N ratio did not reveal significant spatial (p=0.104) and seasonal (p=0.418) variations (Figs. 3B and 4B). The  $\delta^{13}C_{org}$  values were significantly less negative in the downstream lakes (p<0.001) (Fig. 3C). In lake Curuai the mean value was  $-27\pm1\%$  and in lake Cabaliana  $-33\pm2\%$ . No significant (p=0.968) seasonal variation was observed for the  $\delta^{13}C_{org}$  values (Fig. 4C).

The  $\delta^{13}C_{org}$  values in "terra firme" soils and wetland soils varied between -29 and -19‰ (n=7). These samples were collected in the Amazon River margin between Canaçari and Curuai. The C:N ratio values varied between 6 and 16 (n=7) (Table 3). The C<sub>4</sub> macrophytes samples (*Paspalum repens*) showed values of  $\delta^{13}C_{org}$  between -14 and -13‰ and values of the C:N ratio between 15 and 27. The C<sub>3</sub> macrophytes (*Eleocharis sp.* and *Pistia stratiotes*) had  $\delta^{13}C_{org}$  values between -30‰ and -33‰ and C:N ratios between 15 and 24 (Table 2).

#### 4.2 Lignin phenols

No significant changes (p=0.392) were observed along the upstreamdownstream transect for the mean values of  $\lambda$ 8 (i.e. a proxy for the amount of lignin normalized to OC); The mean value of  $\lambda$ 8, a proxy for the amount of lignin normalized to OC, for the SOC was 44±29 mg g<sub>oc</sub><sup>-1</sup>. No significant changes (p=0.392) were observed along the upstream-downstream transect for the mean values of  $\lambda$ 8); However,  $\lambda$ 8 values revealed significant seasonal changes (p=0.001). The higher values were observed in the RW (56±30 mg g<sub>oc</sub><sup>-1</sup>) and in the FW seasons (62±34 mg g<sub>oc</sub><sup>-1</sup>) compared to the HW (23±9 mg g<sub>oc</sub><sup>-1</sup>) and LW (29±12 mg g<sub>oc</sub><sup>-1</sup>) seasons (Fig. 4G). The C:V ratio showed no significant seasonal (p=0.609) and spatial variation (p=0.214), and the mean value for all sediments was 0.7±0.4 (Figs. 3D and 4D). The values of the S:V ratio also does not show significant spatial (p=0.568) or seasonal (p=0.08) differences. The mean values for the lakes were approximately 1.1±0.1 and the mean seasonal values varied between 0.9±0.1 and 1.1±0.2 (Fig. 4E and Table 3). The mean value of (Ad:Al)v ratio for the different lakes does not show spatial variation (p=0.137) (Fig. 3F), however, it was higher in the LW (1.5±0.4) and HW (1.7±0.5) seasons (p<0.001) (Fig. 4F).

For the C<sub>3</sub> macrophytes,  $\lambda 8$  values varied between 26–67 mg g<sub>oc</sub>.<sup>1</sup> and between 48–94 mg g<sub>oc</sub>.<sup>1</sup> for the C<sub>4</sub> macrophytes. The S:V ratio varied between 0.6 and 0.9 for C<sub>3</sub> macrophytes and between 0.4 and 0.7 for the C<sub>4</sub> macrophytes. The range of C:V ratio was 0.4 to 3.7 for the C<sub>3</sub> macrophytes and 1.7 to 4.0 for the C<sub>4</sub> macrophytes. The (Ad:Al)v ratio varied between 0.2 and 0.8 for all macrophyte samples (Table 3). For the "Terra Firme" soil and wetland soil samples, the  $\lambda 8$  values varied between 9 and 88 mg g<sup>-1</sup>. The S:V ratio varied between 0.5 and 1.1, the C:V ratio varied between 0.2 and 0.5, and the (Ad:Al)v ratio varied between 0.6 and 1.5.

#### 4.3 BrGDGTs and crenarchaeol

Along the upstream-downstream transect, no significant changes (p=0.371) were observed for the mean values of brGDGTs concentrations (Fig. 3H). The lowest value was found in Curuai ( $31\pm14 \ \mu g \ g_{oc}^{-1}$ ) and the highest one in Canaçari ( $44\pm22 \ \mu g \ g_{oc}^{-1}$ ). The average concentration of crenarchaeol was higher in Canaçari ( $12\pm6 \ \mu g \ g_{oc}^{-1}$ ) when compared to Janauaca ( $4\pm3 \ \mu g \ g_{oc}^{-1}$ ). However, no significant difference (p=0.127) was observed between the upstream (Cabaliana and Janauaca) lakes and the downstream lake (Curuai) (Fig. 3H and 3I). On the other hand, brGDGTs concentrations showed significant seasonal changes (p=0.025). The highest mean value for brGDGTs concentrations was found in the FW season ( $45\pm23 \ \mu g \ g_{oc}^{-1}$ ), and the lowest mean concentration was found in the HW season ( $24\pm16 \ \mu g \ g_{oc}^{-1}$ ). The RW and LW seasons showed intermediate mean concentrations ( $35\pm12 \ \mu g \ g_{oc}^{-1}$  and  $38\pm16 \ \mu g \ g_{oc}^{-1}$ , respectively) and no significant difference (p=0.335) was observed if compared to the FW and HW seasons (Fig. 4H).

The concentrations of crenarchaeol did not reveal significant changes (p=0.096) over the hydrological seasons (Fig. 4I). The mean values varied between  $4\pm4 \ \mu g \ g_{oc}^{-1}$  and  $10\pm6 \ \mu g \ g_{oc}^{-1}$  in the HW and LW seasons, respectively. The percentage of IPL brGDGTs and IPL crenarchaeol was significantly higher (p=0.002 and p<0.001, respectively) in the LW season (19 $\pm7$  and 23 $\pm9\%$ , respectively). In the other three seasons, it showed values around 10 $\pm2\%$  of IPL brGDGTs and IPL crenarchaeol with no significant variability (Table 3).

### 4.4 Long-chain *n*-alkanes

The results of n-alkane analyses are summarized in Table 4. The carbon preference indices (CPI), calculated according to Bray and Evans (1961), were high, confirming a plant wax origin of the higher n-alkanes. A somewhat lower CPI (3.5) was found in the downstream lake compared to those in the upstream lake (5.5). A

significant increase in the  $\delta^{13}$ C values of the long-chain n-alkanes (C<sub>27</sub>, C<sub>29</sub>, C<sub>31</sub>) was observed downstream. In the upstream lake, the mean  $\delta^{13}$ C for the long-chain n-alkanes was  $-34.1\pm0.5\%$  and in the downstream lake the mean value was  $-31.6\pm0.6\%$  (Table 4). This represents a difference of 2.5‰ from upstream to downstream.

#### **5 Discussion**

#### 5.1 Sources of sedimentary organic matter in the floodplain lakes

To determine the origin of the SOM in the floodplain lakes, we considered five potentially significant sources in the Central Amazon Basin (Hedges et al., 1986; Moreira-Turcq et al. 2013; Mortillaro et al., 2011): (1) the terrestrial Andean claybounded and refractory SPOM, which may be transferred to the floodplain lakes via the Solimões-Amazon and Madeira rivers (Hess et al., 2003), (2) "terra firme" soils and littesr of the Amazonian lowland forests (non-floodable forests), which will be transferred to the floodplain lakes via local streams, (3) the wetland soils (flooded forests) and litters (leaves, grasses, woods etc.), transferred to the floodplain lakes during the receding waters (FW season) or in the rainy season (RW season) (Schöngart et al., 2010), (4) the wetland aquatic and semi-aquatic macrophyte vegetation of the floodplain lakes (Junk, 1997; Moreira-Turcq et al., 2004; Mortillaro et al., 2011), and (5) phytoplankton from the river or produced in the lake itself (Moreira-Turcq et al., 2004; Mortillaro et al., 2011). The biomarkers analyzed, lignin phenols and GDGTs, enabled us to identify most of these sources of OM, except for planktonic sources. However, in this case, some information can be obtained using bulk parameters, i.e. the  $\delta^{13}C_{org}$  and C:N ratio. Our results were compared with data reported previously (Hedges et al., 1986; Martinelli et al., 1994; Meyers, 1994; Martinelli et al., 2003; Aufdenkampe et al., 2007; Zell et al., 2013b) and with specific OM sources sampled and analyzed in this work, such as macrophytes, wetland soil and "terra firme" soil (Table 3), in order to identify the main sources of SOM in the floodplain lakes.

The average values of the various parameters of the river SPOM (Ertel et al., 1986; Hedges et al., 1986), wetland soils, "terra firme" soils and the potential biological OM sources (phytoplankton, macrophytes, grass, leaves and wood) are compared with those of the SOM of the floodplain lakes in Fig. 5 and Table 5. Data for the riverine SPOM is subdivided into fine particulate organic matter (FPOM) and coarse particulate organic matter (CPOM). For the interpretation of these data, it is important to note that the amount of CPOM in the Amazon River has been reported to be approximately eight times lower than that of the FPOM (Richey et al., 1990). The averages of important lignin parameters ( $\lambda$ 8, S:V ratio) but also the C:N ratio of the wood samples are significantly different (p<0.001) from those for the sediments, which clearly indicates only a minor contribution of woody material to the SOM. Furthermore, the  $\lambda 8$  of riverine FPOM is substantially lower than that of the SOM of the floodplain lakes, indicating that riverine SPOM is not an important source of lignin for the SOM of the floodplain lakes either. In terms of lignin parameters, the SOM is distinguished by two clear characteristics. Firstly, the (Ad:Al)v ratio is high with an average value of 1.25 (Fig. 5). Such a high value is only noted in the wetland and "terra firme" soils. However, this ratio is affected by the oxidation state of the lignin and thus, cannot be used as a source characteristic of the lignin. Secondly, the SOM is characterized by a substantially elevated C:V ratio (Fig. 6; cf. Hedges et al., 1982). Since all of the potential lignin sources, except macrophytes, have a much lower value, this indicates that macrophyte lignin and, thus accordingly, macrophyte OM (since average  $\lambda$ 8 values of macrophyte OM and the SOM do not substantially differ) largely contribute to the SOM. Since the S:V ratio of macrophyte OM is relatively lower than that of the lignin component of the SOM (Fig. 5), some contributions of lignin derived from other fresh plant OM (i.e. grasses/leaves) or wetland soils might explain the elevated S:V ratio of the SOM.

Further information with respect to sources of SOM can be obtained from the GDGT concentrations. The concentrations of both brGDGTs and crenarchaeol are higher in the riverine SPOM than in the SOM, pointing to a contribution of riverine SPOM to the SOM, in contrast to what was shown by the lignin phenols. However, the concentrations of brGDGTs in the wetland soils and river SPOM are statistically indistinguishable and, thus, it is not possible to use the brGDGTs as a specific OM source indicator. This is in line with the idea that brGDGTs can be produced in soils (e.g., Weijers et al., 2006), rivers (e.g., Zell et al., 2013a; De Jonge et al., 2015) and lake waters (e.g., Tierney et al., 2010; Buckles et al., 2014). On the other hand, riverine SPOM is the most likely OM source for the substantially increased concentration of crenarchaeol in the SOM of the floodplain lakes if compared to other sources (Fig. 5). Crenarchaeol is indeed produced in the Amazon River by nitrifying archaea that consume ammonium produced from degrading OM (Zell et al., 2013b). However, it is known that crenarchaeol is also produced in lakes (Blaga et al., 2011; Tierney and Russell, 2009), indicating that it may also be produced in the floodplain lakes. Crenarchaeol is, therefore, considered as an indicator of aquatic OM in this system. The enhanced concentrations of crenarchaeol in SOM thus indicate an increased contribution from riverine and/or lacustrine SPOM.

In terms of bulk parameters, the C:N ratio in the SOM shows intermediate values between the riverine SPOM and the various OM sources but, with no distinct average values between them. Moreover, the average values of  $\delta^{13}C_{org}$  are statistically equal for sediments and most sources of OM (except for the wetland soils), and the

20

TOC do not show any significant difference between the soils samples (p=1.241), riverine SPOM (p=1.044) and lake sediments. Thus, it is not possible to discriminate any specific source of SOM based on the average values of the bulk parameters.

We have argued that the C:V ratio and the crenarchaeol concentration are the only two parameters that clearly point out one specific source of SOM (i.e., macrophytes and aquatic OM from rivers or floodplain lakes, respectively). Consequently, these parameters can be applied to a two end-member model to estimate the fractions of each of these two sources in the SOM. According to this approach (Martinelli et al., 2003), the average C:V values of macrophytes and the average values of other OM sources (wetland and non-flooded soils and SPOM) can be used to estimate the contribution of macrophyte OM to the SOM (Eq. 1). Similarly, the concentration of crenarchaeol in the riverine SPOM and its concentration in soil samples can be used to estimate the contribution of aquatic OM to the SOM (Eq. 2).

$$F_{\text{macrophytes}} = \frac{\text{C:V}_{\text{SOM}} - \text{C:V}_{(\text{SPOM+forest})}}{\text{C:V}_{\text{macrophyte}} - \text{C:V}_{(\text{SPOM+forest})}} \times 100$$
(Eq. 1)

$$F_{\text{SPOM}} = \frac{\text{Cren}_{\text{SOM}} - \text{Cren}_{|\text{forest+macrophyte}|}}{\text{Cren}_{\text{SPOM}} - \text{Cren}_{|\text{forest+macrophyte}|}} \times 100$$
(Eq. 2)

$$F_{\text{macrophyte}} + F_{\text{SPOM}} + F_{\text{forest}} = \text{SOM} (100\%)$$
 (Eq. 3)

In Eqs. (1) and (2), the  $F_{macrophytes}$  and  $F_{SPOM}$  represent the estimated fractional abundance of macrophytes and aquatic OM in SOM, respectively. C:V<sub>SOM</sub> and Cren<sub>SOM</sub> are the average values of each parameter found in the sediment samples, C:V<sub>macrophytes</sub> and Cren<sub>SPOM</sub> are the values of the predominant source of the respective parameter and C:V<sub>(SPOM+forest)</sub> and Cren<sub>(forest+macrophyte)</sub> are the values of the other possible OM sources. As discussed above, the high values of (Ad:Al)v indicate that lignin components of the SOM is partially degraded, which may affect the values of the C:V ratio. There are also

numerous complications with the application of crenarchaeol as an indicator of aquatic matter in this ecosystem. Therefore, the presented mixing model should be considered as estimations. The results of Eq. 1-3 indicate that 25–35% of the SOM is derived from macrophytes and 20–30% from aquatic OM sources (riverine and lacustrine SPOM) . Consequently, the remaining 35–55% of the SOM might be derived from the wetlands and non-flooded forests (Eq. 3). The periodic floods link the floodplain lakes and the wetland vegetation and soil. Thus, the seasonal and spatial contrasts in the SOM should be investigated in order to better understand the connectivity between these compartments.

#### 5.2 Spatial differences in the composition of sedimentary organic matter

Along the longitudinal transect, from upstream to downstream, most bulk geochemical parameters (i.e. TOC content and  $\delta^{13}C_{org}$ ) show significant differences between the upstream and downstream lakes (Figs. 3A, 3C), while most of the measured biomarker parameters ( $\lambda$ 8, S:V, (Ad:Al)v and brGDGTs) do not show such a pattern (Figs. 4E, 4F, 4G, and 4H). On the other hand, the biomarker parameters show, in some cases, a clear seasonal contrast, which is not observed for the bulk parameters. Consequently, the bulk parameters apparently mix and homogenize the long time scale (year), while the biomarkers are more sensible to changes in short time scale (months) at the sediment surface. This observation is in agreement with previous studies about earlier diagenesis of organic molecules (Harvey, 2006). It is important to note that the results must be interpreted taking into consideration the high sedimentation rates in the floodplain lakes, typically 1-2 cm y<sup>-1</sup> (Moreira-Turcq et al., 2004), and the fact that resuspension is induced by storms during the LW and RW seasons or by currents during the receding waters (FW). These events may have a substantial effect on the material

comprising the first 2 cm of sediments of floodplain lakes, which are mixed with newly arrived SOM from the water column, and are re-oxygenated favoring the degradation.

The percentage of TOC in the sediment samples shows a decrease from 3.3 (wt. %) upstream (Cabaliana) to 2.1 (wt.%) downstream (Curuai) (Fig. 3A). Furthermore, over the transect of lakes the average  $\delta^{13}C_{org}$  values increase by ca. 5‰ (Fig. 3C). However, the average C:N ratio does not show any significant changes over the transect (Fig. 3B). These results are in good agreement with previous studies in the central Amazon Basin (Victoria et al., 1992; Martinelli et al., 2003). The increasing trend in  $\delta^{13}C_{org}$  from upstream to downstream lakes may be caused by an increased contribution of C<sub>4</sub> macrophytes to the SOM, whose abundance increases in open water lakes and floodplains. Alternatively, since the  $\delta^{13}C_{org}$  values in the downstream lakes come closer to the  $\delta^{13}C_{org}$  of the Solimões-Amazon SPOM (~-23 to -30‰), an increased input of riverine organic matter may also explain this. To disentangle whether this trend in the  $\delta^{13}C_{org}$  values is caused by an increased contribution of C<sub>4</sub> plants or of riverine SPOM, the isotopic composition ( $\delta^{13}$ C) of long-chain *n*-alkanes, markers for higher plants, in sediments from the upstream lake Janauaca and the downstream lake Curuai, both collected during the LW season, was measured. The results (Table 4) show that the longchain n-alkanes  $\delta^{13}$ C signature are more like those of C<sub>3</sub> higher plants (Castañeda et al., 2009) for both lakes although for the Curuai the values are slightly less negative. If one considers the values of  $\delta^{13}$ C in the *n*-alkane C<sub>29</sub> in the leaf waxes of C<sub>3</sub> and C<sub>4</sub> plants, one can calculate the contribution of C<sub>4</sub> plants sedimentary *n*-alkanes according to the following equation:

$$C_{4} \text{ plants (\%)} = \frac{\delta^{13}C_{\text{org}} C_{29} (C_{3} \text{ plants}) - \delta^{13}C_{\text{org}} C_{29} (\text{sediment})}{\delta^{13}C_{\text{org}} C_{29} (C_{3} \text{ plants}) - \delta^{13}C_{\text{org}} C_{29} (C_{4} \text{ plants})} \times 100$$
 (Eq. 4)

where the end member value for  $\delta^{13}C_{org} C_{29}$  (C<sub>3</sub> plants) is -34.7‰ and for  $\delta^{13}C_{org} C_{29}$  (C<sub>4</sub> plants) is -21.7‰ (Castañeda et al., 2009). The measured values for  $\delta^{13}C_{org}$  of the C<sub>29</sub> *n*-

alkane in the sediments of Janauaca and Curuai are listed in Table 4. Accordingly, the fraction of C<sub>4</sub> plants in the SOM in the upstream lake is only 3%, but for the downstream lake 22%. The difference in  $\delta^{13}C_{org}$  for C<sub>4</sub> and C<sub>3</sub> higher plants is ca. 20‰. A switch from almost 100% C<sub>3</sub> macrophytes to a 78% contribution would result in a change in the isotopic composition of the macrophyte "pool" of the SOM of 4-4.5‰. Since this pool is estimated to represent 20-30% of the SOM, this cannot fully explain the observed 5‰ shift (Fig. 3C). However, it should be considered that this increasing contribution of C<sub>4</sub> higher plants in the downstream lake may not solely be the consequence of the change in the floodplain soil, mainly covered by shrubs and grass vegetation, may contribute to the observed shift in  $\delta^{13}C_{org}$  of SOM.

# 5.3 Seasonal changes in the composition of sedimentary organic matter

The two centimeters of surface sediment we have characterized in this study potentially integrate more than one year of sedimentation in such floodplain environment (Moreira-Turcq et al. 2004). However, because of the occurrence of pulsated inputs as well as resuspension, mixing and degradation processes in these superficial sediments (Moreira-Turcq et al. 2013), changes in the composition of superficial sediment apparently occurred at the seasonal scale (Figure 4 and Table 3). Indeed, The  $\lambda$ 8 values showed significantly higher values in the RW and FW seasons than in the LW and HW seasons in all lakes (Figs. 4E, 4G, and 6A). The mean concentrations of brGDGTs also showed higher values in the FW season than in the HW season (Figs. 4H and 6B). The co-occurrence of these two types of molecules indicates that litter, traced by lignin phenols, and superficial soils, traced by brGDGTs, are preferentially deposited in the floodplain lakes during rising and receding waters. In addition, the seasonal mean values of (Ad:Al)v showed remarkably lower values in the

RW and FW seasons (Fig. 4F), an inverse pattern if compared to the  $\lambda$ 8 and brGDGTs. This suggests that less degraded lignin phenols were present in the surface sediments in the RW and FW seasons. Thus, in this case, the increase in the concentrations of the organic compounds was not a consequence of the re-suspension of the sediments, but due to a sudden arrival of fresher OM. In the HW and LW seasons, more degraded lignin phenols (i.e. higher values of (Ad:Al)v) were present in the sediments concomitant with lower amounts of  $\lambda 8$ . A possible process which is responsible for the  $\lambda$ 8 and brGDGTs transfer to the lakes sediments is the connection of the Amazon River main stem with the local catchment areas such as wetlands and non-flooded forests during the RW and FW seasons. The lignin concentration could also increase as a consequence of the macrophyte communities while the brGDGTs could increase due to the in situ production in the floodplain lakes. However, the concentrations of crenarchaeol and IPL brGDGTs as well as C:V ratio do not reveal significant seasonal changes (Table 3 and Fig. 4). Based on these observations, we interpret that these changes in the lignin phenols in the RW and FW seasons and the brGDGTs in the FW season were not derived from the lake in situ production but from soil and leaf runoffs.

Previous studies postulated that Andean and lowland soils are mainly transferred to the lakes via the Amazon River main stem, in particular, during the RW and HW seasons and that they would be the main source of SOM in the floodplain lakes (e.g., Victoria et al., 1992; Moreira-Turcq et al., 2004; Mortillaro et al., 2011). However, according to our results, the lignin phenols increased their concentration in the RW and the FW seasons. The hydrodynamics of floodplain lakes and their connections to the local drainage flooded forests and the main stem (Bourgoin et al., 2007) and the analusis of biomarkers applied in this study, suggest that in the RW and FW seasons, these organic molecules are mainly derived from the drainage of local wetlands and lowland "terra firme" soils. This is more evident for the upstream lakes surrounded by larger flooded forests than for the downstream lakes surrounded mainly by grass vegetation and shrubs. Even if in lake Curuai, phytoplankton primary production and the riverine SPOM are potentially important sources of SOM (Moreira-Turcq et al., 2004; Zocatelli et al., 2013), thia material is not predominant in the sediments, compared to the material coming from the interface between the lake and the wetland is determinant for the sedimentation of the organic compounds.

# 6 Conclusion

Our results suggest that the vegetation coverage of the wetlands (flooded forests) and "terra firme" (non-floodable forests) in the local catchment area of each lake investigated is the most important source of SOM in floodplain lakes of the central Amazon basin. The macrophyte community is also an important source of SOM whereas aquatic OM (i.e., riverine and lacustrine SPOM) contributes to a somewhat lesser extent. In upstream lakes, higher TOC contents in the surface sediments are observed, if compared to the downstream large open lakes. The differences observed in the vegetation coverage of the wetlands, affect the quality of SOM in the floodplain lakes. This pattern could only be observed in a longitudinal transect approach, with the application of molecular isotope technique apart from multiple biomarkers analysis. The sedimentation of OM in the floodplain lakes are strongly linked to the periodic floods. The rain season (RW season), with substantially increased soil runoff, and the receding of waters (FW season), when OM is transported from the flooded soils to the floodplain lakes, are the most important hydrological factors for the sedimentation of OM in the wetlands of the central Amazon basin. Hence, together with wetland and non-flooded vegetation, the hydrodynamics of the floodplain seems to be the most important controlling factor on the composition of SOM in the floodplain lakes of the central Amazon basin.

#### Acknowledgments

This work was conducted in collaboration with the carbon cycle in the Amazon river (CARBAMA) project, funded by the French national research agency (grant n° 08-BLANC-0221) in the framework of the HYBAM Observatory (INSU/IRD). It was conducted by an international cooperation agreement between the National Council for Scientific and Technological Development-Brazil (CNPq) and the Institute for Research and Development-France (IRD) and Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES). The research leading to these results has also received funding from the European Research Council (ERC) under the European Union's Seventh Framework Program (*FP7/2007-2013*) / ERC grant agreement n° [*226600*]. We would like to thank the Companhia de Pesquisa dos Recursos Minerais (CPRM) technical groups for their help during the sampling expeditions. We also would like to thank the INCT-TMCOcean Project (CNPq Proc. 573601/2008-9) for analytical support.

#### References

Abril, G., Martinez, J.-M., Artigas, L.F., Moreira-Turcq, P., Benedetti, M.F., Vidal, L., Meziane, T., Kim, J.-H., Bernardes, M.C., Savoye, N., Deborde, J., Souza, E.L., Alberic, P., Landim de Souza, M.F. and Roland, F. Amazon River carbon dioxide outgassing fueled by wetlands. Nature 505, 395–398, 2014.

Aufdenkampe, A.K., Mayorga, E., Hedges, J.I., Llerena, C., Quay, P.D., Gudeman, J., Krusche, A.V. and Richey, J.E. Organic matter in the Peruvian headwaters of the Amazon: Compositional evolution from the Andes to the lowland Amazon mainstem. Organic Geochemistry 38, 337–364, 2007.

Battin, T.J., Luyssaert, S., Kaplan, L.A., Aufdenkampe, A.K., Richter, A. and Tranvik, L.J. The boundless carbon cycle. Nature Geoscience 2, 598–600, 2009.

Belicka, L.L. and Harvey, H.R. The sequestration of terrestrial organic carbon in Arctic Ocean sediments: A comparison of methods and implications for regional carbon budgets. Geochimica et Cosmochimica Acta 73, 6231–6248, 2009.

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. and Hedges, J.I. Riverine organic matter composition as a function of land use changes, Southwest Amazon. Ecological Applications 14, 263–279, 2004.

Blaga, C.I., Reichart, G.J., Vissers, E.W., Lotter, A.F., Anselmetti, F.S., Sinninghe Damsté, J.S., Seasonal changes in glycerol dialkyl glycerol tetraether concentrations and fluxes in a perialpine lake: implications for the use of the TEX<sub>86</sub> and BIT proxies. Geochimica et Cosmochimica Acta 75, 6416–6428, 2011.

Bligh, E.G. and Dyer, W.J. A Rapid Method of Total Lipid Extraction and Purification. Canadian Journal of Biochemistry and Physiology 37, 911–917, 1959.

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

Bourgoin, L.M., Bonnet, M.P., Martinez, J.M., Kosuth, P., Cochonneau, G., Moreira-Turcq, P., Guyot, J.L., Vauchel, P., Filizola, N. and Seyler, P. Temporal dynamics of water and sediment exchanges between the Curuai floodplain and the Amazon River, Brazil. Journal of Hydrology 335, 140–156, 2007.

Bray, E.E. and Evans, E.D. Distribution of n-paraffins as a clue to recognition of source beds. Geochimica et Cosmochimica Acta 22, 2–15, 1961.

Buckles, L. K., Weijers, J. W. H., Tran, X.-M., Waldron, S., and Sinninghe Damsté, J. S. Provenance of tetraether membrane lipids in a large temperate lake (Loch Lomond, UK): implications for glycerol dialkyl glycerol tetraether (GDGT)-based palaeothermometry. Biogeosciences 11, 5539–5563, 2014.

Callede, J., Kosuth, P., Guyot, J.L. and Guimaraes, V.S. Discharge determination by acoustic doppler current profilers (ADCP): A moving bottom error correction method and its application on the River Amazon at Obidos. Hydrological Sciences Journal 45, 911–924, 2000.

Castañeda, I.S., Mulitza, S., Schefu, E., Santosa, R.A.L.d., Sinninghe Damsté, J.S. and Schouten, S. Wet phases in the Sahara/Sahel region and human migration patterns in North Africa. Proceedings of the National Academy of Sciences of the United States of America 106, 20159–20163, 2009.

Cole, J.J., Prairie, Y.T., Caraco, N.F., McDowell, W.H., Tranvik, L.J., Striegl, R.G., Duarte, C.M., Kortelainen, P., Downing, J.A., Middelburg, J.J. and Melack, J. Plumbing the Global Carbon Cycle: Integrating inland waters into the terrestrial carbon budget. Ecosystems 10, 171–184, 2007.

De Jonge, C., Stadnitskaia, A., Hopmans, E.C., Cherkashov, G., Fedotov, A., and Sinninghe Damsté, J.S. In situ produced branched glycerol dialkyl glycerol tetraethers in suspended perticulate matter from the Yennisei River, Eastern Siberia. Geochimica et Cosmochimica Acta 150, 476–491, 2015.

Devol, A.H., Zaret, T.M. and Forsberg, B.R. Sedimentary organic matter diagenesis and its relation to the carbon budget of tropical Amazon floodplain lakes. Verhandlungen Internationale Vereinigung für theoretische und angewandte Limnologie 22, 1299–1304, 1984.

Ertel, J.R., Hedges, J.I., Devol, A.H., Richey, J.E. and Ribeiro, M.d.N.G. Dissolved humic substances of the Amazon River system. Limnology and Oceanography 31, 139–154. 1986.

Goñi, M.A. and Hedges, J.I. Lignin Dimers - Structures, Distribution, and Potential Geochemical Applications. Geochimica et Cosmochimica Acta 56, 4025–4043, 1992.

Goulding, M., Barthem, R. and Ferreira, E.J. The Smithsonian Atlas of Amazon. Princeton Editorial Associates, Oklahoma City, 2003.

Harvey, H.R. Sources and Cycling of Organic Matter in the Marine Water Column, in: Volkman, J.K. (Ed.), Marine Organic Matter: Biomarkers, Isotopes and DNA. Springer, New York, 2006.

Hedges, J.I., Clark, W.A., Quay, P.D., Richey, J.E., Devol, A.H. and Santos, U.D. *Compositions and Fluxes of Particulate Organic Material in the Amazon River.* Limnology and Oceanography. Oceanogr. 31, 717–738, 1986.

Hedges, J.I., Cowie, G.L., Richey, J.E., Quay, P.D., Benner, R., Strom, M. and Forsberg, B.R. Origins and processing of organic matter in the Amazon River as indicated by carbohydrates and amino acids. Limnol. Oceanogr. 39, 743–761, 1994.

Hedges, J.I. and Ertel, J.R. Characterization of Lignin by Gas Capillary Chromatography of Cupric Oxide Oxidation Products. Analytical Chemistry 54, 174– 178, 1982.

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

Herfort, L., Schouten, S., Boon, J.P., Woltering, M., Baas, M., Weijers, J.W.H. and Damsté, J.S.S. Characterization of Transport and Deposition of Terrestrial Organic Matter in the Southern North Sea Using the BIT Index. Limnology and Oceanography 51, 2196–2205, 2006.

Hess, L.L., Melack, J.M., Novo, E.M.L.M., Barbosa, C.C.F. and Gastil, M. Dual-season mapping of wetland inundation and vegetation for the central Amazon basin. Remote Sensing of Environment 87, 404–428, 2003.

Hopmans, E.C., Schouten, S., Pancost, R.D., van der Meer, M.T.J., Sinninghe Damsté, J.S. Analysis of intact tetraether lipids in archaeal cell material and sediments by high performance liquid chromatography/atmospheric pressure chemical ionization mass spectrometry. Rapid Communications in Mass Spectrometry 14, 585–589, 2000.

Hopmans, E.C., Weijers, J.W.H., Schefu, E., Herfort, L., Sinninghe Damsté, J.S. and Schouten, S. A novel proxy for terrestrial organic matter in sediments based on branched and isoprenoid tetraether lipids. Earth and Planetary Science Letters 224, 107–116, 2004.

Huguet, C., Hopmans, E.C., Febo-Ayala, W., Thompson, D.H., Sinninghe Damsté, J.S. and Schouten, S. An improved method to determine the absolute abundance of glycerol dibiphytanyl glycerol tetraether lipids. Organic Geochemistry 37, 1036–1041, 2006.

Junk, W.J. The Central Amazon Floodplain: Ecology of a Pulsing System, Ecological Studies. Springer, Berlin, p. 529, 1997.

Junk, W.J., Piedade, M.T.F., Parolin, P., Wittmann, F. and Schöngart, J. Ecophysiology, Biodiversity and Sustainable Management of Central Amazonian Floodplain Forests: A Synthesis, in: Junk, W.J., Piedade, M.T.F., Wittmann, F., Schöngart, J., Parolin, P. (Eds.), Amazonian Floodplain Forests: Ecophysiology, Biodiversity and Sustainable Management. Springer, New York, pp. 511–540, 2010.

Kim, J.-H., Zell, C., Moreira-Turcq, P., Pe´rez, M.A.P., Abril, G.l., Mortillaro, J.-M., Weijers, J.W.H., Meziane, T. and Sinninghe Damsté , J.S. Tracing soil organic carbon in the lower Amazon River and its tributaries using GDGT distributions and bulk organic matter properties. Geochimica et Cosmochimica Acta 90, 163–180, 2012.

Kuzyk, Z.Z.A., Goñi, M.A., Stern, G.A. and Macdonald, R.W. Sources, pathways and sinks of particulate organic matter in Hudson Bay: Evidence from lignin distributions. Marine Chemistry 112, 215–229, 2008.

Martinelli, L.A., Victoria, R.L., Camargo, P.B.d., Piccolo, M.d.C., Mertes, L., Richey, J.E., Devol, A.H. and Forsberg, B.R. Inland variability of carbon–nitrogen concentrations and  $\delta^{13}$ C in Amazon floodplain (várzea) vegetation and sediment. Hydrological Processes 17, 1419–1430, 2003.

Martinelli, L.A., Victoria, R.L., Forsberg, B.R. and Richey, J.E. Isotopic Composition of Majors Carbon Reservoirs in the Amazon Floodplain. International journal of Ecology and environmental Sciences 20, 31–46, 1994.

Melack, J.M. and Forsberg, B.R. Biogeochemistry of Amazon Floodplain Lakes and Associated Wetlands, in: McClain, M.E., Victoria, F.L., Richey, J.E. (Eds.), The Biogeochemistry of the amazon Basin. Oxford University Press, New York, pp. 235–274, 2001.

Melack, J.M. and Hess, L.L. Remote Sensing of the Distribution and Extent of Wetlands in the Amazon Basin, in: Junk, W.J., Piedade, M.T.F., Wittmann, F., Schöngart, J., Parolin, P. (Eds.), Amazonian Floodplain Forests Ecophysiology, Biodiversity and Sustainable Management. Springer, New York, pp. 511–540, 2011.

Meyers, P.A. Preservation of elemental and isotopic source identification of sedimentary organic matter. Chemical Geology 114, 289–302, 1994.

Moreira, L.S., Moreira Turcq, P., Kim, J.-H., Turcq, B., Cordeiro, R.C., Caquineau, S., Mandengo-Yogo, M., Sinninghe Damsté, J.S. A Mineralogical and Organic Geochemical Overview of the Effects of Holocene Changes in Amazon River Flow on Three Floodplain Lakes. Palaeogeography Palaeoclimatology Palaeoecology 415, 152–164, 2014.

Moreira-Turcq, P., Jouanneau, J.M., Turcq, B., Seyler, P., Weber, O. and Guyot, J.L. Carbon sedimentation at Lago Grande de Curuai, a floodplain lake in the low Amazon Region: insights into sedimentation rates. Palaeogeography Palaeoclimatology Palaeoecology 214, 27–40, 2004.

Moreira-Turcq, P., Bonnet, M.-P., Amorim, M., Bernardes, M., Lagane, C., Maurice, L., Perez, M., and Seyler, P. Seasonal variability in concentration, composition, age, and fluxes of particulate organic carbon exchanged between the floodplain and Amazon River. Global Biogechemical Cycles 27, 119–130, 2013.

Mortillaro, J.M., Abril, G., Moreira-Turcq, P., Sobrinho, R.L., Perez, M. and Meziane, T. Fatty acid and stable isotope ( $\delta^{13}$ C,  $\delta^{15}$ N) signatures of particulate organic matter in the lower Amazon River: Seasonal contrasts and connectivity between floodplain lakes and the mainstem. Organic Geochemistry 42, 1159–1168, 2011.

Pitcher, A., Hopmans, E.C., Shouten, S. and Sinninghe Damsté, J.S. Separation of core and intact polar archaeal tetraether lipids using silica columms: Insights into living and fossil biomass contributions. Organic Geochemistry 40, 12–19, 2009.

Quay, P.D., Wilbur, D.O., Richey, J.E., Hedges, J.I. and Devol, A.H. Carbon Cycling in the Amazon River: Implications from the <sup>13</sup>C Compositions of Particlesand Solutes. Limnology and Oceanography 37, 857–871, 1992.

Raymond, P.A., Hartmann, J., Lauerwald, R., Sobek, S., McDonald, C., Hoover, M., Butman, D., Striegl, R., Mayorga, E., Humborg, C., Kortelainen, P., Dürr, H., Michel Meybeck, Ciais, P. and Guth, P. Global carbon dioxide emissions from inland waters. Nature 503, 355–359, 2013.

Richey, J.E., Hedges, J.I., Devol, A.H., Quay, P., Victoria, R., Martinelli, L. and Forsberg, B.R. Biogeochemistry of Carbon in the Amazon River. Liminology and Oceanography 32, 352–371, 1990.

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. Persistence of soil organic matter as an ecosystem property. Nature 478, 49-56, 2011.

Schöngart, J., Wittmann, F. and Worbes, M. Biomass and Net Primary Production of Central Amazonian Floodplain Forests, Amazonian Floodplain Forests Ecophysiology, Biodiversity and Sustainable Management. Springer, New York, pp. 347–388, 2010

Schouten, S., Huguet, C., Hopmans, E.C., Kienhuis, M.V.M., and Sinninghe Damsté, J.S. Analytical methodology for  $TEX_{86}$  paleothermometry by high-performance liquid chromatography/atmospheric pressure chemical ionization-mass spectrometry. Analytical Chemistry 79, 2940–2944, 2007.

Sinninghe Damsté, J.S., Shouten, S., Hopmans, E.C., van Duin, A.C.T., Geenevasen, J.A.J. Crenarchaeol: The characteristic core glycerol dibiphytanyl glycerol tetraether membrane lipid of cosmopolitan pelagic crenarchaeota. Journal of Lipid Research 43,1641–1651, 2002.

Sinninghe-Damsté J.S., Verschuren D., Ossebaar, J., Blokker, J., van Houten, R., van de Meer, M., Plessen, B., Schouten, S. A 25,000-year record of climate-induced changes in lowland vegetation of eastern equatorial Africa revealed by the stable carbon-isotopic composition of fossil plant leaf waxes, Earth and Planetary Science Letters, 302, 236-246, 2011.

Sioli, H. Das wasser in Amazonasgebeit. Forschung Fortchrift 26, 274–280, 1950.

Smith, R.W., Bianchi, T.S. and Savage, C. Comparison of lignin phenols and branched/isoprenoid tetraethers (BIT index) as indices of terrestrial organic matter in Doubtful Sound, Fiordland, New Zealand. Organic Geochemistry 41, 281–290, 2010.

Tierney, J.E., Russell, J.M., Eggermont, H., Hopmans, E.C., Verschuren, D. and Sinninghe Damsté, J.S. Environmental controls on branched tetraether lipid distributions in tropical East African lake sediments. Geochimica et Cosmochimica Acta 74, 4902–4918, 2010.

Tierney, J. E., and Russell, J. M. Distributions of branched GDGTs in a tropical lake system: Implications for lacustrine application of the MBT/CBT paleoproxy. Organic Geochemistry 40, 1032–1036, 2009.

Tranvik, L.J., Downing, J.A., Cotner, J.B., Loiselle, S.A., Striegl, R.G., Ballatore, T.J., Dillon, P., Finlay, K., Fortino, K., Knoll, L.B., Kortelainen, P.L., Kutser, T., Larsen, S., Laurion, I., Leech, D.M., McCallister, S.L., McKnight, D.M., Melack, J.M., Overholt, E., Porter, J.A., Prairie, Y., Renwick, W.H., Roland, F., Sherman, B.S., Schindler, D.W., Sobek, S., Tremblay, A., Vanni, M.J., Verschoor, A.M., von Wachenfeldt, E. and Weyhenmeyera, G.A. Lakes and reservoirs as regulators of carbon cycling and climate. Limnology and Oceanography 54, 2298–2314, 2009.

Victoria, R.L., Martinelli, L.A., Trivelin, P.C.O., Matsui, E., Forsberg, B.R., Richey, J.E. and Devol, A.H. The use of stable isotopes in studies of nutrient cycling: Carbon Isotope composition of Amazon varzea sediments. Biotropica 24, 240–249, 1992.

Ward, N.D., Keil, R.G., Medeiros, P.M., Brito, D.C., Cunha, A.C., Dittmar, T., Yager, P.L., Krusche, A.V. and Richey, J.E. Degradation of terrestrially derived macromolecules in the Amazon River. Nature Geosciences 6, 530–533, 2013.

Weijers, J.W.H., Schouten, S., Spaargaren, O.C. and Sinninghe Damsté, J.S. Occurrence and distribution of tetraether membrane lipids in soils: Implications for the use of the TEX<sub>86</sub> proxy and the BIT index. Organic Geochemistry 37, 1680–1693, 2006.

White, D.C., Davis, W.M., Nickels, J.S., King, J.D., Bobbie, R.J. Determination of the sedimentary microbial biomass by extractable lipid phosphate. Oecologia 40, 51–62, 1979.

Zell, C., Kim, J.-H., Abril, G., Sobrinho, R.L., Dorhout, D., Moreira-Turcq, P. and Sinninghe Damsté, J.S. Impact of seasonal hydrological variation on the distributions of tetraether lipids along the Amazon River in the central Amazon basin: implications for the MBT/CBT paleothermometer and the BIT index. Frontiers in microbiology 24, 1–14, 2013a.

Zell, C., Kim, J.-H., Moreira-Turcq, P., Abril, G., Hopmans, E.C., Bonnet, M.-P., Sobrinho, R.L. and Sinninghe Damsté, J.S. Disentangling the origins of branched tetraether lipids and crenarchaeol in the lower Amazon River: Implications for GDGT-based proxies. Limnology and Oceanography 58, 343–353, 2013b.

Zell, C., Kim, J.-H., Dorhout, D., Baas, M., and Sinninghe Damsté, J.S. Sources and distributions of branched tetraether lipids and crenarchaeol along the Portuguese continental margin: Implications for the BIT index, Continental Shelf Research, 96, 34–44, 2015.

Zocatelli, R., Cecanho, F., Amorim, M., Bernardes, M., Moreira-Turcq, P., Turcq, B., Sifeddine, A. and Cordeiro, R.C. 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 Amazonica 41, 195–204, 2011.

Zocatelli, R., Moreira-Turcq, P., Bernardes, M., Turcq, B., Cordeiro, R.C., Gago, S., Disnar, J.R. and Boussafir, M. Sedimentary evidence of soil organic matter input to the Curuai Amazonian floodplain. Organic Geochemistry 63, 40–47, 2013.

Table 1. Localization and summary of geomorphology, biogeography, and water physicochemical of the five floodplain lakes. Data of temperature, conductivity and pH represent the maximum and minimum values measured *in situ* for four hydrological seasons.

	Cabaliana	a Janauaca Mirituba Cana		Canaçari	Curuai
Latitude (S) Longitude (W)	3°18'46'' 60°40'15''	3°23'20" 60°16'26"	3°20'50'' 58°23'60''	2°58'60" 58°15'40"	2°09'44'' 55°27'53''
Approx. area (km <sup>2</sup> )	300	85	360	290	1050
Shape	Ellipsoid	Ravine dendritic	Round	Ellipsoid	Triangular
Wetland Vegetation Type	Forests	Forests	Forests/ Woodlands	Forests/ Woodlands	Woodlands/ Shrubs
Water	White/Black	White/Clear	White	White/Black	White
Conductivity (µS)	10-80	33-71	43-65	10-54	41-69
Temperature (°C)	28-34	29-33	28-34	29-34	30-36
pH	5.0-7.5	6.1-8.0	6.2-8.5	5.9-9.4	7.3-10.1
					······

Obs.: All várzeas receive white water from the Solimões-Amazon River in the flooding season.

Table 2	2. Value	es of bulk	parameters	and ligr	nin phenols	in wetlands,	, terrestrial (	("terra
firme"	) and ac	juatic sou	rces of sedi	imentary	OC.			

	TOC (wt.%)	C:N	δ <sup>13</sup> C <sub>org</sub> (‰ VPDB)	$\lambda 8$ (mg g <sub>OC</sub> <sup>-1</sup> )	C:V	S:V	(Ad:Al) v
Macrophytes							
Eleocharis sp. (root)	27.2	15	-32.3	47.5	3.7	0.6	0.1
Eleocharis sp.	42.3	24.1	-30.5	56.6	3.1	0.6	0.8
Pistia stratiotes	37.1	15	-29.7	25.9	0.4	0.9	0.1
Paspalum repens	41.6	14.9	-12.6	47.9	1.7	0.4	0.1
Paspalum repens (root)	38.5	27.1	-13.6	93.9	4	0.7	0.2
Wetland soil							
Janauaca	0.6	6.1	-27.3	25.5	0.2	0.6	1.2
Janauaca stream	0.4	8.3	-27.8	63.2	0.5	1.1	1
Amazon river	0.4	9.8	-28.4	9	0.5	1	1.3
Amazon river	1	6.7	-18.7	67.5	0.5	0.9	0.9
Soil (Terra Firme)							
Canaçari	2.1	16.3	-27.4	36.6	0.3	0.8	0.6
Amazon river	4.2	14	-28.7	9.7	0.5	0.5	1.4
Amazon river	2.3	11.1	-29	88.3	0.4	0.5	1.5

Table 3. Average values for the seasonality and spatiality of bulk parameters, lignin phenols and GDGTs in sediment samples from the floodplain lakes

	n	TOC (wt.%)	C:N	δ <sup>13</sup> C <sub>org</sub> (‰ VPDB)	Λ8 (mg g <sub>oc</sub> <sup>-1</sup> )	C:V	S:V	(Ad:Al)v	brGDGTs (µg g <sub>oc</sub> -1)	Crenarchaeol $(\mu g g_{OC}^{-1})$	IPL brGDGTs (%)	IPL crenarchaeol (%)
Cabaliana	10	3.3±0.8	10.9±0.8	-33.0±1.6	32±17	0.6±0.3	1.0±0.2	1.6±0.5	33±19	6±6	15±8	9±8
Janauaca	11	2.7±1.0	10.9±1.4	-32.2±1.5	50±41	0.6±0.3	1.1±0.1	1.2±0.5	41±23	4±3	14±9	15±9
Mirituba	11	2.3±1.0	11.3±1.7	-29.3±0.9	57±26	0.7±0.4	1.0±0.2	1.4±0.5	33±16	8±6	14±8	18±10
Canaçari	10	2.0±0.6	10.9±1.4	-30.0±1.3	42±38	0.9±0.6	1.1±0.2	1.2±0.6	44±21	12±6	9±8	11±9
Curuai	15	2.1±0.4	10.0±0.9	-27.0±0.8	41±21	0.9±0.3	1.1±0.2	1.0±0.4	31±14	9±7	9±7	15±10
LW	12	2.3±0.2	10.2±1.2	-30.0±2.3	29±12	0.7±0.4	0.9±0.1	1.5±0.4	38±16	10±6	19±7	23±9
RW	15	2.7±0.8	10.6±1.3	-30.1±2.5	56±30	0.8±0.5	1.1±0.2	1.0±0.5	35±12	7±5	8±6	10±5
HW	12	2.6±1.1	11.1±1.5	-29.7±3.0	23±9	0.8±0.4	1.0±0.2	1.7±0.5	24±16	4±4	10±9	17±16
FW	18	2.2±0.9	11.0±1.3	-30.2±2.4	62±33	0.6±0.2	1.1±0.2	1.1±0.4	45±23	9±7	9±9	8±7

Table 4. Values of long-chain *n*-alkanes  $\delta^{13}$ C in surface sediment samples from the upstream lake Janauaca and the downstream lake Curuai. The samples were collected in the LW season.

	C <sub>27</sub>	C <sub>29</sub>	C <sub>31</sub>
Janauaca	-33.7±0.2	-33.8±0.2	-34.8±0.2
Curuai	-31.2±0.3	-31.5±0.3	-32.2±0.3
Table 5. Average values of biomarkers and bulk parameters in the possible sources of SOM and in sediment samples. The data was obtained in the present work and in the literature (Hedges et al., 1986; Hedges and Mann, 1979; Aufdenkampe et al., 2007).

	TOC	C·N	$\delta^{13}C_{org}$	C·W	c.V		$\lambda 8$	brGDGTs	crenarchaeol
	(WL.%)	C:N	( <sup>%</sup> VPDD)	C:V	5:V	(Au:Al)v	(ing goc)	(µg goc)	(µg goc)
Wetland soil	0.9	8.3	-27.0	0.4	0.9	1.1	41.3	39.6	2.9
Soil (terra firme)	1.6	10.5	-27.6	0.4	0.6	1.2	44.9	21.1	0.5
River (CPOM)	1.4	4.8	-31.4	0.1	0.7	0.2	40.0		
River (FPOM)	2.2	7.2	-29.9	0.1	0.9	0.6	16.1	77.4	25.9
Macrophyte	36.6	28.7	-24.7	1.9	0.6	0.3	59.0		
Grass/Leave	46.7	28.1	-30.1	0.4	1.1	0.2	37.2		
Phytoplankton	13.9	6.7	-31.1						
Wood	46.5	217.7	-27.6	0.0	1.5	0.1	193.3		
Sediment	2.4	10.7	-30.0	0.7	1.1	1.3	43.6	36.1	7.8

Table 6. Data used in equations 1 and 2 to estimate the fraction of OM derived frommacrophytes and aquatic OM to the SOM.

					Estimated
Parameter	SOM	Macrophytes	<b>Rive SPOM</b>	Other OM sources	fraction (%)
C:V	0.7±0.4	1.9		0.2	29
crenarchaeol					
$(\mu g g_{OC}^{-1})$	7.8±6.0		26.0	1.2	27

## **Figure captions**

Figure 1. Study area of the central Amazon basin (A) showing five floodplain lakes (várzeas) in squares B, C, and D.

Figure 2. Seasonal water level changes of the Amazon River main stem at the town Óbidos (RW=rising water, HW=high water, FW=falling water, LW=low water).

Figure 3. Box plots of bulk OC parameters, lignin phenols, and GDGTs along the upstream-downstream transect. The midpoint of a boxplot is the mean. The 25% and 75% quartiles define the hinges (end of the boxes), and the difference between the hinges is the spread. Letters indicate statistically significant groups of data (p < 0.05).

Figure 4: Box plots of total lignin phenols ( $\lambda$ 8), and GDGTs for four hydrological seasons. The midpoint of a boxplot is the mean. The 25% and 75% quartiles define the hinges (end of the boxes), and the difference between the hinges is the spread. Letters indicate statistically significant groups of data (*p* <0.05).

Figure 5: Box plots of average values of multiple biomarkers and bulk parameters in sediment samples and in potential sources on SOM. Data is based on previous studies (Hedges et al., 1986; Aufdenkampe et al., 2007; Zell et al., 2013) and the present work (Table 3). Letters over the boxes indicate significant differences (p<0.05) between the means.

Figure 6. Box plots of seasonal average values of total lignin phenol and brGDGTs. Letters indicate statistically significant groups of data (p<0.05).

List of major Reviews

Original text
---------------

P.4,L.8 and further, the organic matter (OM) produced in the floodplain lakes fuels the outgassing CO<sub>2</sub> in the river system (Abril et al., 2014). Periodical floods intensify the exchange of organic compounds, nutrients and minerals between rivers, lakes and flooded soils (Junk, 1997).

P.4. In the Amazon basin, many studies L.15 have characterized the OM in the In the system is forests and upstream Andean Andean soils soils

P4, in the floodplains in general, and L21 in floodplain lakes in particular, and the contribution of the multiple sources of OM (up- land soils, flooded forest, macrophytes, aquatic and phytoplankton) remain uncertain

P4-5, The spatiality and the seasonality of L.25 the hydrology in the Amazon basin strongly influence the dynamics and the quality of OC in the surface sediments. Most of the SOM is supposed to be transported to the floodplain lakes via river main streams during the high water season (Hedges et al., 1986; Victoria et al., 1992; Moreira Turcq et al., 2004; Mortillaro et al., 2012; Moreira-Turcq et al., 2013). However, a significant increase in vertical flux of OC was observed in lake Curuai during the falling water season, which is interpreted as the result of a process of concentration of In downstream lakes, higher values of (Victoria et al., 1992) bulk  $\delta^{13}$ C were found in the sediments, when compared to upstream lakes (Victoria et al., 1992).

**Reviewed text** 

Inputs of CO<sub>2</sub> from plant respiration and reactive OC produced in floodplain lakes are significant sources of CO<sub>2</sub> outgassed in the central Amazon basin (Abril et al., 2014). The periodic floods intensify the exchange of organic compounds, nutrients and minerals between rivers, lakes and flooded soils (Junk, 1997).

Amazon basin, many studies have suspended particulate organic matter characterized the suspended particulate organic (SPOM) in the rivers system and in the matter (SPOM) in the rivers and the floodplain floodplain lakes and concluded that the lakes and concluded that the main source of OM to main source of OC to the aquatic the aquatic system is the forests and the upstream

> in the floodplain lakes in general, and in particular, the contribution of the multiple sources of OM (upland soils, flooded and non-flooded forests, aquatic macrophytes, and phytoplankton) remains uncertain

The seasonality and the spatiality of the wetlands in the Amazon basin strongly influence the dynamics and the quality of OM in the surface sediments of floodplain lakes. Most of the SOM is supposed to be transported to the floodplain lakes via Amazon River mainstem during the rising and high water seasons (Hedges et al., 1986; Victoria et al., 1992; Moreira-Turcq et al., 2004; Mortillaro et al., 2011; Moreira-Turcq et al., 2013). However, a significant increase in the vertical flux of OM was observed in lake Curuai during the falling water season, which is interpreted as the result of resuspended sediments when the lake becomes smaller and shallower periodically resuspended sediments as (Moreira-Turcq et al., 2004). In terms of the lakes are becoming smaller and spatiality, the downstream lakes present higher shallower (Moreira-Turcq et al., 2004). values of  $\delta^{13}C_{org}$  in comparison to the upstream lakes

|--|

P.5, L15

> recalcitrant organic Lignin is а macromolecule composed of phenolic molecules and produced by vascular The products of CuO plants. degradation of lignin (Hedges and Ertel, 1982) have been widely applied as biomarkers to trace plant material to aquatic systems (Hedges et al., 1986; Bernardes et al., 2004; Aufdenkampe et al., 2007; Kuzyk

> component of fossil OC in floodplain lakes (Zocatelli et al., 2013) but also a relevant source for the outgassing of  $CO_2$  in the Amazon River (Ward et al., 2013). BrGDGTs and crenarchaeol have been applied to quantify the OC proportion originating from soils (Hopmans et al., 2004; Herfort et al., 2006; Belicka and Harvey, 2009; Smith et al., 2010) and have recently and aquatic sources of OC in rivers and (Kim et al., 2012; Zell et al., 2013a; Moreira et al., 2014).

Lignin is produced by vascular plants. It is composed of phenolic compounds and general considered as a recalcitrant organic molecule. As a consequence, the products of CuO degradation of lignin (Hedges and Ertel, 1982) have been widely applied as biomarkers to trace plant material to aquatic systems (Hedges et al., 1986; Bernardes et al., 2004; Aufdenkampe et al., 2007; Kuzyk et al., 2008). Previous works in the Amazon basin showed that lignin is an important component of fossil OC in floodplain lakes (Zocatelli et al., 2013) but also a relevant carbon source for the outgassing of CO<sub>2</sub> in the Amazon River (Ward et al., 2013). This apparently contradiction reflects the relevance of environmental conditions on the degradation of organic molecules (Schmidt et al. 2012) which must considered in the application of these be et al., 2008). It can be an important biomarkers. The GDGTs are membrane lipids mainly composed of acyclic or cyclic biphytane core lipids with two glycerol head groups (Hopmans et al., 2000). The head groups are easily degraded while the two biphytanyl core lipids are well preserved in sediments and soils (White et al., 1979; Harvey et al., 1986). The GDGTs are found in diverse environments worldwide but, the brGDGTs are mainly produced in the soil, by the bacteria domain (e.g., Weijers et al., 2006), and the crenarchaeol is predominant in the aquatic been applied to identify the terrestrial environments and produced by Thaumarchaeota (Sinninghe Damsté et al., 2002). Accordingly, the floodplain lakes of the Amazon basin relative amount of brGDGTs to the crenarchaeol, so-called the Branched and Isoprenoid Tetraether (BIT) index, have been proposed to quantify the OC proportion originating from soils and aquatic environments (e.g., Hopmans et al., 2004; Herfort et al., 2006; Belicka and Harvey, 2009; Smith et al., 2010). Previously, this method was successfully applied in rivers and floodplain lakes of the Amazon basin (e.g., Kim et al., 2012; Zell et al., 2013a; Moreira et al., 2014).

P.6, white water (e.g., Solimões, Madeira, and Amazon L.11 rivers), black water (e.g., Negro River), and clear and Amazon), black water (e.g. Negro), and clear water (e.g. Tapajós) water (e.g., Tapajós River)

the Manacapuru River discharges clear water while in the southern region, the white water brought by the Solimões River, mixes with clear water. Janauaca has a peculiar morphology with a ravine shape surrounded by flooded forests (Fig. 1b). Solimões water comes through the channel in the north, and clear water comes through the stream system in the south. Mirituba has a round shape	the Manacapuru River discharges black water while in the southern region, the white water brought by the Solimões River, mixes with black water. Janauaca has a peculiar morphology with a ravine shape surrounded by flooded forests (Fig. 1B). Solimões water comes through the channel in the north, and some clear water comes through the stream system in the south. Conductivity values in Lakes Cabalina and Janauaca are close to that of the Solimões river, evidencing that white waters predominate. Mirituba has a round shape
It receives white water from the Amazon River through small channels, apart from the main channel in the eastern side. There are small contributions of black water streams in its most Southeastern part.	It receives in majority white water from the Amazon River through channels connected to the main stem, .Small contributions of black water streams occur in the Curuai floodplain, but remain spatially restricted to their most Southeastern region.
The four hydrological seasons were targeted during different research cruises with a small research vessel (Fig. 2): CBM5 in June and July 2009 covered the High Water (HW) season; CBM6 in October 2009 covered the Low Water (LW) season, CBM7 in August 2010 covered the Falling Water (FW) season and CBM8 in January 2011 covered the Rising Water (RW) season. In each floodplain lake, sediment samples were collected at three stations in each season.	The four hydrological seasons were targeted during different research cruises with a small vessel (Fig. 2): in June and July 2009, which covered the high water (HW) season; in October 2009 the low water (LW) season, in August 2010 the falling water (FW) season and in January 2011 the rising water (RW) season. In each floodplain lake, sediment samples were collected at three stations in each season.
However, sometimes only two samples were collected when stations were not accessible during a specific season.	However, sometimes only two samples were collected when stations were not accessible during a specific season. Thus, it was obtained approximately 12 samples in each lake and 15 samples per season. The samples were collected in the three most distinct sites of each lake: near the connecting channel, in the meddle and near the flooded forests. Most of the he sampling sites were located in areas flooded by the Solimões-Amazon river water (white water). In Cabaliana and Canacari one station was located near the black water streams in order to represent the heterogeneity of the lakes.
	the Manacapuru River discharges clear water while in the southern region, the white water brought by the Solimões River, mixes with clear water. Janauaca has a peculiar morphology with a ravine shape surrounded by flooded forests (Fig. 1b). Solimões water comes through the channel in the north, and clear water comes through the stream system in the south. Mirituba has a round shape It receives white water from the Amazon River through small channels, apart from the main channel in the eastern side. There are small contributions of black water streams in its most Southeastern part. The four hydrological seasons were targeted during different research cruises with a small research vessel (Fig. 2): CBM5 in June and July 2009 covered the High Water (HW) season; CBM6 in October 2009 covered the Low Water (LW) season, CBM7 in August 2010 covered the Falling Water (FW) season and CBM8 in January 2011 covered the Rising Water (RW) season. In each floodplain lake, sediment samples were collected at three stations in each season.

P.7, L.24	Four riverbank sediments and three soils from well above the inundations known as "terra firme"	Four wetland soils and three nonfloodable soils from well above the maximum inundations levels, known as "terra firme"
P.8, L.5	Total carbon (TC), total nitrogen (TN), and $\delta^{13}$ C for the samples obtained during the CBM5 and CBM6 cruises	For the samples collected during the HW and LW seasons, total carbon (TC), total nitrogen (TN), and $\delta^{13}$ C of TC
P.8, L.9	For samples obtained during the CBM7 and CBM8 cruises were analysed	Other samples gathered during the FW and RW cruises were analyzed for TC, TN, and $\delta^{13}$ C of TC
P.8, L.16	TC (wt. %) and correlated very well with TOC (wt. %) with a +0.16 intercept (R 2 = 0.96; p < 0.001; n = 16). This indicates that TC in floodplain lakes sediments investigated was mostly TOC. Therefore, we considered TC as TOC in this study. In order to assess contribution of inorganic nitrogen (NH +4 + NO 2 + NO 3 ) to TN, TN (wt. %) and TOC (wt. %) were correlated (R = 0.89; p < 0.001; n = 57). It showed that	TC (wt.%) correlated very well with TOC (wt.%) with an intercept no significant from 0 (R <sup>2</sup> =0.96, p<0.001, n=16). This indicates that TC in floodplain lakes sediments investigated was predominantly TOC and the fraction of carbonates was minor. Therefore, TC was considered as TOC in this study. In order to assess contribution of inorganic nitrogen (NH <sup>+4</sup> + NO <sup>-2</sup> + NO <sup>-3</sup> ) to TN, TN (wt.%) and TOC (wt.%) were correlated (R <sup>2</sup> =0.89; p<0.001; n=16). The interception of the correlation line on the TN axis (0.06) was interpreted as the percentage of inorganic nitrogen, suggesting that
P.9, L.2	Approximately 500 mg of freeze-dried sediments and macrophytes were analyzed for lignin monomers using the alkaline CuO oxidation method	The lignin phenols were extracted from approximately 500 mg lake sediment and soil samples and from 50 mg of macrophytes samples. The samples were freeze-dried and the extraction method applied was the alkaline CuO oxidation
P.9, L.13	The recovery factor was calculated using he internal standard ethyl vanillin added prior to analysis (values above 60 % were considered).	The recovery factor was calculated using the internal standard ethyl vanillin added after the CuO oxidation and prior to analysis (values above 60% were considered).

P.9, L.16	To confirm the identification of each lignin phenol, eight selected samples were analyzed with an Agilent 7890A gas chromatography coupled to an Agilent 5975C VL MSD mass spectrometer using a selective ion monitoring (SIM) at NIOZ (The Netherlands).	To confirm the identification of each lignin phenol, eight selected samples were analyzed with an Agilent 7890A gas chromatograph (GC-FID) coupled to an Agilent 5975C VL MSD mass spectrometer using a selective ion monitoring (SIM) at NIOZ (The Netherlands).
P. 10, L.23	The core lipids and IPL-derived GDGTs were analyzed using high performance liquid chromatography- atmospheric pressure positive ion chemical ionization mass spectrometry (HPLC-APCI-MS) in selected ion monitoring (SIM) mode according to Schouten et al. (2007).	The core lipids and IPL GDGTs were analyzed using high performance liquid chromatograph- atmospheric pressure positive ion chemical ionization-mass spectrometry (HPLC-APCI-MS, an Agilent 1100 series LC/MSD SL, Alltech Prevail Cyano column (150 × 2.1 mm × 3 $\mu$ m)) in a selected ion monitoring (SIM) mode according to Schouten et al. (2007).
P.11, L.8	The n-alkanes in the apolar fraction were identified by a Thermo Finnigan Trace DSQ gas chromatography (GC- MS) and quantified with an HP 6890 GC system.	n-alkanes in the apolar fractions were identified by a Thermo Finnigan Trace DSQ gas chromatograph- mass spectrometry (GC-MS) and quantified with an HP 6890 GC system.
P.11, L.14		Thermo Delta V Advantage and the results were obtained using the software Isodat 3.0. Isotope

Thermo Delta V Advantage and the results were obtained using the software Isodat 3.0. Four injections were performed for each sample to calculate the analytical error.

Thermo Delta V Advantage and the results were obtained using the software Isodat 3.0. Isotope values were measured against calibrated external reference gas and performance of the instrument was monitored by daily injects of a mixture of a  $C_{20}$  and a  $C_{24}$  perdeuterated *n*-alkane with known isotopic compositions. The  $\delta^{13}$ C values for the *n*-alkanes are reported in the standard delta notation against the Vienna Pee Dee Belemnite (VPDB) standard. All samples were run four times with an average standard deviation of 0.3 ‰ for the  $C_{25}$  *n*-alkane, 0.3 ‰ for the  $C_{29}$  *n*-alkane, and 0.2 ‰ for the  $C_{31}$  *n*-alkane (Sinninghe-Damsté et al., 2011).

P.12, L.3	The TOC content showed lower mean value (Table 2) in the downstream lake Curuai (2.0 $\pm$ 0.6 wt. %) and the highest mean value was found in Cabaliana (3.3 $\pm$ 0.8 wt. %; Fig. 3a). No significant seasonal variation was observed (Fig. 4a). The C:N ratio did not reveal significant spatial and seasonal variations (Figs. 3B and 4b). The lowest mean value was found in Curuai (10 $\pm$ 1) and the highest one in Mirituba (11 $\pm$ 2). The $\delta^{13}C_{org}$ values were significantly less negative in the downstream lakes (Fig. 3c). In Curuai the mean value was $-27 \pm 1 \%$ and in Cabaliana $-33 \pm 2 \%$ . No significant seasonal variation was observed for the $\delta^{13}C_{org}$ values (Fig. 4c)	The TOC content was the lowest in the downstream lake Curuai (2.0±0.6 wt.%) and the highest in lake Cabaliana (3.3±0.8 wt.%) (Fig. 3a, Table 3). No significant seasonal variation was observed (p=0.145) (Fig. 4A). The C:N ratio did not reveal significant spatial (p=0.104) and seasonal (p=0.418) variations (Figs. 3B and 4B). The $\delta^{13}C_{org}$ values were significantly less negative in the downstream lakes (p<0.001) (Fig. 3C). In lake Curuai the mean value was $-27\pm1\%$ and in lake Cabaliana $-33\pm2\%$ . No significant (p=0.968) seasonal variation was observed for the $\delta^{13}C_{org}$ values (Fig. 4C).
P.12, L.11	The $\delta^{13}$ C values in soils and	The $\delta^{13}C_{org}$ values in "terra firme" soils and wetland
	riverbank sediment samples varied between $-29$ and $-19 \%$ (n = 7)	soils varied between –29 and –19‰ (n=7). These samples were collected in the Amazon River margin between Canaçari and Curuai.
P.12, L.14	The C <sub>3</sub> macrophytes ( <i>Eleocharis</i> sp. And <i>Pistia stratiotes</i> ) had $\delta^{13}C_{org}$ values of $-30 \%$ and values of the C:N ratio between 15 and 24 (Table 2).	The C <sub>3</sub> macrophytes ( <i>Eleocharis sp.</i> and <i>Pistia stratiotes</i> ) had $\delta^{13}C_{org}$ values between $-30\%$ and $-33\%$ and C:N ratios between 15 and 24 (Table 2).

- P.12, No significant changes were observed
- L.18 along the transect for the mean values of  $\lambda 8$  (i.e. a proxy for the amount of lignin); the mean value of  $\lambda 8$  for the SOC was 44  $\pm$  29 mg g<sub>oc</sub><sup>-1</sup> . However,  $\lambda$ 8 values revealed significant seasonal changes. The higher values were observed in the RW ( $56 \pm 30 \text{ mg g}_{oc}^{-1}$ ) and in the FW seasons (62  $\pm$  34 mg  $g_{oc}^{-1}$ ) compared to the HW (23  $\pm$  9 mg  $g_{oc}^{-1}$ ) and LW (29  $\pm$  12 mg  $g_{\rm oc}{}^{\rm -1}$  ) seasons (Fig. 3g). The C:V ratio showed no significant seasonal and spatial variation, and the mean value for all sediment samples was  $0.7 \pm 0.4$  (Figs. 3d and 4d). The values of the S:V ratio did not show significant spatial differences either but higher mean values in the RW season  $(1.1 \pm 0.1)$  and in the FW season  $(1.2 \pm$ 0.2) were observed in comparison to that in the LW season (0.9  $\pm$  0.1; Fig. 4e). The mean value of (Ad:Al)v ratio for the different lakes did not show spatial variation (Fig. 3f), however, it was higher in the LW  $(1.5 \pm 0.4)$  and HW (1.7  $\pm$  0.5) seasons for most lakes (Fig. 4f).
- P.13, For the  $C_3$  macrophytes,  $\lambda 8$  values L.4 varied between 50–60 mg  $g_{OC}^{-1}$  and between 70-160 mg  $g_{OC}^{-1}$  for the C<sub>4</sub> macrophyte samples. The S:V ratio varied between 0.4 and 0.6 for C<sub>3</sub> macrophytes and between 0.4 and 0.8 for the C<sub>4</sub> macrophyte. The range of C:V ratio was 1.0 to 3.1 for the  $C_3$ macrophytes and 1.4 to 2.7 for the  $C_4$ macrophytes. The (Ad : Al)v ratio varied between 0.2 and 0.8 for all macrophyte samples (Table 3). For the riverbank and wetland soil samples, the  $\lambda 8$  values varied between 8 and 88 mg  $g_{OC}^{-1}$ . The S:V ratio varied between 0.5 and 1, the C:V ratio varied between 0.2 and 0.5, and the (Ad: Al)v ratio varied between 0.6 and 1.5.

upstream-downstream No significant changes (p=0.392) were observed along the upstream-downstream transect for the mean values of  $\lambda 8$  (i.e. a proxy for the amount of lignin normalized to OC); The mean value of  $\lambda 8$ , a proxy for the amount of lignin normalized to OC, for the SOC was 44±29 mg g<sub>oc</sub><sup>-1</sup>. No significant changes (p=0.392) were observed along the upstream-downstream transect for the mean values of  $\lambda 8$ ); However,  $\lambda 8$  values revealed significant seasonal changes (p=0.001). The higher values were observed in the RW (56 $\pm$ 30 mg g<sub>oc</sub><sup>-1</sup>) and in the FW seasons (62±34 mg g<sub>oc</sub><sup>-1</sup>) compared to the HW (23±9 mg  $g_{oc}^{-1}$ ) and LW (29±12 mg  $g_{oc}^{-1}$ ) seasons (Fig. 4G). The C:V ratio showed no significant seasonal (p=0.609) and spatial variation (p=0.214), and the mean value for all sediments was 0.7±0.4 (Figs. 3D and 4D). The values of the S:V ratio also does not show significant spatial (p=0.568) or seasonal (p=0.08) differences. The mean values for the lakes were approximately 1.1±0.1 and the mean seasonal values varied between 0.9±0.1 and 1.1±0.2 (Fig. 4E and Table 3). The mean value of (Ad:Al)v ratio for the different lakes does not show spatial variation (p=0.137) (Fig. 3F), however, it was higher in the LW  $(1.5\pm0.4)$  and HW  $(1.7\pm0.5)$ seasons (p<0.001) (Fig. 4F).

For the  $C_3$  macrophytes,  $\lambda 8$  values varied t etween 26–67 mg  $g_{OC}^{-1}$  and between 48–94 mg  $g_{OC}^{-1}$  for the  $C_4$  macrophytes. The S:V ratio varied between 0.6 and 0.9 for  $C_3$  macrophytes and between 0.4 and 0.7 for the  $C_4$  macrophytes. The range of C:V ratio was 0.4 to 3.7 for the  $C_3$  macrophytes and 1.7 to 4.0 for the  $C_4$  macrophytes. The (Ad:Al)v ratio varied between 0.2 and 0.8 for all macrophyte samples (Table 3). For the "Terra Firme" soil and wetland soil samples, the  $\lambda 8$  values varied between 9 and 88 mg g<sup>-1</sup>. The S:V ratio varied between 0.5 and 1.1, the C:V ratio varied between 0.6 and 1.5.

P. 13, Along the upstream-downstream L.13 transect, no significant changes were observed for the mean values of brGDGTs concentrations (Fig. 3h). The lowest value was found in Curuai (31 ± 14  $\mu g \ g_{\text{oc}}^{-1}$  ) and the highest one in Canaçari (44 ± 22  $\mu$ g g<sub>oc</sub><sup>-1</sup>). The mean concentrations of crenarchaeol were higher in Canaçari (115 ± 57 µg  $g_{oc}^{-1}$ ) when compared to Janauaca ( $34 \pm 33$ )  $\mu g_{oc}^{-1}$ ). However, no significant difference was observed between the upstream (Cabaliana and Janauaca) lakes and the downstream lake (Curuai; Fig. 3h and i). On the other hand, brGDGTs concentrations showed significant seasonal changes. The highest mean value for brGDGTs concentrations was found in the FW season (45  $\pm$  23 µg g<sub>oc</sub><sup>-1</sup>), and the lowest mean concentration was found in the HW season (24  $\pm$  16 µg g<sub>oc</sub><sup>-1</sup>). The RW and LW seasons showed intermediate mean concentrations (35  $\pm$  12 and 38  $\pm$  16 µg g<sub>oc</sub><sup>-1</sup>, respectively) and no significant difference was observed if compared to the FW and HW seasons (Fig. 4h). The concentrations of crenarchaeol did not reveal significant changes over the hydrological seasons (Fig. 4i). The mean values varied between  $5 \pm 4$  and  $10 \pm 6 \ \mu g \ g_{oc}^{-1}$  in the HW and LW seasons, respectively. The percentage of IPL brGDGTs and IPL crenarchaeol was significantly higher in the LW season (19  $\pm$  7 and 23  $\pm$  9 %, respectively). In the other three seasons, it showed values around 10  $\pm$ 2 % of IPL brGDGTs and IPL crenarchaeol significant with no variability (Table 3).

Along the upstream-downstream transect, no significant changes (p=0.371) were observed for the mean values of brGDGTs concentrations (Fig. 3H). The lowest value was found in Curuai (31±14 µg  $g_{oc}^{-1}$ ) and the highest one in Canaçari (44±22 µg  $g_{oc}^{-1}$ <sup>1</sup>). The average concentration of crenarchaeol was higher in Canaçari ( $12\pm 6 \mu g g_{oc}^{-1}$ ) when compared to Janauaca ( $4\pm3 \ \mu g \ g_{oc}^{-1}$ ). However, no significant difference (p=0.127) was observed between the upstream (Cabaliana and Janauaca) lakes and the downstream lake (Curuai) (Fig. 3H and 3I). On the other hand, brGDGTs concentrations showed significant seasonal changes (p=0.025). The highest mean value for brGDGTs concentrations was found in the FW season (45±23  $\mu$ g g<sub>oc</sub><sup>-1</sup>), and the lowest mean concentration was found in the HW season  $(24\pm16 \ \mu g \ g_{oc}^{-1})$ . The RW and LW seasons showed intermediate mean concentrations (35±12 µg  $g_{oc}^{-1}$ and  $38\pm16 \ \mu g \ g_{oc}^{-1}$ , respectively) and no significant difference (p=0.335) was observed if compared to the FW and HW seasons (Fig. 4H).

The concentrations of crenarchaeol did not reveal significant changes (p=0.096) over the hydrological seasons (Fig. 4I). The mean values varied between  $4\pm4 \ \mu g \ g_{oc}^{-1}$  and  $10\pm6 \ \mu g \ g_{oc}^{-1}$  in the HW and LW seasons, respectively. The percentage of IPL brGDGTs and IPL crenarchaeol was significantly higher (p=0.002 and p<0.001, respectively) in the LW season ( $19\pm7$  and  $23\pm9\%$ , respectively). In the other three seasons, it showed values around 10±2% of IPL brGDGTs and IPL crenarchaeol with no significant variability (Table 3).

P.14,

L.5 summarized in Table 6.

The results of n-alkane analyses are The results of n-alkane analyses are summarized in Table 4.

P.14, L.10	In the upstream lake, the mean $\delta$ 13 C for the long-chain n-alkanes was -34.1 $\pm$ 0.5 ‰ and in the downstream lake the mean value was -31.6 $\pm$ 0.6 ‰ (Table 6)	In the upstream lake, the mean $\delta^{13}$ C for the long- chain n-alkanes was $-34.1\pm0.5\%$ and in the downstream lake the mean value was $-31.6\pm0.6\%$ (Table 4)
P.14,		
L.15	To determine the origin of the SOM in	To determine the origin of the SOM in the
	the floodplain lakes, we considered	floodplain lakes, we considered five potentially
	five potential sources: (1) the terrestrial	significant sources in the Central Amazon Basin
	Andean clay-bounded and refractory	(Hedges et al., 1986; Moreira-Turcq et al. 2013;
	SPOM, which may be transferred to	Mortillaro et al., 2011): (1) the terrestrial Andean

the floodplain lakes via the Solimões-Amazonas and Madeira rivers (Hess et transferred to the floodplain lakes via the Solimõesal., 2003), (2) "terra firme" soil and Amazon and Madeira rivers (Hess et al., 2003), (2)

	litter of the Amazonian lowland forest , which will be transferred to the floodplain lakes via streams, (3) the wetland soil (flooded forests) and litter (leaves, grasse, wood etc.)	"terra firme" soils and littesr of the Amazonian lowland forests (non-floodable forests), which will be transferred to the floodplain lakes via local streams, (3) the wetland soils (flooded forests) and litters (leaves, grasses, woods etc.),
P.15, L.12	are compared with those of the SOM of the floodplain lakes in Fig. 5 and Table 4	are compared with those of the SOM of the floodplain lakes in Fig. 5 and Table 5
P.15, L.18	wood samples are significantly different from those	wood samples are significantly different (p<0.001
P. 16, L.19	Crenarchaeol is, therefore, considered as an (indirect) indicator of aquatic primary production. The enhanced concentrations of crenarchaeol in SOM thus indicate a contribution from this source.	Crenarchaeol is, therefore, considered as an indicator of aquatic OM production in this system. The enhanced concentrations of crenarchaeol in SOM thus indicate an increased contribution from riverine and/or lacustrine SPOM.
P.16, L.26	soils samples, riverine SPOM and lake sediments.	soils samples (p=1.241), riverine SPOM (p=1.044) and lake sediments.
P.17, L.2	specific source of SOM (i.e., macrophytes and aquatic production in the rivers or floodplain lakes, respectively)	specific source of SOM (i.e., macrophytes and aquatic OM from rivers or floodplain lakes, respectively).

#### P.17, L.5

el al., 2003), the C:V values of macrophytes and the average values of soil and riverine SPOM samplescan be used to estimate the contribution of macrophyte OM to the SOM.

According to this approach (Martinelli et al., 2003), the average C:V values of macrophytes and the According to this approach (Martinelli average values of other OM sources (wetland and non-flooded soils and SPOM) can be used to estimate the contribution of macrophyte OM to the SOM (Eq. 1). Similarly, the concentration of crenarchaeol in the riverine SPOM and its concentration in soil samples can be used to estimate the contribution of aquatic OM to the SOM (Eq. 2).

$$F_{\text{macrophytes}} = \frac{\text{C:V}_{\text{SOM}} - \text{C:V}_{(\text{other})}}{\text{C:V}_{\text{macrophyte}} - \text{C:V}_{(\text{other})}} \times 1 \quad F_{\text{macrophytes}} = \frac{\text{C:V}_{\text{SOM}} - \text{C:V}_{(\text{SPOM+forest})}}{\text{C:V}_{\text{macrophyte}} - \text{C:V}_{(\text{SPOM+forest})}} \times 100$$

$$F_{\text{aquatic}} = \frac{\text{Cren}_{\text{SOM}} - \text{Cren}_{(\text{other})}}{\text{Cren}_{\text{SPOM}} - \text{Cren}_{(\text{other})}} \times 100 \quad F_{\text{SPOM}} = \frac{\text{Cren}_{\text{SOM}} - \text{Cren}_{(\text{forest+macrophyte})}}{\text{Cren}_{\text{spoM}} - \text{Cren}_{(\text{forest+macrophyte})}}} \times 100$$

$$F_{\text{wetlands}} = 100 - (F_{\text{aquatic}} + F_{\text{macrophytes}}) \quad F_{\text{macrophyte}} + F_{\text{SPOM}} + F_{\text{forest}} = \text{SOM} (100\%)$$

#### P.17, L.14

In Eqs. (1) and (2), the  $F_{macrophytes}$  and F<sub>aquatic</sub> represent the estimated fractional abundance in SOM of macrophytes and SPOM, respectively. C:V<sub>SOM</sub> and Cren<sub>SOM</sub> are the average values of each parameter found in the sediment samples, C:V<sub>macrophytes</sub> and Cren<sub>SPOM</sub> are the values of the source of the respective parameter and C:V<sub>other</sub> and Cren<sub>other</sub> are the values of the other possible sources (Table 5). These calculations indicate that 20-30 % of the SOM is derived from macrophytes and 20–30 % from the aquatic production either in the river or in the floodplain lake itself. Consequently, the remaining 40-60 % of the SOM might be derived from other sources of OM such as the flooded forests (Eq. 3). floods link The periodical the floodplain lakes and the wetland vegetation and soil. Thus, the seasonal and spatial contrasts in the SOM should be investigated in order to better understand the connectivity between these compartments.

In Eqs. (1) and (2), the  $F_{macrophytes}$  and  $F_{SPOM}$ represent the estimated fractional abundance of macrophytes and aquatic OM in SOM, respectively. C:V<sub>SOM</sub> and Cren<sub>SOM</sub> are the average values of each parameter found in the sediment samples, C:V<sub>macrophytes</sub> and Cren<sub>SPOM</sub> are the values of the predominant source of the respective parameter and C:V<sub>(SPOM+forest)</sub> and Cren<sub>(forest+macrophyte)</sub> are the values of the other possible OM sources. As discussed above, the high values of (Ad:Al)v indicate that lignin components of the SOM is partially degraded, which may affect the values of the C:V ratio. There also numerous complications with the are application of crenarchaeol as an indicator of aquatic matter in this ecosystem. Therefore, the presented mixing model should be considered as estimations. The results of Eq. 1-3 indicate that 25-35% of the SOM is derived from macrophytes and 20-30% from aquatic OM sources (riverine and lacustrine SPOM). Consequently, the remaining 35-55% of the SOM might be derived from the wetlands and non-flooded forests (Eq. 3). The periodic floods link the floodplain lakes and the wetland vegetation and soil. Thus, the seasonal and spatial contrasts in the SOM should be investigated in order to better understand the connectivity between these compartments.

P.19, are listed in Table 6. Accordingly, the Table 4. Accordingly, the fraction of C<sub>4</sub> plants in the percentage of C 4 plants in the SOM in the upstream lake is only 3%, but for the upstream lake is only 3 %, but for the downstream lake 22%.

P.20, L.2

> The  $\lambda 8$  values and the S : V ratio show significantly higher values in the RW and FW seasons (Figs. 4e, g, and 6a) in all lakes. The mean concentrations of brGDGTs also show higher values in the FW season (Figs. 4h and 6b). The co occurrence of these two types of molecules indicate that litter, traced by lignin phenols, and superficial soil, traced by brGDGTs, are preferentially deposited during rising and receding waters, which increases the wetland soil runoff. Besides, the seasonal mean values of (Ad : Al)v show remarkably lower values in the RW and FW seasons (Fig. 4f), an inverse pattern if compared to the S : V ,  $\lambda 8$  and brGDGTs. This means that less degraded lignin is present in the surface sediments in the RW and FW seasons. Thus, the increase in the concentrations of the organic compounds is not a consequence of the re-suspension of the sediments, but to the arrival of fresher OM. In the HW and LW seasons, more degraded lignin phenols (higher values of (Ad : Al)v) present in the sediments are concomitant with lower amounts of  $\lambda 8$ and S : V ratio. Since the concentration of crenarchaeol (a marker for aquatic production) and the C:V ratio (manly affected by aquatic macrophytes; see above) do not reveal significant seasonal changes, we conclude that such increase in the concentration of the lignin phenols in the RW and FW seasons and the brGDGTs in the FW season is not derived from the water column, riverine SPOM or in situ production but from the soil and leaf runoff.

The two centimeters of surface sediment we have characterized in this study potentially integrate more than one year of sedimentation in such floodplain environment (Moreira-Turcq et al. 2004). However, because of the occurrence of pulsated inputs as well as resuspension, mixing and degradation processes in these superficial sediments (Moreira-Turcq et al. 2013), changes in the composition of superficial sediment apparently occurred at the seasonal scale (Figure 4 and Table 3). Indeed, The  $\lambda 8$  values showed significantly higher values in the RW and FW seasons than in the LW and HW seasons in all lakes (Figs. 4E, 4G, and 6A). The mean concentrations of brGDGTs also showed higher values in the FW season than in the HW season (Figs. 4H and 6B). The co-occurrence of these two types of molecules indicates that litter, traced by lignin phenols, and superficial soils, traced by brGDGTs, are preferentially deposited in the floodplain lakes during rising and receding waters. In addition, the seasonal mean values of (Ad:Al)v showed remarkably lower values in the RW and FW seasons (Fig. 4F), an inverse pattern if compared to the  $\lambda 8$  and brGDGTs. This suggests that less degraded lignin phenols were present in the surface sediments in the RW and FW seasons. Thus, in this case, the increase in the concentrations of the organic compounds was not a consequence of the re-suspension of the sediments, but due to a sudden arrival of fresher OM. In the HW and LW seasons, more degraded lignin phenols (i.e. higher values of the (Ad:Al)v) were present in sediments concomitant with lower amounts of  $\lambda 8$ . A possible process which is responsible for the  $\lambda 8$  and brGDGTs transfer to the lakes sediments is the connection of the Amazon River main stem with the local catchment areas such as wetlands and nonflooded forests during the RW and FW seasons. The lignin concentration could also increase as a consequence of the macrophyte communities while the brGDGTs could increase due to the in situ production in the floodplain lakes. However, the concentrations of crenarchaeol and IPL brGDGTs as well as C:V ratio do not reveal significant seasonal changes (Table 3 and Fig. 4). Based on these observations, we interpret that these changes in the lignin phenols in the RW and FW seasons and the brGDGTs in the FW season were not derived from the lake in situ production but from soil and leaf runoffs.

- P.20, Previous works postulated that Andean
- L.20 and low land soil material is mainly transferred to the lakes via river main stream, in particular, during the RW and HW seasons and that would be the main source of SOM of the floodplain lakes (e.g., Victoria et al.,1992; Moreira-Turcq et al., 2004; Mortillaro et al., 2011). However, according to our results, the lignin phenols increase their concentration in the RW and the FW seasons. Thus, based on the hydrodynamics of floodplain lakes and the concentration of the biomarkers applied in this study, in the RW and FW seasons, these organic molecules are mainly derived from the drainage of local wetlands soils. This is more evident for the upstream lakes, which are surrounded by flooded forests and by larger flooded area, than for the downstream lakes. which are surrounded mainly by grass vegetation and shrubs. However, even in lake Curuai, where the primary production and the riverine SPOM is admittedly an important source of SOM (Moreira-Turcq et al., 2004; Zocatelli et al., 2013), the interface between the floodplain lake and the flooded soil drives the sedimentation of the organic compounds.

Previous studies postulated that Andean and lowland soils are mainly transferred to the lakes via the Amazon River main stem, in particular, during the RW and HW seasons and that they would be the main source of SOM in the floodplain lakes (e.g., Victoria et al., 1992; Moreira-Turcq et al., 2004; Mortillaro et al., 2011). However, according to our results. the lignin phenols increased their concentration in the RW and the FW seasons. The hydrodynamics of floodplain lakes and their connections to the local drainage flooded forests and the main stem (Bourgoin et al., 2007) and the analusis of biomarkers applied in this study, suggest that in the RW and FW seasons, these organic molecules are mainly derived from the drainage of local wetlands and lowland "terra firme" soils. This is more evident for the upstream lakes surrounded by larger flooded forests than for the downstream lakes surrounded mainly by grass vegetation and shrubs. Even if in lake Curuai, phytoplankton primary production and the riverine SPOM are potentially important sources of SOM (Moreira-Turcq et al., 2004; Zocatelli et al., 2013), thia material is not predominant in the sediments, compared to the material coming from the interface between the lake and he wetland is determinant for the sedimentation of the organic compounds.

P.21, L.7	The vegetation coverage of the wetlands (flooded forests) are the most important source of SOM in floodplain lakes of the central Amazon basin. The macrophyte community in the floodplain lakes is also an important source of SOM whereas the river SPOM contributes to a minor fraction of it.	Our results suggest that the vegetation coverage of the wetlands (flooded forests) and "terra firme" (non-floodable forests) in the local catchment area of each lake investigated is the most important source of SOM in floodplain lakes of the central Amazon basin. The macrophyte community is also an important source of SOM whereas aquatic OM (i.e., riverine and lacustrine SPOM) contributes to a somewhat lesser extent.
P.21, L.15	The sedimentation of OC in the floodplain lakes are linked to the periodical floods.	The sedimentation of OM in the floodplain lakes are strongly linked to the periodic floods. The rain season (RW season), with substantially increased soil runoff, and the receding of waters (FW season), when OM

P. 22,

References added to the manuscript:

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

Hopmans, E.C., Schouten, S., Pancost, R.D., van der Meer, M.T.J., Sinninghe Damsté, J.S. Analysis of intact tetraether lipids in archaeal cell material and sediments by high performance liquid chromatography/atmospheric pressure chemical ionization mass spectrometry. Rapid Communications in Mass Spectrometry 14, 585– 589, 2000.

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. Persistence of soil organic matter as an ecosystem property. Nature 478, 49-56, 2011.

Sinninghe Damsté, J.S., Shouten, S., Hopmans, E.C., van Duin, A.C.T., Geenevasen, J.A.J. Crenarchaeol: The characteristic core glycerol dibiphytanyl glycerol tetraether membrane lipid of cosmopolitan pelagic crenarchaeota. Journal of Lipid Research 43,1641–1651, 2002.

Sinninghe-Damsté J.S., Verschuren D., Ossebaar, J., Blokker, J., van Houten, R., van de Meer, M., Plessen, B., Schouten, S. A 25,000-year record of climate-induced changes in lowland vegetation of eastern equatorial Africa revealed by the stable carbon-isotopic composition of fossil plant leaf waxes, Earth and Planetary Science Letters, 302, 236-246, 2011.

White, D.C., Davis, W.M., Nickels, J.S., King, J.D., Bobbie, R.J. Determination of the sedimentary microbial biomass by extractable lipid phosphate. Oecologia 40, 51–62, 1979. Responses to Referee #1 and #2

## Referee #1

## **General Comments:**

This study examines a suite of organic biomarkers and bulk chemistry in the surface sediments of the five major floodplain lakes in the central Amazon River during four seasonally distributed expeditions. The primary goal was to determine the relative contribution of upland (e.g. Andean) soils, flooded/non-flooded forests, macrophytes, and phytoplankton to floodplain sediments.

The authors conclude that the majority of floodplain sedimentary organic matter (SOM) is derived from flooded forests and aquatic macrophytes with minimal contributions from all other sources. The most convincing data are the C:V values observed for lignin phenols. The estimation that 20-30% of SOM is derived from macrophytes based on a simple mixing model is reasonable and based on established knowledge of endmember compositions. However, this is the only truly quantitative conclusion that can be made from this dataset as presented.

The other organic parameters measured are not adequate for quantifying the relative contribution of the desired OM sources beyond vague inference. For example, the authors somehow conclude that flooded forest vegetation is the primary source of SOM without any actual quantification of this source presented. The composition of lignin phenols cannot be used to differentiate between flooded versus non flooded forest vegetation/soil sources (or suspended POM for that matter) in this case.

Similarly, the authors estimate the contribution of phytoplankton based on the abundance of crenarchaeol. However, as the authors note, this compound is not produced solely by phytoplankton. Crenarchaeol has been found in nearly every type of environment (e.g. soils, sediments, rivers, lakes, and oceans), making any type of quantitative differentiation between endmembers dubious at best. Illustrating this point, the authors inconsistently state what crenarchaeol was used as a proxy for. For example, the abstract states it was used to identify river suspended POM, the introduction states that it was used to determine soil sources, and the results/discussion state that it was used to "indirectly" quantify aquatic production.

The other main conclusion made is that floodplain hydrodynamics seem to be the most important factor controlling SOM composition. Although this is probably true, the authors provide no discussion or data related to floodplain hydrodynamics. The only hydrologic data presented is discharge at Óbidos, which gives very little insight into the complex floodplain dynamics or possible drainages from the surrounding (non-flooded) landscape. A detailed modeling exercise would be required to adequately represent the complicated floodplain hydrodynamics and watershed inputs. Insights from the literature were not presented in this regard. Further, the collection of sediments at only 2-3 locations per floodplain lake does not provide a robust assessment of these environments, which the co-authors have reported as highly spatially heterogeneous in previous publications.

Overall the manuscript provides data for a collection of organic parameters that may be useful for other researchers in the region. Aside from the estimation of macrophyte contributions to SOM, very little quantitative conclusions are made, which greatly limits the potential impact of this work. The authors state many conclusions that appear to be inferred hypotheses at this point. The manuscript could be improved by describing the ambiguity of the measured parameters in greater detail and moderating/removing conclusions that are not quantitatively grounded. **Reply:** First of all, we would like to thank the referee #1 for the constructive comments. We believe that, after the corrections proposed by the referee #1, the quality of our manuscript will be greatly improved. More detailed rebuttals are provided below.

The principal objective of the present work is to quantify the fractions of the principal sources of sedimentary organic matter (SOM) in floodplain lakes of the central Amazon basin. It is expected that the sedimentation of OM in this ecosystem is linked to the periodic floods (e.g. Junk, 2010). In order to observe any possible changes in the spatiality and seasonality related to such link, we collected 57 surface sediment samples during four periods of the flood cycle (hydrological seasons) in the five major floodplain lakes (Figure 1). This sampling set provided approximately 12 samples per lake and 15 samples per season, collected in distinct sites of each lake. The number of samples was enough to make statistic comparisons (ANOVA) with the data found in the literature and to observe significant (p < 0.05) changes in the spatiality (upstreamdownstream) and seasonality. Differences among sites in each lake might be an important factor. To understand such differences, more detailed studies in each lake should be performed in future. For that purpose, a higher number of samples, including samples from flooded forest soils and non-floodable soils, are necessary. However, this is beyond the major purpose of this study. Here, we rather focused on the general role of the periodic floods on the SOM, by applying a mixing model.

The mathematical approach applied to quantify the sources of SOM was the three end-member modeling. Due to the complexity of the ecosystem and the behavior of each parameter in the environment, only the C:V ratio among the lignin parameters could be applied as an specific source (i.e. macrophytes). However, as observed in Figure 5, the riverine suspended particulate organic matter (SPOM) is a potential source of crenarchaeol to the SOM, since the soils have significantly lower amount of this compoundon. Unfortunately the data of crenarchaeol in the water column of the floodplain lakes are not available to this study. Consequently, it is impossible to disentangle the major crenarchaeol source between the riverine SPOM and the lacustrine SPOM based on the data available. Therefore, we will consistently use a more general term "the aquatic source" for crenarchaeol. It is unexpected that the primary production performed by the phytoplankton in the floodplain lakes is an expressive source of SOM, if compared to the others sources investigated in this work (e.q. Moreira-Turcq et al., 2013). Consequently, we will apply the crenarchaeol to quantify the fraction of aquatic OM or, in other words, as an indicator of the SOM derived from the aquatic environment (both river and lakes) in the revised version. Furthermore, we will make clear that we do not use crenarchaeol to estimate the contribution of phytoplankton since this compound is produced by Thaumarchaeota. On the other hand, it should also be noted that the abundance of crenarchaeol alone is not used to trace the soil input but the ratio of crenarchaeol and brGDGTs.

The rise and recede of waters in the floodplain lakes follow the river main channel, since they are connected. Thus, we believe that the data from Obidos are enough to illustrate the flood cycles and the periods when the sediments were sampled. Finally, we will provide information on the measured parameters in greater detail and will also moderate conclusions in the revised version as requested by the referee#1.

## **Specific Comments:**

- **Comment #1 (P4, L7):** "...the organic matter (OM) produced in the floodplain lakes fuels the out- gassing CO2 in the river system (Abril et al., 2014)."

There are several issues with this statement. First, the cited reference suggests that direct inputs of CO2 from (flooded) plant respiration is a significant source of CO2 to the system, not just the breakdown of OM derived from floodplain plants. Second, the cited reference suggests that the above floodplain CO2 sources are the primary source for CO2 in the central Amazon, not the entire Amazon system. Finally, the cited reference describes floodplains as a source for labile OM, such as lignin macromolecules that have been shown to be quite reactive, but fail to mention that the terrestrial (non-flooded) environment is also a large source of these types of molecules. "Terrestrial" carbon was only attributed to radiocarbon-depleted headland sources rather than vas- cular plants around the drainage basin. This statement should be moderated. For example, consider something along the lines of: "Further, inputs of CO2 from plant respiration and reactive OM produced in floodplain lakes is a significant source of CO2 outgassed in the Central Amazon River."

**Reply:** We agree with the Referee #1 for this point. We will add the sentence suggested by the Referee #1 as follows:

*"Further, inputs of CO<sub>2</sub> from plant respiration and reactive OM produced in floodplain lakes are significant sources of CO<sub>2</sub> outgassed in the Central Amazon River."* 

- **Comment #2 (P4, L22):** "...the contribution of the multiple sources of OM (up- land soils, flooded forest, aquatic macrophytes, and phytoplankton) <u>remain</u> uncertain"

**Reply:** We will change it as recommended by the referee as follows:

"...the contribution of the multiple sources of OM (up- land soils, flooded forest, aquatic macrophytes, and phytoplankton) <u>remains</u> uncertain"

- **Comment #3 (P4, L17):** describes that Suspended POM is primarily derived from forests and upstream soils. Why are forests (non-flooded) not mentioned as a potential source for sedimentary OM in floodplains? **Reply:** The major potential sources of OM are described in P14, L15, and the non-flooded forests are included among them as follows:

"To determine the origin of the SOM in the floodplain lakes, we considered five potential sources: (1) the terrestrial Andean clay-bounded and refractory SPOM, which may be transferred to the floodplain lakes via the Solimões-Amazonas and Madeira rivers (Hess et al., 2003), (2) "terra firme" soil and litter of the Amazonian lowland forest, which will be transferred to the floodplain lakes via streams, …"

We propose to make the necessary change in the text as follows: "To determine the origin of the SOM in the floodplain lakes, we considered five potential sources: (1) the terrestrial Andean clay-bounded and refractory SPOM, which may be transferred to the floodplain lakes via the Solimões-Amazonas and Madeira rivers (Hess et al., 2003), (2) "terra firme" soil and litter of the Amazonian lowland forest (non-floodable forests), which will be transferred to the floodplain lakes via streams,..."

- Comment #4 (P5, L15): "Lignin is a recalcitrant organic macromolecule. . ."

This statement is in conflict with the statement made at P5, L20: "but also a relevant source for the outgassing of CO2 in the Amazon River (Ward et al., 2013)." The cited reference showed that lignin can be very reactive in certain environments such as the Amazon River main stem near the mouth and more studies finding high rates of lignin turnover in other settings are emerging. The authors should consider the evolving philosophy on "recalcitrance/lability" vs. "reactivity" (Schmidt et al., 2011 Nature). Organic compounds are not intrinsically "labile" or "reactive" based only on chemical structure, but, rather, depend on the culmination of ecosystem properties.

**Reply:** We agree with the Referee #1 for this point. We will revised the aspect related to the recalcitrance of the lignin compounds in the revised version.

- **Comment #5 (P5, L21):** BrGDGTs and crenarcheol have been found to be not exclusively of soil origin in different environments around the world. Other potential sources should be described here as was done on P16, L10. The authors note in the discussion that these are not useful indicators for soil OM.

**Reply:** We will add more detailed information on the potential and diverse sources of brGDGTs and crenarcheol in the revised version.

- **Comment #6 (P6, L4):** ". . . provides new insights into the link between the hydrology of the Amazon basin to the sources of SOM in floodplain lakes."

It is not clear what linkages to hydrology were made here in this study. The only hydrologic data provided or discussed was discharge at Obidos during the study period with no discussion of the complex hydrology/hydrodynamics of floodplains. This statement is also made in the abstract (P3, L19) and in the conclusions.

**Rep**ly: The hydrological cycles in the floodplain lakes are strongly linked to the the Amazon River main channel. Therefore, we believe that the data from Obidos are sufficient to illustrate the flood cycles for the investigated areas and the periods when the sediments were sampled. Further, all the lakes in the present study are strongly flooded by the Solimões-Amazon river system in the RW and HW seasons (white waters) with minor contribution (in some cases) by black waters. This reinforces that the data reported in Óbidos are directly linked to the floodplain lakes. The necessary changes in Table 1 will be made to clarify this statement

- **Comment #7 (P7, L21):** Previous studies in these floodplain lakes describe immense spatial variability in biogeochemical characteristics. Do the authors feel that 2 to 3 sediment samples is a robust representation of these systems? Also it is not clear in the text where sampling stations were distributed.

**Reply:** We will provide more precise information on the sampling stations in the revised version. Although we sampled mostly at three sites in each lake for each period, we believe that our sampling site are representative for each lake with the most distinct regions of each lake: near the connecting channel, in the middle and near the floodable forests. This was sufficient to depict the major seasonal and spatial variations from upstream to downstream.

- **Comment #8 (P8, L19):** In order to assess contribution of inorganic nitrogen (NH+4 + NO-2 + NO-3) to TN, TN (wt. %) and TOC (wt. %) were correlated (R2 = 0.89; p < 0.001; n = 57)."

This is a confusing way to calculate inorganic nitrogen...please clarify. Also, does the calculated C:N ratio represent TOC to TON or TOC (i.e. TC in this study) to TN?

**Reply:** The results of TN were corrected by the factor found in the referred correlation (0.06 wt.%). Once the values of TON were determined, the C:N ratio was calculated. The sentence will be corrected as follows: "In order to assess contribution of inorganic nitrogen (NH+4 + NO-2 + NO-3) to TN, TN (wt. %) and TOC (wt. %) were correlated (R2 = 0.89; p < 0.001; n = 57). The interception of the trend line in the TN axis (0.06) was interpreted as the percentage of inorganic nitrogen. Thus, this correlation showed that....."

- **Comment #9 (P9, L1):** "Approximately 500 mg of freeze-dried sediments and macrophytes were ana- lyzed for lignin monomers using the alkaline CuO oxidation method"

This method is not for analyzing lignin monomers. The purpose of the CuO oxidation is to break apart macromolecules into monomers that can be analyzed, and, thus, represents the combination of macromolecules and monomers. Free lignin monomers typically make up less than 1% of the total lignin content.

**Reply:** We will modify this part in the revised version.

- **Comment #10 (P9, L12):** What type of detector was used on the gas chromatograph (e.g. GC-FID)?

**Reply:** We used the GC-FID. We will add this information in the revised version.

- **Comment #11 (P9, L13):** Please clarify whether the recovery standard was added before CuO oxidation/extraction or before analysis on the GC.

**Reply:** The recovery standard was added after the CuO oxidation following the methodology presented in the literature (Hedges and Ertel, 1982; Goni and Hedges, 1992). We will add this information in the revised version as follows: "The recovery factor was calculated using the internal standard ethyl vanillin added after the CuO oxidation and prior to analysis (values above 60 % were considered)."

- **Comment #12 (P12, L6):** "The C:N ratio did not reveal significant spatial and seasonal variations (Figs. 3b and 4b)"

Could this possibly be related to the fact that a "correlation" with TOC was used to calculate inorganic (and subsequently organic) N concentrations? Do the calculated C:N values represent real C:N values, or simply C:C (multiplied by some factor)?

**Reply:** We also looked at the C/N ratio without the correction of the inorganic nitrogen, which revealed the same pattern. Hence, we believe that it is not probable that the correction used for the TON could affect the results presented.

- **Comment #13 (P12, L11):** These are large ranges. It would be interesting to know the spatial distribution (e.g. where was -19 per mil and where was -29 per mil).

**Reply:** We will mention the spatial distribution of this parameter in the revised version.

- **Comment #14 (P15, L17):** "The averages of important lignin parameters (λ8, S : V ratio) but also the C : N ratio of the wood samples are significantly different from those for the sediments, which clearly indicates only a minor contribution of woody material to the SOM."

The authors should note that source signatures for lignin phenols are obscured by processes such as leaching, sorption, and biodegradation (e.g. Hernes et al. 2008, GRL and others). Vascular plant-derived OM will not have the same signature as a plant end-member after it has been mobilized into streams and altered by biological processes.

**Reply:** This is a good point. It will be incorporated the literature mentioned by the Referee#1 into the revised version. Indeed, this subject is discussed in the text based on the (Ad:Al)v results (P15, L21). The effects of degradation can alter the composition of lignin phenols of each source and also in the sediment samples, where the (Ad:Al)v values are high. However, we could not find an appropriate approach to quantify this process. Based on that, we considered an error of 10% in our estimations of each fraction.

- **Comment #15 (P16, L20):** "Crenarchaeol is, therefore, considered as an (indirect) indicator of aquatic primary production. The enhanced concentrations of crenarchaeol in SOM thus indicate a contribution from this source."

This relationship seems dubious, especially considering that in the introduction it was stated that crenarchaeol was used as a soil OM indicator and in the abstract it was stated that crenarchaeol was used as a suspended POM indicator.

**Reply:** The crenarchaeol (iGDGT) itself is not indicative of soil input as mentioned above. It is typically produced in the water column, but can also be produced in other compartments such as soils and sediments. In the present work we interpret, based on Figure 5 and in the percentage of IPL fraction (Table 3), that the main source of crenarchaeol to the sediments of the floodplain lakes is the riverine SPOM (no data from lacustrine SPOM available). Consequently, our interpretation is based on the fact that, most of the crenarchaeol found in the river SPOM was produced in situ (Zell et al. 2013). Since none of our proxies trace the plankton primary production, we cannot estimate its fraction in the SOM but the crenarchaeol can be an indicator of the OM derived from the river and lake waters.

- **Comment #16 (P17, L20):** "Consequently, the remaining 40–60 % of the SOM might be derived from other sources of OM such as the flooded forests (Eq. 3)"

There is no quantitative basis for this statement. You could just as easily say 40-60% might be derived from terra firme, headland, and/or SPOM sources. This sounds like a "guess."

**Reply:** Equations 1 and 2 estimate the fractions of macrophytes and SPOM. Based on that, the remaining fraction must come from other sources, namely flooded and non-flooded forests. We will revised this sentence in the revised version as follows:

"Consequently, the remaining 40–60 % of the SOM might be derived from the wetlands and non-flooded forests (Eq. 1). The periodic floods link the floodplain lakes and the wetland vegetation and soil. Thus, the seasonal and spatial contrasts in the SOM should be investigated in order to better understand the connectivity between these compartments."

- **Comment #17 (P17, Line 24):** "Thus, the seasonal and spatial contrasts in the SOM should be investigated in order to better understand the connectivity between these compartments."

Some insight from the authors on what else could be done would be appreciated. This study claims to address this and "provide new insights", but not many revealing trends were observed aside from the contribution of macrophytes. How can we improve on this?

**Reply:** The mixing model presented the relevance of the interface between the lakes and the surrounding wetlands and non-flooded forests. To understand how this interaction occurs we investigated the contrasts in the spatiality and in the seasonality. The only clear trend in the spatiality was the  $\delta^{13}C$  values (Figure 3C), which was further investigated by the molecular  $\delta^{13}C$  in n-alkanes (P19). It showed that such trend was caused by changes in  $C_4$  plants communities (macrophytes and grass vegetation), more abundant in downstream open lakes. Based on that, we concluded that the interaction between the river main stream and the lakes is not the most relevant factor for the composition of SOM, but the macrophyte population and the surrounding soils (wetlands and "terra firme"). This corroborates the results indebted in the mixing

model. Further, the seasonality showed significant changes in the  $\lambda 8$  and brGDGTs values. The higher concentration of these compounds in the FW season (more clear for the  $\lambda 8$  values) indicates that they are transported from the wetlands to the sediments. Finally, these observations, point to the conclusion that the river SPOM is not the main source of SOM, but the macrophytes and the local catchment area of each lake. This observation is presented as a general pattern for the central Amazon basin and not for some specific lake. Based on what is presented on previous works (e.g.: Victoria et al., 1992; Hedges et al.,1986; Mortillaro et al., 2011; Moreira-Turcq et al., 2013, Moreira et al., 2014) we understand that these conclusions are new insights concerning to this topic.

- **Comment #18 (P18, L8):** "Consequently, the bulk parameters apparently mix and homogenize the long time scale (year), while the biomarkers are more sensible to changes in short time scale (months) at the sediment surface."

How was this determined? All parameters were only measured 4 times over a 2 year period, not monthly.

**Reply:** The bulk parameters did not show any seasonal changes, which implies that these parameters do not change on these time scale (months). On the other hand some biomarkers do show significant change in these periods according to the periodical floods. Based on that we stated that in surface sediment samples, the bulk parameters cannot be applied to observe seasonal changes in the SOM but some biomarkers do so.."

- **Comment #19 (P20, L15):** "Since the concentration of crenarchaeol (a marker for aquatic production). . ."

The introduction states that crenarchaeol was/is typically used as a marker for soil OM. The abstract states crenarchaeol was used as an indicator for SPOM. Previously the authors mention that crenarchaeol can be found in nearly any type of environment.

**Reply #19:** It is not correct that crenarchaeol was/is typically used as a marker for soil OM since this compound is predominantly produced in the aquatic environment. Nonetheless, it is true that the crenarchaeol can be produced in various environments. According to the literature and our own results (Figure 5), the concentration of crenarchaeol in the SPOM of the Amazon River is significantly higher than any other compartment. Based on this observation, we interpreted that this compound is mainly produced in the rivers and consequently, this is the main source of crenarchaeol to the SPOM and thus SOM. Thus, we applied the crenarchaeol as an indicator of SPOM. We will modify the sentences which were written ambiguously as follows:

P5, L21: The ratio between brGDGTs and crenarchaeol have been applied to quantify the OC proportion originating from soils and aquatic environments (Hopmans et al., 2004; Herfort et al., 2006; Belicka and Harvey, 2009; Smith et al., 2010) and have recently been applied in rivers and floodplain lakes of the Amazon basin (Kim et al., 2012; Zell et al., 2013a; Moreira et al., 2014).

- **Comment #20 (P20, L16):** ". . .we conclude that such increase in the concentration of the lignin phenols in the RW and FW seasons and the brGDGTs in the FW season is not derived from the water column, riverine SPOM or in situ production but from the soil and leaf runoff."

How was this determined quantitatively, or is it just assumed/hypothesized?

**Reply:** The conclusions of the present work are based on the comparison of multiple biomarkers. This comparison consider the contrasts of each biomarker

according to the spatiality and the seasonality. Apart of it, the mean values of the sediment samples were compared to other compartments of the floodplains ecosystem (data found in the literature), in order to qualify and to quantify the sources of SOM. The C:V ratio and the crenarchaeol were the only biomarkers which clearly indicate specific sources of SOM (macrophytes and SPOM respectively). Consequently, they were applied to quantify the fraction of these two sources of SOM, according to the end member approach (Eq. 1 and 2). The other biomarkers could also indicate the sources of SOM but not quantitatively.

In terms of seasonality (which is linked to the periodic floods), the literature postulates that during the RW and HW the river transfer and deposits its SPOM into the lakes and this process is the main source of SOM. However, the crenarchaeol (the principal indicator of riverine SPOM) does not change seasonally. On the other hand, the  $\lambda 8$  and the brGDGTs do change. Both can be transported to the lakes from the river SPOM in the RW season, based on the hydrology of this ecosystem (Bouraoin et al.. 2007). In the FW water season, the only possible sources of these compounds are the sorrounding forests (wetland and "terra firme") in the catchment area of each lake. One can speculate that the macrophyte production of lignin and the in situ production of brGDGTs could cause such changes. If one considers this hypothesis, it is expected that the C:V ratio and the IPL brGDGTs would also increase in these same seasons, which was not observed (Figure 4D and Table 3). Thus, we interpret that in the RW season, the riverine SPOM and the soil runoff (as a consequence of the rain) are the causes of the increase in the  $\lambda 8$  values. In the FW season, the main source is the receding waters, which transfer the forest OM into the lakes. We will modify the sentences as follows:

P20: "The  $\lambda 8$  values showed significantly higher values in the RW and FW seasons (Figs. 4e, g, and 6a) in all lakes. The mean concentrations of brGDGTs also show higher values in the FW season (Figs. 4h and 6b) if compared to the HW season. The co-occurrence of these two types of molecules indicate that litter, traced by lignin phenols, and superficial soil, traced by brGDGTs, are preferentially deposited during rising and receding waters. Besides, the seasonal mean values of (Ad:Al)v show remarkably lower values in the RW and FW seasons (Fig. 4f), an inverse pattern if compared to the  $\lambda$ 8 and brGDGTs. This means that less degraded lignin is present in the surface sediments in the RW and FW seasons. Thus, the increase in the concentrations of the organic compounds is not a consequence of the re-suspension of the sediments, but to the arrival of fresher OM. In the HW and LW seasons, more degraded lignin phenols (higher values of (Ad : Al)v) are present in the sediments concomitant with lower amounts of  $\lambda 8$ . Consequently, the possible process via which the  $\lambda 8$  and brGDGTs can be transferred to the lakes sediments are the connection with the river main stream and with the local catchment area (wetland and "terra firme"). The liquin concentration could also increase as a consequence of the macrophyte communities, while the brGDGTs could increase due to the in situ production. However, the concentration of crenarchaeol, C : V ratio and the IPL brGDGTs do not reveal significant seasonal changes (Table 3 and Fig. 4). Based on these observations, we interpret that these changes in the lignin phenols in the RW and FW seasons and the brGDGTs in the FW season is not derived from the water column, riverine SPOM or in situ production but from soil and leaf runoff.

Previous works postulated that Andean and low land soil material is mainly transferred to the lakes via river main stream, in particular, during the RW and HW seasons and that would be the main source of SOM of the floodplain lakes (e.g., Victoria et al., 1992; Moreira-Turcq et al., 2004; Mortillaro et al., 2011). However,

according to our results, the lignin phenols increase their concentration in the RW and the FW seasons. Thus, based on the hydrodynamics of floodplain lakes (Bourgoin et al., 2007) and the comparison between the biomarkers applied in this study, in the RW and FW seasons, these organic molecules are mainly derived from the drainage of local wetlands and "terra firme" soils. This is more evident for the upstream lakes, which are surrounded by flooded forests and by larger flooded area, than for the downstream lakes, which are surrounded mainly by grass vegetation and shrubs. However, even in lake Curuai, where the primary production and the riverine SPOM is admittedly an important source of SOM (Moreira-Turcq et al., 2004; Zocatelli et al., 2013), the interface between the lake and the wetland and non-flooded forests are determinant for the sedimentation of the organic compounds."

- **Comment #21 (P20, L25):** "Thus, based on the hydrodynamics of floodplain lakes and the concentration of the biomarkers applied in this study, in the RW and FW seasons, these organic molecules are mainly derived from the drainage of local wetlands soils. "

This study did not include any assessment of "hydrodynamics." How was this conclusion reached?

**Reply:** The literature postulates that in the RW and in the HW seasons the flux of suspended material goes from the river main stream to the lakes. In FW season the flux goes in the opposite direction, from the flooded forests and lakes into the river. Finally, in the LW season, these interactions are very small (Bourgoin et al., 2007). Consequently, the increasing values in  $\lambda 8$  and brGDGTs, could be caused by the intense exchange of material between these compartments. As presented in Reply #20, after comparing these data with other biomarkers, we interpret that, the main cause for such seasonal changes are the interface between the lakes and the local wetland and non-flooded forests.

- **Comment #22 (P21, L2):** "However, even in lake Curuai, where the primary production and the riverine SPOM is admittedly an important source of SOM (Moreira-Turcq et al., 2004; Zocatelli et al., 2013), the interface between the floodplain lake and the flooded soil drives the sedimentation of the organic compounds." How was this conclusion quantitatively determined?

**Reply:** This statement could not be determined quantitatively. However, the mixing model results showed that the SPOM fraction is 20-30%, which indicates that the majority of the SOM does not come from this source. Further, the increasing values observed in  $\lambda 8$  (RW and FW seasons) and brGDGTs (FW season), and the comparison with other biomarkers, such as the C:V ratio, crenarchaeol and IPL brGDGTs, leads to the interpretation that the interface between the lakes and the local catchment area are determinant for the composition of the SOM. This process occurs in both upstream and downstream lakes since no spatial changes were observed for any biomarker, except the bulk  $\delta^{13}C$ .

- **Comment #23 (P21, L6):** "The vegetation coverage of the wetlands (flooded forests) are the most important source of SOM in floodplain lakes of the central Amazon basin. The macrophyte community in the floodplain lakes is also an important source of SOM whereas the river SPOM contributes to a minor fraction of it."

It is not clear how this conclusion was reached. The authors provide no quantitative index for flooded forests. Further any such index would be obscured by the contribution of terra firme forests. No modeling or spatial analysis was used to determine potential inputs from flooded forests vs. terra firme forests. The only endmember model that the authors provide is for macrophytes and phytoplankton and it is assumed that the remaining 40-60% of SOM is derived from flooded forest with no regard for SPOM or "terra firme" forests.

**Reply:** Both wetlands and non-flooded forests ("terra firme") can be relevant sources of SOM. We interpret that the wetlands are the most important among them, since it is nearer to the lakes and direct linked to them due to the periodic floods. However, the end member estimations are not conclusive about the fraction of each of these two sources to the SOM. We modified sentences as follows:

"The vegetation coverage of the wetlands (flooded forests) and "terra firme" (non-floodable forests), in the local catchment area of each floodplain lake, are the most important source of SOM in floodplain lakes of the central Amazon basin. The macrophyte community is also an important source of SOM whereas the river SPOM contributes to a minor fraction of it."

- **Comment #24 (P21, L15):** "The sedimentation of OC in the floodplain lakes are linked to the periodical floods."

Hydrology/hydrodynamics are not discussed beyond discharge at Óbidos. This study makes minimal, if any, connections between OM sources and hydrology as stated. Further, most results were reported to not vary "significantly" over space and time.

**Reply:** It has been widely reported in the literature the role of the periodic floods in the biogeochemistry and ecology of the floodplain lakes, in particular to the composition of the SOM (e.g., Hedges et al., 1986; Junk, 1997, Moreira-Turcq et al., 1013). The present work reinforces that the periodic floods are determinant for the sedimentation of OM in the lakes. However, we propose that the interface between the lakes and the local catchment area is more relevant than the interface between the lakes and the river main stream.

- **Comment #25 (P21, L19):** "Hence, together with wetland vegetation, the hydrodynamics of the flood- plain seems to be the most important controlling factor on the composition of SOM in the floodplain lakes of the central Amazon basin. See above comment.

**Reply:** The hydrodynamics of the floodplain lakes is driven by the periodic floods, which is well reported in the literature (e.g., Hedges et al., 1986; Junk, 1997, Moreira-Turcq et al., 1013, Bourgoin et al., 2007). The present work emphasizes that the periodic floods are determinant for the sedimentation of OM in the lakes. However, we propose that the interface between the lakes and the local catchment area is more relevant than the interface between the lakes and the river main stream.

# **Technical Corrections:**

-TC #1 (P4, L7): Capitalize "and"

**Reply**: We will correct it as follows: "The primary production is performed by the flooded forest, macrophytes, phytoplankton and periphyton (Junk et al., 2010) and further, the organic matter (OM) produced in the floodplain lakes fuels the outgassing  $CO_2$  in the river system (Abril et al., 2014). "

**-TC #2 (P6, L11):** "rivers" or "River" should be added after the river names (e.g. "Tapajós River") P6, L21: Capitalize "River"

**Reply:** We will correct it as follows:

P12, L11: "white water (e.g. Solimões, Madeira and Amazon rivers), black water (e.g. Negro River), and clear water (e.g. Tapajós River)."

P21, L3: "However, even in Curuai, where the primary production and the riverine SPOM is admittedly an important source of SOM..."

**-TC #3 (P7, L18):** It is unclear what the "CBM" code is referencing. Why not just refer to cruises as HW, LW, etc as was done in Figure 1 for better clarity?

**Reply:** The necessary changes will be made in the text as follows:

"The four hydrological seasons were targeted during different research cruises with a small research vessel (Fig. 2): in June and July 2009, which covered the High Water (HW) season; in October 2009 covering the Low Water (LW) season, in August 2010 covering the Falling Water (FW) season and in January 2011, which covered the Rising Water (RW) season. In each floodplain lake, sediment samples were collected at three stations in each season."

P8, L5: "Total carbon (TC), total nitrogen (TN), and  $\delta^{13}C$  for the samples obtained during the HW and LW cruises..."

P8, L9: "Four samples obtained during the FW and RW cruises were analysed...."

# -TC #4 (P8, L17): Delete "and"

**Reply:** We will delete it as follows: "TC (wt.%) correlated very well with TOC (wt. %) with a +0.16 intercept ( $R^2$ = 0.96; p < 0.001; n = 16)."

-TC #5 (P9, L18): Change "chromatography" to "chromatograph"

**Reply:** We will change it as follows: "To confirm the identification of each lignin phenol, eight selected samples were analyzed with an Agilent 7890A gas chromatograph coupled to an Agilent 5975C VL MSD mass spectrometer using a selective ion monitoring (SIM) at NIOZ (The Netherlands)."

**-TC #6 (P10, L25):** What brand/model HPLC-APCI-MS was used? P10, L9: Change "chromatography" to "chromatograph"

**Reply:** We will change it as follows: "The core lipids and IPL-derived GDGTs were analyzed using high performance liquid chromatograph-atmospheric pressure positive ion chemical ionization-mass 25 spectrometry (Agilent 1100 series LC/MSD SL, Alltech Prevail Cyano column ( $150 \times 2.1 \text{ mm}$ ;  $3\mu \text{m}$ )) in selected ion monitoring (SIM) mode according to Schouten et al. (2007)."

P11, L9: "The *n*-alkanes in the apolar fraction were identified by a Thermo Finnigan Trace DSQ gas chromatograph (GC-MS) and quantified with an HP 6890 GC system."

**-TC #7 (P10, L18):** It doesn't seem as though any statistical information has been reported in the results other than the number of samples. Perhaps p values should be reported.

**Reply:** We will report p values in the revised version.

-TC #8 (P11, L4): Change "value" to "values"

Reply: We will change it in the revised version.

**-TC #9 (P12, L19):** It should be noted that Lambda 8 is the "amount of lignin" normalized to OC.

**Reply:** We will add this information in the revised version as follows: "No significant changes were observed along the upstream-downstream transect for the mean values of  $\lambda 8$  (i.e. a proxy for the amount of lignin normalized to OC);..."

**-TC #10 (P12, L20) (and more):** The term "significant" was used 8 times in page 12, 6 times on page 13, 2 times on page 14, and 1 time on pages 15 and 16 with no statistical information given. Perhaps p values should be reported as was alluded to in the brief methods section (3.6). The use of "significant" is quite redundant.

**Reply:** The term "Significant" means p < 0.05 as described in session 3.6. We will report p values in the revised version.

**-TC #11 (P17, L7):** Add a space to "samples can" P17, Line 22: "Periodical" should be replaced with "periodic" here and elsewhere.

**Reply:** We will revise the sentence suggested by the referee#1 as follows: "According to this approach (Martinelli el al., 2003), the C : V values of macrophytes and the average values of soil and riverine SPOM sample scan be used to estimate the contribution of macrophyte OM to the SOM."

We will also revise the following sentences:

*P17*, *L22*: "The periodic floods link the floodplain lakes and the wetland vegetation and soil."

*P4, L8: "Periodic floods intensify the exchange of organic compounds, nutrients and minerals between rivers, lakes and flooded soils (Junk, 1997)."* 

*P21, L15: "The sedimentation of OC in the floodplain lakes are linked to the periodic floods..."* 

**-TC #12 (P19, L14):** "Accordingly, the percentage of C4 plants in the upstream lake is only 3 %, but for the downstream lake 22 %."

This should say "the contribution of C4 plants to SOM." It sounds as if the authors are stating that C4 plants only make up 3% of plant biomass in the region, when they are actually referring to the amount of C4-derived SOM.

**Reply:** We will correct it as suggested by the Refefee#1 as follows: "Accordingly, the fraction of  $C_4$  plants in the SOM in the upstream lake is only 3 %, but for the downstream lake 22 %."

-TC #13 (P20, L15): Change "manly" to "mainly"

**Reply:** We will corrected it as suggested by the Refefee#1 as follows: "Since the concentration of crenarchaeol (a marker for SPOM) and the C:V ratio (mainly affected by aquatic macrophytes; see above)..."

#### References:

*Reply:* We will add the following references in the revised version.

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.: Persistence of soil organic matter as an ecosystem property, Nature, 478, 49-56, 2011.

Bonnet, M.P., Barroux, G., Martinez, J.M., Syeler, F., Moreira-Turcq, P., Cochonneau, G., Melack, J.M., Boaventura, G., Maurice-Bougoin, L., León, J.G, Roux, E., Calmant,

S., Kosuth, P., Guyot, J.L., Seyler, P.: Floodplain hydrology in an Amazon floodplain lake (lago Grande de Curuai), Journal of Hydrology, 349, 18-30, 2008.

Bourgoin, L.M., Bonnet, M.P., Kosuth, P., Cochonneau, G., Moreira-Turcq, P., Guyot, J.L., Vauchel, P., Filizola, N., Seyler, P.: Temporal dynamics of water and sediment exchanges between the Curuai floodplain and the Amazon River, Brazil, Journal of Hydrology, 335, 140-156, 2007

## Referee#2

- **Comment #1:** Spatial heterogeneity within each lake: All lakes (except for Mirituba) are described as receiving water and sediments from multiple sources (i.e. white waters, black waters, clear waters), yet only 2-3 samples were collected for each lake in each season. There is no discussion as to the spatial variability of SOM within each lake for a given season, although this could have large impacts on the observed seasonal variability. For example, Moreira-Turcq et al., 2004 state that, "[sediment] fluxes were highly variable in space [within the Curuai lake], precluding extrapolation from a few measurements to a single value for the whole várzea." This heterogeneity was also reflected in %OC, C/N ratio, mineralogy, etc. The role of spatial heterogeneity within each lake should be addressed here.

**Reply:** The present work does not intend to evaluate the heterogeneity of each lake. For such, an effort, it would be necessary to examine a higher number of samples and a more detailed description of each lake, in terms of hydrological and physicchemical characteristics, as pointed out by referee #2. Instead, our aim was to understand the seasonal and spatial variations with respect to the sedimentary organic carbon in the floodplain lakes of the central Amazon basin. To this end, we selected three sampling sites in each of the five major floodplain lakes during four hydrological seasons. The selected sites were the most distinct regions of each lake: near the connecting channel, in the middle and near the floodable forests. As a result, we have approximately twelve samples for each lake for characterizing the spatiality along the transect from upstream to downstream, and we have approximately fifteen samples per season for characterizing the seasonality. In total, 57 sediment samples were analyzed in this study. This gives us a robust sampling set to investigate changes in spatiality and seasonality and to compare the results from the sediment samples with other compartments of the ecosystem and sources of OM, based on the statistics. In order to illustrate the variability in our data we presented the error bars in Figure 4 and all mean values used in the variance analyses (ANOVA) were presented in box plots. In the revised version we will make the aim of this study clearer.

- **Comments** #2: Sample collection and analysis: Naming conventions are not consistent throughout the manuscript. For example, the authors state that, "four riverbank sediments ... were also collected during the LW season," (p8753, line 24-25) however these samples are also referred to as "wetland soils" (e.g. in Table 2). Bulk carbon % and  $\delta$ 13C values reported are for raw samples – i.e. not decarbonated – although a subset of decarbonated samples resulted in similar carbon content with an offset of 0.16% ( $\delta$ 13C not compared). Assuming this 0.16% is inorganic carbon, this could explain ~0.8‰ of the observed downstream  $\delta$ 13C SOM enrichment. This is not a large difference (12% of the total observed), but should be addressed explicitly. Similarly,  $\delta$ 13C should therefore not be referred to as  $\delta$ 13Corg throughout the manuscript. For n-alkane quantification, peak areas should be calibrated against an external standard, with an internal standard only used for calculating extraction recovery. More detail should be given for GC-IRMS methods, such as column used, standard reproducibility, calibration method (i.e. using pulses of CO2 with known  $\delta$ 13C?), etc.

**Reply:** We thank the referee for spotting the inconsistencies of sample names and will correct them in the revised version of our manuscript. We will also provide more detailed description of the GC-IRMS method in the revised version. However, it is not clear for us why we should use an external standard if we use the internal standard, which is more common exercise in our field, for the quantification. - **Comment #3:** Reporting of results: Significant inconsistency exists between the results reported in the Results section of the main text and Tables 2-3, and tables / figures are mislabeled throughout the main text.

**Reply:** Thanks for spotting such mistakes. We will go through the text, figures and tables carefully and correct such mistakes thoroughly.

For example: Page 8758, Line 3: ". . . lower mean value (Table 2) in the downstream Lake Curuai," should refer to Table 3 and Lake Canaçari.

**Reply:** This will be corrected in the revised version as follows: "The TOC content was the lowest in the downstream lake Curuai ( $2.0 \pm 0.6$  wt. %) and the highest in lake Cabaliana ( $3.3 \pm 0.8$  wt. %) (Fig. 3a, Table 3)." ....

Page 8758, Line 6-7: "The lowest mean value was found in Curai (10  $\pm$  1) and the highest one in lake Mirituba (11  $\pm$  2)." These values are statistically identical.

**Reply:** This will be corrected in the revised version as follows: The sentence: "The lowest mean value was found in Curuai  $(10 \pm 1)$  and the highest one in Mirituba  $(11 \pm 2)$ " will be deleted.

Page 8758, Line 11: "Riverbank sediments" is referred to as "Wetland Soils" in Table 2. âA c Page 8758, Line 12: Table 3 should read Table 2.

**Reply:** This will be corrected in the revised version as follows: "The  $\delta^{13}C_{org}$  values in soils and wetland soils varied between -29 and -19 % (n = 7)..."

Page 8758, Line 15: "The C3 macrophytes ...  $\delta$ 13C values of -30‰'" The range reported in Table 2 is - 30‰ to -32‰

**Reply:** This will be corrected in the revised version as follows: "The  $C_3$  macrophytes (Eleocharis sp. and Pistia stratiotes) had  $\delta^{13}C_{org}$  values between -30% and -33% and values of C: N ratio between 15 and 24 (Table 2). ...."

Page 8758, Line 23: Fig. 3g should read Fig. 4g

**Reply:** This will be corrected in the revised version as follows: "...compared to the HW (23±9 mg  $g_{oc}^{-1}$ ) and LW (29±12 mg  $g_{oc}^{-1}$ ) seasons (Fig. 4g). ...."

Page 8758, Line 25 – Page 8759, Line 1: "The values of S:V ratio did not show significant spatial differences either but higher mean values in the RW season (1.1  $\pm$  0.1) and in the FW season (1.2  $\pm$  0.2) were observed in comparison to that of the LW season (0.9  $\pm$  0.1)." These values are statistically identical.

**Reply:** This will be corrected in the revised version:

Page 8759, Line 4-11: All numbers reported here are inconsistent with the values reported in Table 2. Again, "riverbank and wetland soils" is referred to only as "Wetland Soil" in Table 2.

**Reply:** This will be corrected in the revised version as follows: "For the  $C_3$  macrophytes,  $\lambda 8$  values varied between 26–67 mg g<sup>-1</sup> and between 48–94 mg g<sup>-1</sup> for the  $C_4$  macrophyte samples. The S : V ratio varied between 0.6 and 0.9 for  $C_3$  macrophytes and between 0.4 and 0.7 for the  $C_4$  macrophyte. The range of C : V ratio was 0.4 to 3.7 for the  $C_3$  macrophytes and 1.7 to 4.0 for the  $C_4$  macrophytes. The (Ad : Al)v ratio varied between 0.2 and 0.8 for all macrophyte samples (Table 3). For the "Terra Firme" soil and wetland soil samples, the  $\lambda 8$  values varied between 9 and 88 mg g<sup>-1</sup>. The S:V

ratio varied between 0.5 and 1.1, the C : V ratio varied between 0.2 and 0.5, and the (Ad : Al)v ratio varied between 0.6 and 1.5."...

Page 8759, Line 14-17: Unclear whether this is referring to core GDGTs, IPL GDGTs, or both. Values of crenarchaeol reported in the main text, ". . .higher in Canaçari (115  $\pm$  57 µg gOC-1) when compared to Janauaca (34  $\pm$  33 µg gOC-1)," are an order of magnitude higher than the values reported in Table 3.

**Reply:** This will be corrected in the revised version as follows: BrGDGT refers to the CL fraction and IPL brGDGts to the IPL fraction as presented in Table 3.

Page 8759, Line 26: ". . . mean values varied between 5  $\pm$  4. . ." while mean value reported in Table 3 is 4.

**Reply:** This will be corrected in the revised version as follows: "The mean values varied between  $4 \pm 4$  and  $10 \pm 6 \ \mu g \ g^{-1}$  in the HW and LW seasons, respectively."

Page 8760, Line 5-12: n-alkane results reported in Table 6 although Tables 4 and 5 have not been introduced yet. Tables should therefore be re-arranged for clarity.

**Reply:** We will rearrange the Tables according to the appearance in the text in the revised version as follows: "...and the C : N ratio values varied between 6 and 16 (n = 7; Table 2)." ....

Additionally, only average values are reported in Tables 2 and 3, while uncertainty is reported and interpreted in the main text. Analytical uncertainty should be reported in Table 2, and standard deviations about the mean values should be reported in Table 3.

**Reply:** We will report the uncertainties in Table 2 and 3 in the revised version as follows: "The mean concentrations of crenarchaeol were higher in Canaçari ( $12 \pm 6 \mu g g^{-1}$ ) when compared to Janauaca ( $4 \pm 3 \mu g g^{-1}$ )."

P8760 L.5-12: Rearrangement of table numbering:

**Reply:** We will rearrange the table numbers as follow:

Table 6 will become Table 4

Table 4 will become Table 5

Table 5 will become Table 6

Other changes in the text:

**Reply:** *We will correct them as follow:* 

P8761 L.12 - "...those of the SOM of the floodplain lakes in Fig. 5 and Table 5."

P8763 L.10 - "...the SOM (Eq. 1–3, Table 6)."

P8763 L.18 - "...the values of the other possible sources (Table 6)"

P8765 L.5 - "The results (Table 4) show that the long-chain n-alkanes  $\delta^{13}$ C signature..." P8765 L.13 - "...sediments of Janauaca and Curuai are listed in Table 4."
Table 2 - Analytical error will be reported

Table 3 - Standard deviation will be reported

- **Comment #4:** Interpretation of end members: One major concern is the inconsistency in interpre-tation of end members and the biomarkers used to infer them. For example, lignin is referred to both as a "recalcitrant organic macromolecule" as well as a "relevant source for the outgassing of CO2 from the Amazon River," implying that lignin is labile (p8751, line 15-20).

**Reply:** Although lignin is generally considered more recalcitrant in comparison to other organic compounds, recent studies also showed that this compounds can be degraded during the transport (Schmidt et al., 2011 Nature). Therefore, it is not inconsistency to interpret our data in this regard. However, we also see that the link between theses point is not well described in the current version and thus we will incorporate this point made by the referee in the revised version.

Discussion of lignin parameters (p8761, line 9 – p8762, line 4) does not discuss the fact that mixing of C:V, S:V and (Ad:Al)v is highly nonlinear between sources due to their variable  $\lambda$ 8 values.

**Reply:** We understand that this is a very pertinent comment about the nonlinearity of the lignin phenols in this approach. It will be properly discussed in the revised version.

Additionally, brGDGTs are said to track soil OM (p8749, line 10) as well as in situ production (p8762, line 10-13), while the authors state that, "riverine SPOM is the only possible OM source to explain a substantially increased concentration of crenarchaeol, in the SOM of the floodplain lakes if compared to other sources" (p8762, line 13-15). However, crenarchaeol is then used "as an (indirect) indicator of aquatic primary production." (p8762, line 20). Riverine SPOM is itself a complicated mixture of OC with highly variable contribution by phytoplankton production depending on the type of river (i.e. white, black, clear) and the water stage (e.g. Kim et al., 2012 GCA). Therefore, the simplification that crenarchaeol tracks riverine SPOM contribution used here should be refined.

**Reply:** In general, brGDGTs are mainly produced in soils, while crenarchaeol is predominantly produced in aquatic environments such as lakes and rivers. And thus brGDGTs can be used to trace soil OM input from land to the aquatic environment and crenarchaoel as an indicator for the aquatic production. Although this fact can be complicated since it turned out that brGDGTs are also produced in the aquatic environments and crenarchaeol in soils, it has been shown that detailed studies in a given area can give us detailed information from where these compounds are mainly originated and thus we can use them to trace the source of sedimentary OM. We will make this point clearer and try to avoid any inconsistency in the text in the revised version.

- **Comment #5:** Mixing model: The linear mixing model approach used here is underconstrained and nonlinear, and therefore invalid as presented. As an example of nonlinearity, a mixture of 50% macrophyte-derived OC and 50% riverine SPOMderived OC will bias toward the macrophyte end-member due to the contrasted lignin concentrations ( $\lambda$ 8 values) between these end members, resulting in a C:V of the mixture of ~1.6 rather than 0.75 if mixing was linear. Additionally, this model is inherently a 3 end-member mixing (rather than 2 as stated): macrophyte, aquatic, and "other" (also referred to as wetlands?). Thus, determining Fmacrophyte and Faquatic independently and solving for Fwetlands by difference is invalid, for example due to the influence of C:Vaquatic to the total C:VSOM which is not incorporated into the model as presented. Instead, this should be simultaneously solved as a system of 3 equations with 3 unknowns. Lastly, it is unclear how the authors chose end-member values for the "other" source or how they determined the uncertainty in the resulting fractional contributions.

**Reply:** To make possible the estimations of each fraction of SOM based on the end member approach we grouped the principal sources of SOM in Macrophytes, SPOM (riverine and lacustrine) and the surrounding forests (wetlands and "terra firme" soils, leaf, grass and wood). The calculation used only the biomarkers which were characteristics of one specific source. In this sense, the fraction of the calculated source was distinct from any other sources. For example, the C:V ratio indicates macrophytes, since its concentration in macrophyte samples are higher than any other source. Thus, C:V in the riverine SPOM does not affect the results. The same is expected for the calculations of SPOM based on the crenarchaeol. There is no specific biomarker to calculate the forest fraction. Thus, it was estimated based on the results of the two other fractions as follows:

 $F_{\text{macrophyte}} + F_{\text{SPOM}} + F_{\text{forest}} = \text{SOM} (100\%)$ 

$$F_{\text{SPOM}} = \frac{\text{Cren}_{\text{SOM}} - \text{Cren}_{(\text{forest+macrophyte})}}{\text{Cren}_{\text{SPOM}} - \text{Cren}_{(\text{forest+macrophyte})}} \times 100$$

$$F_{\text{macrophytes}} = \frac{\text{C:V}_{\text{SOM}} - \text{C:V}_{(\text{SPOM+forest})}}{\text{C:V}_{\text{macrophyte}} - \text{C:V}_{(\text{SPOM+forest})}} \times 100$$

The values of  $Cren_{(forest+macrophyte)}$  and  $C:V_{(SPOM+forest)}$  was determined on the average value of the respective biomarker in these sources. The data used to calculate these averages are presented on Table 4 and the averages on Table 5 as " $OC_{other}$ " (this will also be modified in the final version). Finally, we understand that present version of the formulas and the text in the manuscript were not clear about our mathematical background and our interpretation, thus we propose to do the necessary changes as it has been exemplified it the responses to referee #1 and #2.

The necessary modifications in the text will be done in the text as follows: "In Eqs. (2) and (3), the  $F_{SPOM}$  and  $F_{macrophytes}$  represent the estimated fractional abundance in SOM of macrophytes and SPOM, respectively. C:V<sub>SOM</sub> and Cren<sub>SOM</sub> are the average values of each parameter found in the sediment samples, C:V<sub>macrophytes</sub> and Cren<sub>SPOM</sub> are the values of the source of the respective parameter and C:V<sub>(SPOM+forest)</sub> and Cren<sub>(forest+macrophyte)</sub> are the values of the other possible sources (Table 5). These calculations indicate that 20–30 % of the SOM is derived from macrophytes and 20–30 % from. Consequently, the remaining 40–60 % of the SOM might be derived from the wetlands and non-flooded forests (Eq. 1). The periodical floods link the floodplain lakes and the wetland vegetation and soil. Thus, the seasonal and spatial contrasts in the SOM should be investigated in order to better understand the connectivity between these compartments."

The authors dedicate most of the results and discussion section to presenting data which show differences between lakes or seasons, i.e. increasing  $\delta 13C$  and decreasing %OC downstream, lower  $\lambda 8$  during LW and HW, lower brGDGTs during HW. However, none of these differences are incorporated into the mixing model presented here. There is no justification given for grouping all locations and seasons into a single mixing model despite their disparate bulk and biomarker values. In fact, this is contradictory to the observed downstream increase in bulk and n-alkane  $\delta 13C$ .

**Reply:** Even considering the spatial and seasonal variability for some biomarkers, which is not the case for crenarchaeol and C:V, the mixing model intends to compare the different compartments of the ecosystem and sources of SOM with the sediments. In this case, the seasonality and the spatiality should not be taken into account.

Additionally, the statement that, "bulk parameters apparently mix and homogenize the long time scale (year), while the biomarkers are more sensible to changes in short time scale (months) at the sediment surface," (p8764, line 7-10) is highly speculative and requires justification.

**Reply:** The bulk parameters did not show any seasonal changes, which implies that these parameters do not change on these time scale (months). On the other hand some biomarkers do show significant change in these periods according to the periodical floods. Based on that we stated that in surface sediment samples, the bulk parameters cannot be applied to observe seasonal changes in the SOM but some biomarkers do so.