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Input of particulate organic and dissolved inorganic carbon from the Amazon to the Atlantic Ocean

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[1] We report concentrations and isotope measurements (radiocarbon and stable carbon) of dissolved inorganic carbon (DIC) and suspended particulate organic carbon (POC) in waters collected from the mouth of the Amazon River and the North Brazil Current. Samples were collected in November 1991, when the Amazon hydrograph was at its annual minimum and the North Brazil Current had retroflected into the equatorial North Atlantic. The DIC $\Delta^{14}C$ results revealed postbomb carbon in river and ocean waters, with slightly higher values at the river mouth. The low DIC $\delta^{13}C$ signature of the river end-member (−11‰) demonstrates that about half of the DIC originated from the remineralization of terrestrially derived organic matter. A linear relationship between DIC and salinity indicates that DIC was mixed nearly conservatively in the transition zone from the river mouth to the open ocean, though there was a small amount ($\leq$10%) of organic matter remineralization in the mesohaline region. The POC $\Delta^{14}C$ values in the river mouth were markedly lower than those values from the western Amazon region (Hedges et al., 1986). We conclude that the dominant source of POC near the river mouth and in the inner Amazon plume during November 1991 was aged, resuspended material of significant terrestrial character derived from shelf sediments, while the outer plume contained mainly marine-derived POC.

Components: 3892 words, 4 figures, 1 table.

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Index Terms: 4805 Oceanography: Biological and Chemical: Biogeochemical cycles, processes, and modeling (0412, 0414, 0793, 1615, 4912); 4806 Oceanography: Biological and Chemical: Carbon cycling (0428); 4860 Oceanography: Biological and Chemical: Radioactivity and radioisotopes.

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1. Introduction

[2] The Amazon River accounts for 19% of global river water discharge and 10% of the river suspended load exported to the world ocean [*Milliman*, 1991]. According to *Richey et al.* [1990], the average export of particulate and dissolved organic carbon (POC and DOC) is 1.4 × 10^{13} g/yr and 2.2 × 10^{13} g/yr, respectively, which is ~10% of the global river input of organic carbon to the oceans. Thus the mixing of organic carbon in Amazon River water with seawater has a potentially large influence on the biogeochemistry of carbon in the Atlantic Ocean.

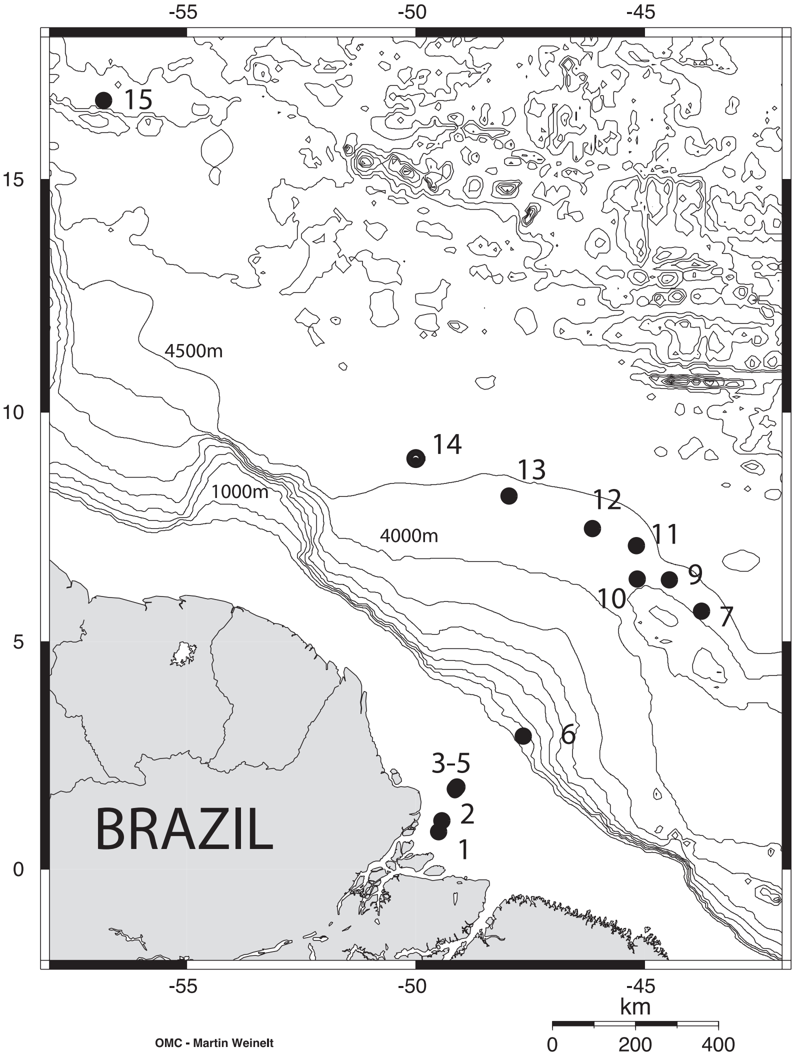

Figure 1. Map of station locations of samples collected during the November 1991 cruise. The predominant mean surface flow off Brazil during November is toward the northwest.

[3] As a part of CAMREX (Carbon in the Amazon River Experiment) [*Ertel et al.*, 1986; *Hedges et al.*, 1986, 1994; *Richey et al.*, 1990], *Hedges et al.* [1986] used Δ^{14}C measurements to show that coarse and fine suspended POC in the Amazon River basin cycled from terrestrial biomass through the watershed on decadal timescales. Several investigators have studied Amazon shelf sedimentation during the multidisciplinary Amazon Shelf Sediment Study (AmasSeds) and found mainly Holocene-aged sediments (<10 Ky) off the coast and north of the Amazon [*Nittrouer et al.*, 1996; *Sommerfield et al.*, 1996].

[4] In the present study, we reveal that POC at the mouth of the Amazon had lower Δ^{14}C values than those reported by *Hedges et al.* [1986], indicating resuspension of aged, sedimentary organic carbon near the river's mouth. The DIC concentrations and isotopic mixing curves reveal nearly conservative mixing in the Amazon plume region.

2. Collection and Analyses

[5] Our samples were collected at 14 sites on the final leg of the AmasSeds cruise from 26 November to 4 December 1991 aboard the R/V *Columbus Iselin* (see Figure 1). Surface waters ranged from a salinity of 0.40‰ at the river's mouth (station 1) to full open ocean values (35.01–35.21‰) at stations 6, 10 and 15 (Table 1). Brackish Amazon plume waters flow northwestward with the North Brazil Current and wind back to the southeast during the retroflection period [*Muller-Karger et al.*, 1988]; our stations 7, 9, 11, 12, 13 and 14 were in this mixing region. The vertical extent of the brackish plume averaged 19 m deep as deduced

Table 1. Isotope and Concentration Measurements of Suspended POC and DIC Collected During the Amazon Cruise in November 1991[a]

Stn	Btm Dep	Depth, m	Coll. Type	Salinity, ‰	*UCID*	[POC], μg/L	Δ^{14}C POC	Mean Δ14C	±, ‰	δ^{13}C POC	*UCID*	[DIC], μM	Δ^{14}C DIC	δ^{13}C DIC
1	<4m	0.5	pump	0.40	*448*		−145	−145	6	−25.6	*1778*	374	120	−11.0
2	10	0.5	Go-flo	4.67	*430*	628	−167	−164	5	−24.8	*1768*	563	100	−7.9
2					*1864*		−160			−25.8				
2					*1865*		−164			−25.9				
3	10	0.5	Go-flo	20.78	*527*	635	−251	−222	20	−25.0	*1770*	1386	103	−2.0
3					*1866*		−207			−25.2				
3					*1867*		−208			−25.3				
4	10	0.5	Go-flo	22.00	*445*	281	−201	−200	5	−24.2	*1765*	1427	99	−1.5
4		0.5			*1868*		−199			−25.4				
5	10	0.5	Go-flo	26.94	*446*	282	−184	−170	15	−23.6	*1773*	1718	110	−0.2
5					*1869*		−156			−24.2				
6	812	10.0	pump	35.01	*541*		82	82	5	−22.1	*1775*	2051	88	1.8
7	3197	100.0	Go-flo		*535*		−130		30	nd	*1776*	2179	56	
7a	3197	10.0	Go-flo	33.79	*529*	30				−21.2	*1766*	1955	63	0.7
9	4549	10.0	Go-flo	34.05	*483*	28	77	77	20	−21.5	*1771*	1983	86	1.7
10	4147	10.0	Go-flo	35.21							*1777*	2048	116	1.6
10		10.0									*1779*		94	1.8
11	4145	10.0	pump	33.78	*526*	37	13	13	5	−19.9				
12	4145	10.0	Go-flo	33.40	*484*	28	112	112	20		*1767*	1941	94	1.7
13	4145	100.0	Go-flo		*528*		−112		20	nd	*1772*	2185	51	
14	4145	10.0	Go-flo	32.64							*1769*	1911	91	1.6
15	4145	10.0	Go-flo	35.03	*447*	22	95	95	20	−19.9	*1764*	2014	89	2.0

[a] Samples from 100 meter depth are underlined, and laboratory identification numbers are italicized.

from periodic CTD profiles (W. Moore, personal communication). Samples collected at stations 1 through 5 were taken from 0.5 to 1 m depth and at the other stations were from 10 m depth. Water from 100 m depth was also collected at stations 7 and 13 to measure parameters below the plume.

[6] The ship was swabbed by Charlene Grall of the University of Miami Tritium Laboratory prior to loading our gear to ensure that contamination from tracer levels of ^{14}C and ^{3}H was not present in the labs or on deck. Dissolved organic and humic carbon concentrations and isotopic measurements will be reported separately (J. E. Bauer et al., manuscript in preparation). Twelve-liter, PVC Go-flo bottles were used to collect seawater for isotopic and concentration measurements of DIC. Prior to use, the 12-l Go-flo bottles were cleaned with 10% HCl, methanol and soaked in seawater. Each water sample was filtered using a precombusted (550°C) quartz fiber filter (142-mm diameter, Whatman ultrapure QM-A 0.8 μm nominal pore diameter) in a PVC holder attached directly to the bottle spigot. The 500 ml water samples for DIC were poisoned with 100 μl of saturated $HgCl_2$ solution, sealed with greased (Apiezon-N) ground-glass stoppers and stored at room temperature in the dark. Suspended POC was collected at stations 1, 6 and 11 (Table 1) using a deck-mounted pumping system to filter 10–110 liters of seawater through a 0.8-μm pore size, precombusted quartz-fiber filter housed in a 125mm diameter PVC filter holder. At all other sites, the entire contents of a 12-l Go-Flo bottle were filtered through the same filter type and housing. All filters were immediately frozen at −20°C in precombusted glass jars.

[7] Processing of DIC samples for carbon isotopic analyses was performed according to *McNichol et al.* [1994]. Briefly, a 250-ml sample was acidified with 85% phosphoric acid and nitrogen gas was used to sparge the sample of carbon dioxide, which was frozen into a trap with liquid nitrogen. Concentrations of DIC (in μmol/l) were determined in the laboratory of Catherine Goyet.

[8] For the POC concentration and carbon isotopic measurements, the filters were acidified with 1% H_3PO_4 in a double quartz tube, pumped dry, and combusted at 850°C with CuO and silver foil in vacuo for 1–2 hours [*Druffel et al.*, 1992]. The CO_2 from DIC and POC was reduced to graphite using hydrogen gas on cobalt catalyst at 560°C [*Vogel et al.*, 1987]. Radiocarbon was measured at the Center for AMS (Accelerator Mass Spectrometry) at Lawrence Livermore National Laboratory and reported as Δ^{14}C in per mil for geochemical samples with no age correction [*Stuiver and*

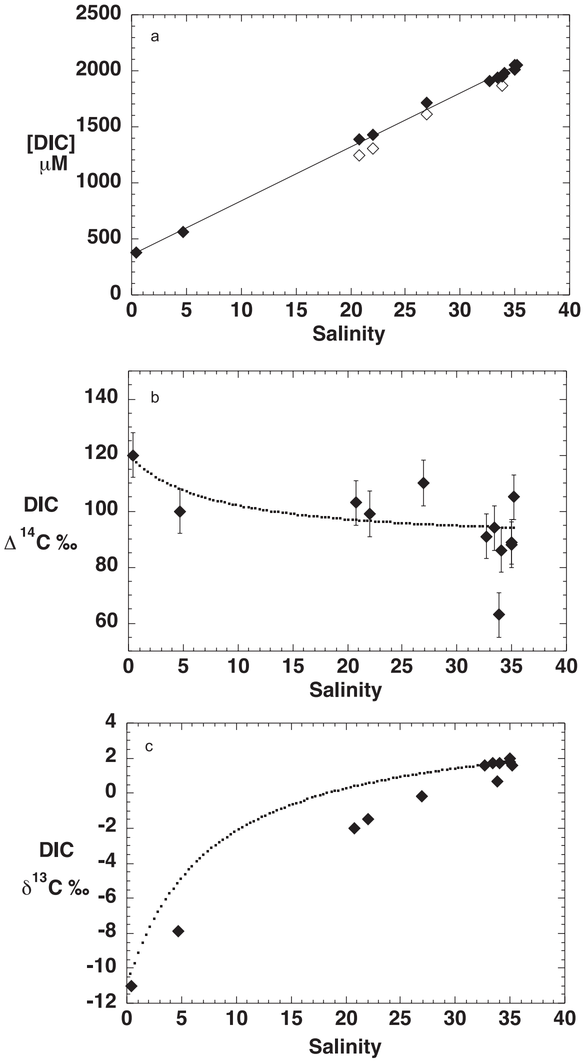

Figure 2. Measurements of salinity versus DIC (a) concentration (in μmol/l), (b) $\Delta^{14}C$ (in ‰), and (c) $\delta^{13}C$ (in ‰) for water samples collected from stations 1 through 15 as described in the text. The open diamonds in Figure 2a represent DIC concentrations corrected for DIC added from remineralization, as described in the text using $\delta^{13}C$ values. The lines represent conservative distributions for DIC (a) concentration, (b) $\Delta^{14}C$, and (c) $\delta^{13}C$ between river water and open ocean water.

Polach, 1977]. Total uncertainties for the $\Delta^{14}C$ measurements were ±10‰ for the DIC samples and as listed in Table 1 for the POC samples. Stable carbon isotope results ($\delta^{13}C$) were performed on splits of CO_2 gas at Woods Hole Oceanographic Institution with an uncertainty of ±0.1‰.

3. Results

[9] The DIC concentration measurements at 12 surface stations ranged from 374 μmol/l at the river mouth (station 1) to 2051 μmol/l at station 6 (Figure 2a). A significant, linear relationship (r = 0.999, line in Figure 2a) is obtained between surface DIC concentration and salinity, which suggests conservative mixing of DIC between river and open ocean waters during November 1991.

[10] The $\Delta^{14}C$ values of surface DIC ranged from a high of 120 ± 10‰ at the river mouth to an average value of 94 ± 10‰ in the three open ocean samples having salinity >35.00‰ (stations 6, 10, 15). The $\delta^{13}C$ values of surface DIC were lowest at the river mouth (−11‰) and highest in the three open ocean samples (average of 1.8‰). The DIC concentration values for the two samples at 100 m depth (2179 and 2185 μmol/l at stations 7 and 13, respectively, Table 1) were higher than in any of the surface water samples, and the DIC $\Delta^{14}C$ values (56‰ and 51‰, respectively) at 100 m depth were lower than surface values.

[11] Concentrations of suspended POC were highest in samples near the river mouth (628−635 μg/L at stations 2 and 3, respectively) and lowest in oceanic samples (22−37 μg/L at stations 7 through 15) (Figure 3a). A least squares fit of POC concentration vs. salinity shows significant inverse correlation (r = 0.90).

[12] The $\Delta^{14}C$ values of surface POC (Figure 3b) ranged from −145‰ at the river mouth, to lower values (−164 to −222‰) in the mesohaline region to a maximum value of 112‰ at station 12. The $\delta^{13}C$ values of surface POC were lowest at station 2 (−24.8 to −25.9‰) and highest at stations 11 and 15 (−19.9‰) (Table 1).

4. Discussion

4.1. Remineralization of Organic Carbon in the Amazon Plume

[13] The linear relationship between DIC concentration and salinity (Figure 2a) suggests that DIC was mixed conservatively across the entire salinity range during November 1991. The least squares fit extrapolated to a salinity value of 0‰ reveals a DIC concentration of 363 μmol/l.

[14] In contrast to DIC concentrations, isotopic signatures expected on the basis of conservative mixing alone must be corrected for variable concentrations of DIC at each site. Since DIC concentrations correlate linearly with salinity (Figure 2a), equation (1) is used to calculate

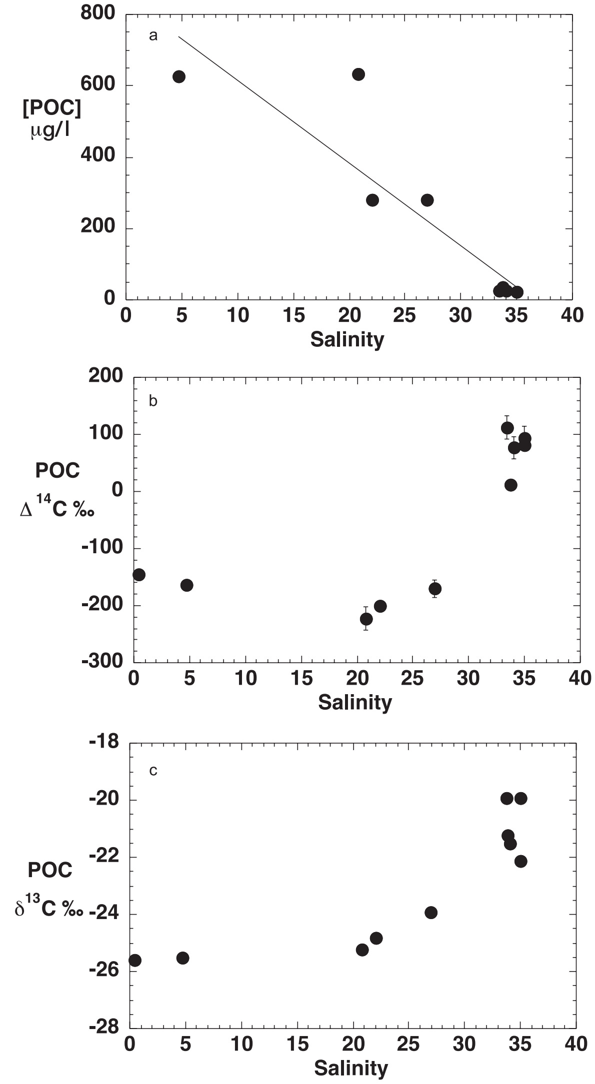

Figure 3. Measurements of salinity versus POC (a) concentration (µg/l), (b) Δ^{14}C (‰), and (c) δ^{13}C (‰) for samples from stations 1–15. There is no POC concentration measurement available at the river mouth station due to frequent clogging of the filters.

conservative mixing (CM) lines for Figures 2b and 2c:

$$X_{CM} = \frac{f_r \bullet X_r + f_o \bullet X_o \bullet [C]_o}{[C]_{CM}}, \tag{1}$$

where f_r and f_o are the fractions of water from the river (r) and ocean (o) ($f_r + f_o = 1$), X_r and X_o are the carbon isotope signatures (Δ^{14}C or δ^{13}C) of DIC in river (+120‰, −11‰) and ocean (94‰, 1.8‰) water, respectively, and [C] values are the DIC concentrations (line in Figure 2a). Though the gradient for DIC Δ^{14}C values between river and ocean water is small (Figure 2b), it appears that DIC Δ^{14}C was conserved in the Amazon mixing zone during November 1991.

[15] The DIC concentration and δ^{13}C value observed at the river mouth (374 µmol/l and −11‰, respectively) are comparable to the ranges (400–700 µmol/l and −16‰ to −12.5‰, respectively) observed at Obidos located 600 km upriver from station 1 [*Quay et al.*, 1992]. *Quay et al.* [1992] attributed the low DIC δ^{13}C values to remineralization of organic matter, where 40% was from C_3 plants (leaves and litter) and 60% from C_4 plants (grasses). High remineralization rates in Amazon River waters were also reported by *Richey et al.* [2002], who estimated that the net flux of CO_2 from the river to the atmosphere was 5×10^{14} gC yr^{-1}, an order of magnitude higher than the fluvial export of organic carbon to the ocean.

[16] The DIC δ^{13}C values in the mesohaline region are offset by 1.5 to 3‰ from the conservative mixing line with salinity and DIC concentration in Figures 2c and 4a, respectively. These low δ^{13}C signatures were likely caused by local remineralization of organic matter (POC and/or DOC) to produce additional DIC in the mixing zone. However the conservative relationship for DIC concentration (Figure 2a) limits the amount of DIC that could have been added by remineralization during mixing across the plume. For example, the 2.1‰ depletion of DIC δ^{13}C at station 4 (with respect to the conservative mixing line in Figure 4) would mean that 10% of the DIC, or 118 µmol/l, had been produced from POC (with a δ^{13}C = −24.8‰). By correcting the DIC δ^{13}C values in this way for stations 3–5 and 7a, we obtain an average offset of

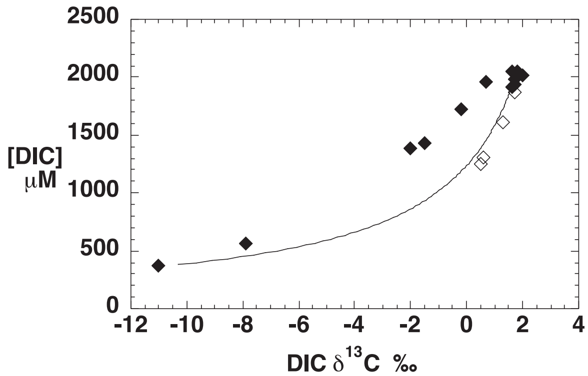

Figure 4. DIC concentration versus DIC δ^{13}C in surface waters from stations 1–15 (solid diamonds). The open diamonds represent DIC concentrations corrected for DIC added from remineralization, as described in the text using DIC δ^{13}C values. The line represents conservative distributions for DIC δ^{13}C between river water and open ocean water.

110 ± 22 μmol/l in DIC concentration (the unfilled diamonds in Figure 4) from the conservative mixing line. These "remineralization-corrected" DIC δ^{13}C values generally agree with the conservative mixing lines in Figures 2a and 4 and are consistent with the production of DIC from remineralization of organic matter in the mesohaline region of the Amazon plume.

[17] From the data presented here, we are unable to determine the source of the remineralized organic matter (e.g., POC or DOC) or the process(es) by which it was oxidized (e.g., photochemical oxidation and/or heterotrophic utilization). However, *Aller* [1998] described the "fluidized bed reactor" where significant terrestrial organic matter is remineralized within the sediments on the shelf. Thus the potential role of shelf sediments in contributing to the overall remineralization of Amazon-transported organic C may equal or exceed that in the plume water column.

4.2. Resuspension of Aged, Terrestrial Sedimentary Organic Carbon (SOC) in the Amazon Plume

[18] Conservative mixing does not apply to particles because solid phases are affected by gravity and flocculation. The AmasSeds Project showed that massive amounts of sediment are transported through the Amazon mouth and coastal shelf, forming fluid-mud deposits that are tidally cycled between the seafloor and water column. Tidal currents in excess of 1 meter/sec [*Curtin*, 1986; *Geyer et al.*, 1991] have been shown to rework the top 150 cm of Amazon shelf sediments [*Kuehl et al.*, 1986], leading to suspended sediment concentrations of several hundred mg/l at ten or more meters above the seabed [*Kineke et al.*, 1991] and tens of mg/l in surface waters [*Nittrouer et al.*, 1986].

[19] The δ^{13}C value for POC at the river mouth (−25.6‰) indicates that most of the POC was terrestrial in origin, though values were even lower for POC collected from the river mouth in May 1983 (−27.3‰ [*Showers and Angle*, 1986]). These results indicate that POC at the river mouth collected during our cruise contained significant carbon derived from marine sources (−20‰), which would be expected during the period of lowest river flow (November).

[20] The Δ^{14}C value of POC from the river mouth (−145‰) is much lower than the postbomb (>1950) values reported for fine and coarse POC (19 ± 19‰ to 227 ± 14‰, respectively) collected from the western Amazon (Rio Solimoes, 2°S, 66°W) from 1982−1984 [*Hedges et al.*, 1986]. This dissimilarity is due to enrichment of vascular plant debris and recent soils in POC at upriver locations. The Δ^{14}C value of surface SOC in the shelf region is −250 ± 30‰ [*Sommerfield et al.*, 1996], similar to the average POC Δ^{14}C value at our mesohaline stations (−197 ± 26‰). The isotope and concentration data reported here suggest that there was massive resuspension of aged, terrestrial OM on the Amazon shelf, which had been stored for variable amounts of time.

4.3. Implications for the Carbon Cycle in the Atlantic Ocean

[21] The Amazon River contributes a trivial amount of DIC to the North Atlantic (2.7×10^{13} gC/yr, <.05%) compared to the input of CO_2 from gas exchange with the atmosphere. In contrast, POC concentration in mesohaline plume water is 20 times higher than that in the open ocean (Table 1), making it a point source of POC to the North Brazil Current during plume retroflection. However, the dissimilarity of the isotopic compositions of the river and oceanic POC pools suggests that organic carbon in open ocean water is primarily modern, marine-derived plankton.

[22] The high annual flux of POC from the Amazon River (1.4×10^{13} gC/y [*Richey et al.*, 1990)] compared to the pool of suspended POC in the Atlantic implies that there is a significant sink (or sinks) for river-derived POC prior to its delivery to the open ocean. *Showers and Angle* [1986] estimated that the accumulation of OC in Amazon shelf surface sediments from terrestrial sources was 0.3×10^{13} g-TOC/yr. Our isotopic study indicates significant resuspension of SOC into the POC pool in the coastal region during November, the period of lowest water outflow. Our results also suggest that the POC near the river mouth contained significant amounts of marine-derived POC, which would be expected during the period of lowest river flow. The small amount of remineralization in the plume, as indicated by the DIC concentration and isotopic data, further suggests that the major fates of this POC is resedimentation or dispersal and dilution with oceanic waters in the outer plume region.

5. Conclusions

[23] Mixing of DIC was generally conserved in the Amazon plume during the low-flow retroflection

period, though low $\delta^{13}C$ values of DIC indicate that remineralization of organic matter in plume waters (salinity values 21–33‰) produced excess DIC approximately 4–10% above ambient levels. Aged, terrestrial POC was added to waters in the mixing zone from resuspended sediment that had low $\Delta^{14}C$. The rapid decrease of POC concentrations and rapid increase of POC $\Delta^{14}C$ values with distance from the river's mouth suggest that the old resuspended terrestrial material was removed by means (resedimentation, dispersal) other than remineralization in the outer plume and did not contribute significantly to the character of equatorial Atlantic POC.

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