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Molecular and isotopic constraints on the sources of suspended particulate organic carbon on the northwestern Atlantic margin

Jeomshik Hwang^{*,1}, Daniel Montluçon, Timothy I. Eglinton

Woods Hole Oceanographic Institution, Woods Hole, MA 02543, USA

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ABSTRACT

The abundance, carbon isotopic composition ($\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$), and lipid biomarker (alkenones and saturated fatty acids) distributions of suspended particulate organic matter were investigated at three stations centered on the 2000, 3000, and 3500 m isobaths over the New England slope in order to assess particulate carbon sources and dynamics in this highly productive and energetic region. Transmissometry profiles reveal that particle abundances exhibit considerable fine structure, with several distinct layers of elevated suspended particulate matter concentration at intermediate water depths in addition to the presence of a thick bottom nepheloid layer at each station. Excluding surface water samples, the $\Delta^{14}\text{C}$ values of particulate organic carbon (POC) indicated the presence of a pre-aged component in the suspended POC pool ($\Delta^{14}\text{C} < +38\text{‰}$). The $\Delta^{14}\text{C}$ values at the 3000 m station exhibited greater variability and generally were lower than those at the other two stations where the values decreased in a more systematic manner with increasing sampling depth. These lower $\Delta^{14}\text{C}$ values were consistent with higher relative abundances of terrigenous long-chain fatty acids at this station than at the other two stations. Two scenarios were considered regarding the potential provenances of laterally transported POC: cross-shelf transport of shelf sediment ($\Delta^{14}\text{C} = -140\text{‰}$) and along-slope transport of the slope sediment proximal to the sampling locations ($\Delta^{14}\text{C} = -260\text{‰}$). Depending on the scenario, isotopic mass balance calculations indicate allochthonous POC contributions ranging between 15% and 54% in the meso- and bathy-pelagic zone, with the highest proportions at the 3000 m station. Alkenone-derived temperatures recorded on suspended particles from surface waters closely matched in-situ temperatures at each station. However, alkenone-derived temperatures recorded on particles from the subsurface layer down to 250 m were lower than those of overlying surface waters, especially at the 3000 m station, implying supply of phytoplankton organic matter originally produced in cooler surface waters. AVHRR images and temperature profiles indicate that the stations were under the influence of a warm-core ring during the sampling period. The low alkenone-derived temperatures in the subsurface layer coupled with the lower $\Delta^{14}\text{C}$ values for the corresponding POC suggests supply of OC on resuspended sediments underlying cooler surface waters distal to the study area, possibly further north or west. Taken together, variations in $\Delta^{14}\text{C}$ values, terrigenous fatty acid abundances, and alkenone-derived temperatures among the stations suggest that input of laterally advected OC is a

* Corresponding author. Tel.: +82 10 8717 6587; fax: +82 54 279 8299.

E-mail address: jhwang@postech.ac.kr (J. Hwang).

¹ Present address: School of Environmental Science and Engineering, Pohang University of Science and Technology, Pohang 790-784, Republic of Korea.

prominent feature of POC dynamics on the NW Atlantic margin, and is spatially heterogeneous on a scale smaller than the distance between the stations (<150 km).

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1. Introduction

Continental margins are highly productive oceanic environments, globally accounting for approximately 10–20% of marine primary productivity and 90% of organic carbon (OC) burial (Hedges, 1992; Behrenfeld and Falkowski, 1997). The dynamics of particulate organic carbon (POC) over continental margins is highly complex with strong spatial and temporal heterogeneity due to gradients in surface ocean productivity, nutrient supply, and current regimes. There is also potential for export of OC from the margins to the deep ocean (Walsh et al., 1981; Churchill et al., 1988; Druffel and Williams, 1990; Bauer and Druffel, 1998). The literature is replete with evidence to indicate the widespread transfer of OC from the continental margins to the ocean interior. This evidence includes: (i) increased material fluxes in deep, relative to shallow sediment traps deployed near continental slopes (Honjo et al., 1982; Biscaye et al., 1988; Freudenthal et al., 2001; Smith et al., 2001); (ii) high suspended particle concentrations in the bottom nepheloid layer or intermediate nepheloid layers detached from the upper slope (McCave, 1983; McCave et al., 2001); (iii) the importance of lateral particle transport of resuspended sediments in simulations of bottom- and intermediate-nepheloid layers in a regional ocean model (Karakas et al., 2006); (iv) imbalances between carbon supply and oxygen consumption in sediments (Jahnke, 1996); (v) old ^{14}C ages of suspended and sinking POC (Druffel et al., 1998; Honda et al., 2000; Hwang et al., 2004); (vi) high aluminum contents of suspended particles (Sherrell et al., 1998); and (vii) the isotopic and molecular composition of organic matter in deep sea sediments (Benthien and Müller, 2000; Ohkouchi et al., 2002; Mollenhauer et al., 2006). Furthermore, the importance of long-range lateral transport of lithogenic material over and beyond the continental margin, known as hemipelagic deposition, in mineral input to the ocean interior has been well recognized (McCave and Tucholke, 1986; Rea and Hovan, 1995). Despite the evidence for widespread lateral transport, the magnitude of export, as well as the type of OC that is exported (i.e., as dissolved or particulate OC, and whether it is marine or terrestrial in origin), remains uncertain.

Redistribution of OC following initial deposition in margin sediments is important in determining the characteristics and quality of OC from several respects. It can re-expose OC to oxic degradation in the water column, enhancing biogeochemical transformations (Keil et al., 2004). Resuspension and across-shelf advection transports OC from more productive continental margins to ocean's interior (Walsh et al., 1981; Churchill et al., 1994; Karakas et al., 2006), and may be important in export of terrestrial OC to abyssal depths and sequestration of this carbon in slope and basin sediments. Remobilization and lateral transport is also an important consideration for

paleoclimate reconstruction from sediment since it can induce spatial and temporal effects between proxy records (Benthien and Müller, 2000; Ohkouchi et al., 2002; Mollenhauer et al., 2005).

The continental slope and rise of the NW Atlantic margin has been recognized as both a high productivity (Yoder et al., 2001; Mouw and Yoder, 2005) and high kinetic energy environment (Gardner and Sullivan, 1981; Hollister and Nowell, 1991). Both geological and physical oceanographic observations indicate that vigorous bottom currents are a common phenomenon in the region between the Gulf Stream and the Nova Scotian margin (Gardner and Sullivan, 1981; Hollister and McCave, 1984). These “benthic storms”, which appear to be associated with the southward flowing deep western boundary current (DWBC), may scour, resuspend and laterally transport sedimentary particles over the slope. Remobilized particles may be entrained in the DWBC and settle out along its path when currents weaken or may be transported basinward through interactions between the DWBC and the deep recirculation gyres in the interior Atlantic basin where they can ultimately settle (McCave and Tucholke, 1986). Although lateral transport of particles from the northwest Atlantic margin to areas of accumulation in the deep Atlantic such as the Bermuda Rise has long been recognized, it has only been recently recognized that organic matter can also survive long-range transport to an extent that it can dominate carbon burial in pelagic sediments (Ohkouchi et al., 2002, submitted). Accumulation of allochthonous OC in Bermuda Rise sediments was revealed by the ^{14}C ages and composition of source-specific biomarker lipids. Notably, alkenones – unsaturated long-chain ketones derived from haptophyte algae – were up to 7000 ^{14}C -years older than planktonic foraminifera isolated from the same sediment horizon (Ohkouchi et al., 2002). Since both alkenones and planktonic forams grow in the surface water, they should have identical ages. The age offset thus indicates that formation of the algal biomarker must substantially predate that of the foram, and this observation can be explained by aging due to temporary storage in distal margin sediments or during transport. Moreover, measurements of the alkenone unsaturation index (U_{37}^k ; Prahl and Wakeham, 1987) suggested that alkenones were produced in colder surface waters than those overlying the Bermuda Rise (Ohkouchi et al., 2002). Stable hydrogen isotope (D) analysis of alkenones in Bermuda Rise sediments suggests that they were derived prominently from subpolar waters to the northwest or north of the drift (i.e., the Scotian Margin) (Englebrecht and Sachs, 2005). Together with available sedimentological information, the preponderance of evidence suggests supply of organic matter originating from the productive waters over the Northwest Atlantic margin.

The physical oceanography of the New England slope region has been, and continues to be intensively studied in efforts to understand the Atlantic meridional overturning circulation, and in particular the role and dynamics of the DWBC, its interaction with the Gulf Stream, and its response to high-latitude climate forcing (e.g., Joyce et al., 2005). At present, activities are focused on a transect between Woods Hole and Bermuda (“Line W”; <http://www.whoi.edu/science/PO/linew/index.htm>). To further our understanding of POC dynamics in the NW Atlantic, we have initiated a study of biogeochemical characteristics of suspended and sinking particulate matter and surface sediments in the vicinity of this physical oceanographic observation program. In this paper, we discuss the biogeochemical properties of suspended particles recovered from different water depths at three stations over the New England slope. Bulk (radiocarbon and stable carbon isotope ratios) and molecular (the alkenone unsaturation index and saturated fatty acid chain length distribution) signatures were used to assess the provenance of POC in continental slope waters.

2. Methods

Suspended POC samples were collected at three locations centered on the 2000 m (39°45N, 69°44W, Station A), 3000 m (39°27N, 68°22W, Station B), and 3500 m (38°36N, 68°50W, Station C) isobaths on the New England slope during a cruise aboard the R/V *Endeavor* in

early summer 2005 (25 June–2 July) (Fig. 1). Sampling sites were aligned with the “Line W” physical oceanographic mooring array that provides hydrographic context for the samples collected and also an assessment of the dynamics of the particle field in the vicinity of the DWBC (Fig. 1). Between 700 and 1700 l of seawater were filtered using submersible pump systems (McLane Research Laboratories Inc.) equipped with pre-combusted 142 mm diameter glass fiber filters (Whatman GF/F). Samples were collected from 10 to 12 depths at each station. The volume of pumped water was determined with the flow meter of each filtration system. Particles in the surface water were also collected from the ship’s uncontaminated seawater intake using 293 mm diameter filters (GelmanScience, type A/E glass fiber). Each filter was folded and stored frozen at -20°C in a pre-combusted aluminum foil pouch until analysis. A quarter of each filter was freeze-dried and used for quantification of POC abundance and carbon isotope analyses. This portion of each filter was decarbonated (in either a combusted 140 mm ID Petri-dish covered with a watch glass, or in a desiccator) using concentrated HCl vapor for 12 h or overnight at room temperature (Hedges and Stern, 1984). Each filter was packed into double quartz tubes with CuO and silver wires, and subsequently the tubes were evacuated, flame-sealed, and combusted at 850°C for 5 h. The resulting CO_2 gas was cryogenically purified, and then quantified manometrically. A procedural blank obtained by processing a quarter of a pre-combusted 142 mm GF/F filter by the same method was $3.3\ \mu\text{gC}$ ($n=3$), corresponding to less than 2% (0.9% on the average) of the

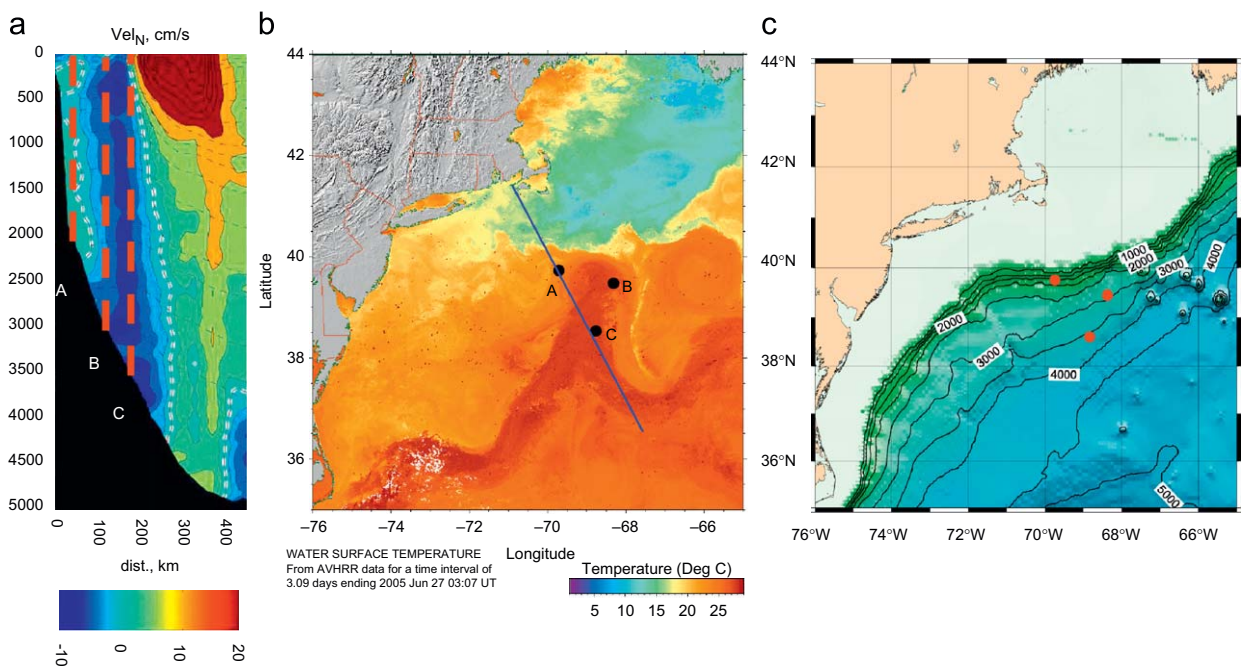


Fig. 1. (a) Average normal velocity (reproduced from Joyce et al., 2005) along a transect shown on (b). The vertical straight lines indicate positions of 2000, 3000, and 3500 m stations (A, B, and C, respectively) occupied for suspended particle sampling. (b) Satellite-measured SST, and (c) Bathymetry and sampling locations on the New England slope in the northwest Atlantic. The AVHRR image was obtained from the Ocean remote sensing group at Johns Hopkins University Applied Physics laboratory (<http://fermi.jhuapl.edu/avhrr/>) and represents a three day composite ending 27 June 2005. It shows the moment when the Gulf Stream engulfs a warm-core ring produced in mid- to late-May.

sample carbon yields. Carbon isotopic measurements were made at the National Ocean Sciences Accelerator Mass Spectrometry facility at Woods Hole Oceanographic Institution using standard techniques (McNichol et al., 1994). The $\Delta^{14}\text{C}$ values were not blank-corrected. In the worst case scenario, incorporation of ^{14}C -dead carbon blank may cause an up to 20‰ decrease in $\Delta^{14}\text{C}$ for a recently produced POC sample. The uncertainty for $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ measurements for this type of samples based on multiple duplicate analyses is smaller than $\pm 10\%$ and $\pm 0.1\%$, respectively.

For lipid biomarker analyses, one half of each filter was subjected to accelerated solvent extraction (DIONEX, ASE200) using a dichloromethane:methanol (93:7 by volume) mixture. Solvent was removed from the resulting total lipid extract by blowing nitrogen gas gently over the samples. The dried extracts were saponified in ~ 10 ml of 6% KOH in methanol at 80°C for 2 h. Roughly the same volume of 10% NaCl solution was then added to the mixture, and a neutral lipid fraction was extracted into a hexane phase at least 3 times. The separated and combined hexane extracts were rinsed twice with 10% NaCl solution (~ 5 ml) to remove water soluble components and any remaining water was removed with anhydrous Na_2SO_4 . A ketone fraction was obtained from the neutral fraction by eluting through a glass column filled with activated silica-gel (1 g) with dichloromethane after hydrocarbons were pre-eluted with hexane. The ketone fraction was injected into a HP-5890II gas chromatograph equipped with either 60 or 30 m capillary column (Varian, CP-SIL 5CB, 30 m \times 0.25 mm ID \times 0.25 μm film thickness) and a flame ionization detector for determination of alkenone abundances and unsaturation index (U_{37}^k ; Prahl and Wakeham, 1987). Alkenone temperatures for suspended POC and core-top sediment samples were calculated according to the corresponding alkenone temperature calibrations by Conte et al. (2006). The uncertainty for alkenone temperature is smaller than $\pm 2^\circ\text{C}$, which includes the mean standard error of estimation of 1.2°C for temperature calibration (Conte et al., 2006). Following the isolation of the neutral fraction from the saponified samples, the aqueous phases were acidified (pH < 1) with concentrated HCl and an acidic fraction was obtained by partitioning into an apolar phase (hexane:DCM, 4:1 v/v, at least 3 times). The solvent was completely removed and the residue was transesterified in 5% HCl/methanol at 70°C for about 12 h (or overnight). Fatty acids were quantified as fatty acid methyl esters (FAMES) with the same GC/FID equipped with a 30 m capillary column (Varian, CP-SIL 5CB, 30 m \times 0.25 mm ID \times 0.25 μm film thickness). The FAMES were identified using a Supelco FAME standard. Recovery efficiencies of the extraction process (by addition of an internal standard) were not determined because of potential subsequent use of the samples for carbon isotopic analysis. Hence, the concentration data should only be regarded as semi-quantitative. However, the percentage of long-chain fatty acids to total fatty acids (see Section 3.5), a relative property, has a much smaller uncertainty than the concentration due to the high degree of internal GC reproducibility.

Transmittance data for assessment of particle abundances were acquired in-situ with a transmissometer (C-STAR, WET Labs Inc.; 25 cm path length, 660 nm), along with fluorescence and other physical parameters using a CTD-Rosette system. Beam attenuation coefficient is a measure of the total cross-sectional area of particles in the beam (Van de Hulst, 1957), and was calculated using calibration parameters provided by the company: $c = -\ln[(19.7790 \times \text{Voltage} - 1.1828)/100]/0.25$.

3. Results

3.1. Temperature, salinity, and abundances of suspended particulate matter and organic carbon

Station A had lower temperature and salinity values than the other two stations, which had similar temperature and salinity profiles extending down to ~ 800 m (Fig. 2). The surface mixed layer depth ranged from 10 m at Station A to 50 m at Station C. The vertical profiles of temperature and salinity reveal distinct fine structure, and imply mixing of different water masses in the upper 500 m. An anomaly centered in temperature and salinity at ~ 320 m is particularly prominent at Station C, and implies an intrusion of colder, less-saline water at that depth (see further discussion in Section 4.1).

Transmissometry profiles also reveal considerable fine structure in water column particle abundances with depth (Fig. 2c and 3a). At each station, beam attenuation decreased rapidly from the surface to 100–150 m; this attenuation in the upper 150 m layer was greatest at the shallowest station (A). Distinct layers of higher beam attenuation (i.e., elevated concentrations of suspended particulate matter) were observed in the upper 1000 m, although discernable lenses of higher beam attenuation were also observed deeper in the water column. These intermediate-depth nepheloid layers (INLs) were most pronounced between 200 and 600 m at the most offshore station (C). Bottom nepheloid layers (BNLs) of 200–400 m thickness were evident by increases in beam attenuation towards the seafloor, and again were most prominent at Station C.

In general, vertical profiles of POC abundance determined by in-situ filtration paralleled those of beam attenuation. POC abundance gradually decreased with increasing depth (Fig. 3b; Table 1) with the highest POC concentrations (15.5 $\mu\text{gC/L}$) observed at the shallowest depth. POC abundances below 500 m ranged from 0.7 to 2.2 $\mu\text{gC/L}$. Although differences in POC abundance exist among stations, these discrepancies were not apparent below 500 m. A common feature at all stations was a higher abundance of POC near the sea floor that extended a few hundred meters above the bottom (mab), and these layers were coincident with the region of enhanced beam attenuation (i.e., BNL). The bottom water maxima underlay an apparent POC minimum between 500 and 1000 mab at each station.

Comparison between beam attenuation coefficients, averaged over 40 m intervals, and POC abundance from corresponding depths, yielded a significant correlation

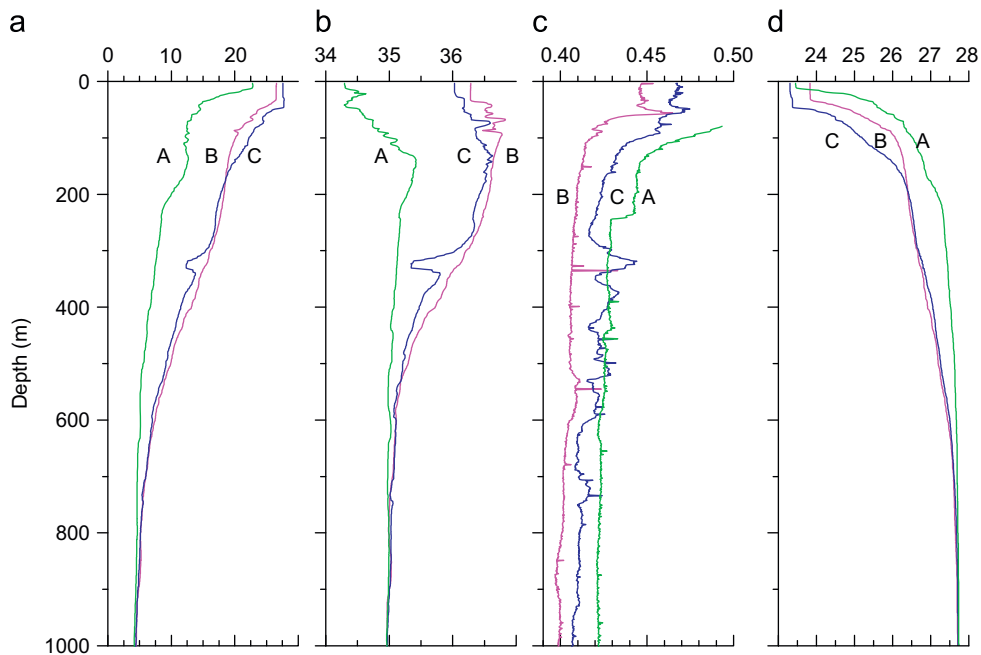


Fig. 2. (a) Temperature in °C, (b) salinity, (c) beam attenuation coefficient, and (d) σ_t shown only in the upper 1000 m. Values for temperature, salinity, and σ_t at each station converge and remain virtually identical below this depth. Complete vertical profiles of beam attenuation coefficients are shown in Fig. 3a. The capital letters on the plots denote station names.

(p -value $< 10^{-10}$, Fig. 4). The overall regression slope (beam attenuation coefficient vs. POC abundance in nmol/L = 2×10^{-4} , and 1.3×10^{-4} when 40 m result was excluded) at the three stations was steeper than those reported in the NE Pacific and in the Equatorial Pacific waters [0.6×10^{-4} , Bishop (1999); Bishop et al. (1999)]. This may have been caused by different optical properties of freshly produced particles and resuspended sediment (Pilskaln, 2006), and high uncertainty for POC determination due to cracks occasionally observed around the periphery of filters upon their recovery.

3.2. $\Delta^{14}\text{C}$ values

The $\Delta^{14}\text{C}$ value of POC collected from the ship's uncontaminated seawater intake at Station B was 67‰ (Fig. 3c). This value is within the range of the observed surface water DIC (dissolved inorganic carbon) $\Delta^{14}\text{C}$ values, between +40‰ and +80‰, in the northwestern Atlantic margin and the Sargasso Sea (Druffel et al., 1992; Bauer et al., 2002). The $\Delta^{14}\text{C}$ value of a suspended POC sample from this station at 40 m depth was 38‰, and the values at all the other depths were lower than this value. This indicates the presence of ^{14}C -depleted OC, of which some fraction pre-dated atmospheric nuclear weapons testing in the middle of the last century. At Stations A and C, the $\Delta^{14}\text{C}$ values gradually decreased with increasing depth with the exception of a more abrupt decrease near the bottom, coincident with the BNL. In contrast, the $\Delta^{14}\text{C}$ values at Station B were highly variable

throughout the water column. At most depths, the $\Delta^{14}\text{C}$ values were much lower at Station B than those at the other stations, with the lower $\Delta^{14}\text{C}$ values observed in the water column at Station B comparable to those observed for 20–50 mab samples in the BNLs at this and the other stations.

Bauer et al. (2002) reported $\Delta^{14}\text{C}$ values as low as -285 ‰ for suspended POC in deep slope waters near our study area ($39^{\circ}49.77'\text{N}$, $70^{\circ}20.781'\text{W}$, water depth = 1200 m), as well as slope waters along the Mid-Atlantic Bight. In this case, POC was collected by filtration of much smaller volumes of water (<30 L) and therefore might carry higher carbon blanks (e.g., from dissolved OC sorbed onto the filter; Liu et al., 2005). Notably, the lowest value observed in our study is significantly higher (-80 ‰). This apparent temporal and spatial variability in $\Delta^{14}\text{C}$ values of suspended POC clearly requires further investigation.

3.3. $\delta^{13}\text{C}$ values

The $\delta^{13}\text{C}$ values of suspended POC ranged between -20 ‰ and -22.5 ‰ (Fig. 3d; Table 1). The values did not vary with depth at each station in a systematic manner, although there is a hint of an increase in the BNLs. The values were slightly lower at Station B than at the other stations (on the average, by ~ 0.4 ‰ without considering the 40 m datum). The lowest value was observed at 40 m depth. In general, the range of $\delta^{13}\text{C}$ values and the shape of the vertical profiles – the lowest values near the surface

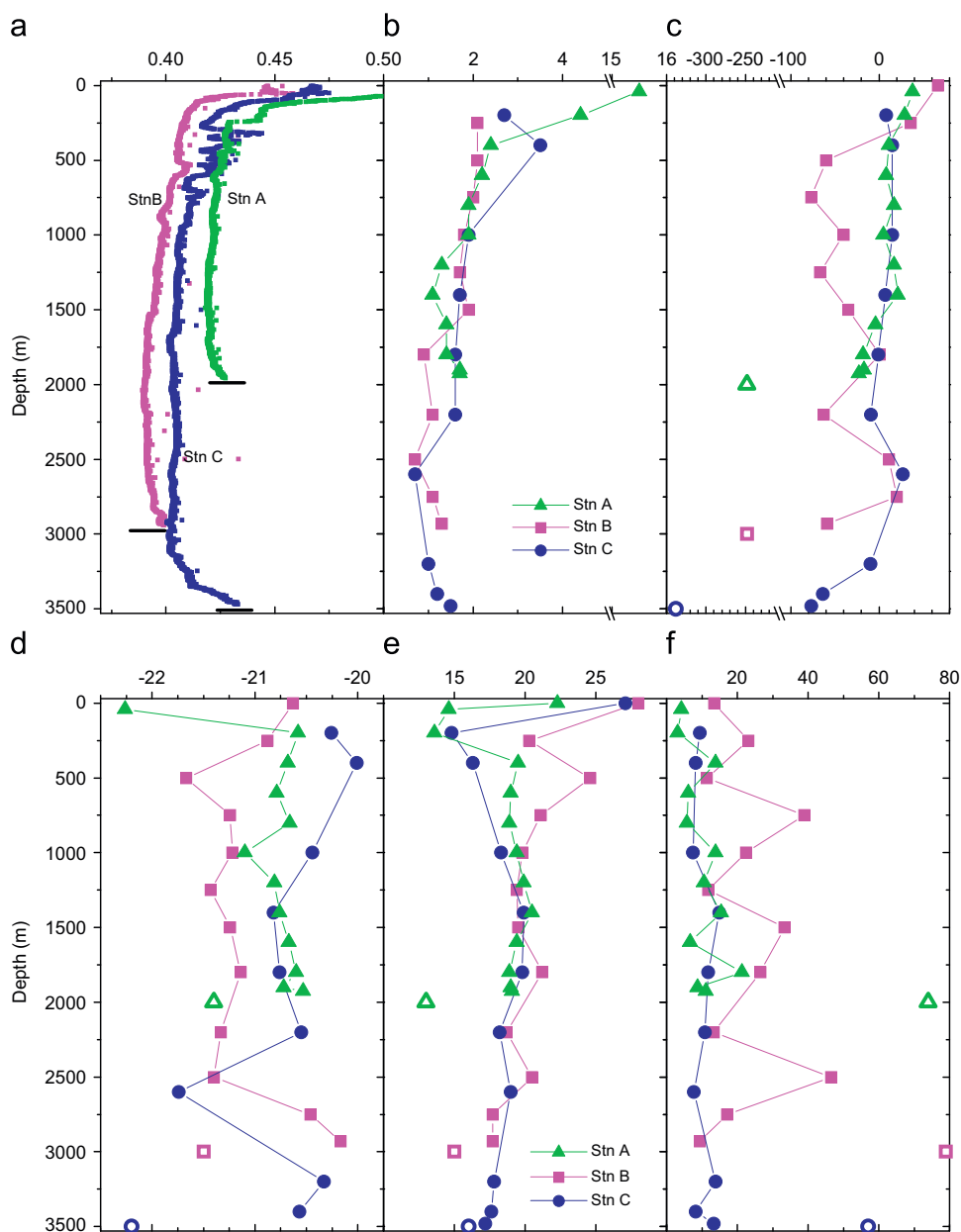


Fig. 3. (a) Beam attenuation coefficient measured in-situ using a transmissometer, (b) POC concentration in $\mu\text{gC/L}$, (c) $\Delta^{14}\text{C}$ values in ‰ (uncertainty = $\pm 10\%$), (d) $\delta^{13}\text{C}$ values in ‰ ($\pm 0.1\%$), (e) alkenone-derived temperature ($\pm 2^\circ\text{C}$), and (f) percentage of long-chain fatty acids in total fatty acids of suspended POC collected at 10–12 depths at each station. The horizontal bars on plot (a) indicate water depths. The open symbols are the values of the core-top (0–1 cm) sediments. Alkenone temperatures were estimated using the calibrations by Conte et al. (2006). Note the scale break for (b) and (c). The lines are for visual guidance and do not represent interpolation.

and a slight increase near the ocean floor – are similar to those observed in the North Central Pacific and the Sargasso Sea (Druffel et al., 1998, 2003).

3.4. Alkenone distribution and unsaturation index

The temperatures estimated from alkenone unsaturation index (U_{37}^k , Conte et al., 2006) of POC collected from surface waters at Stations A, B, and C via the ship's uncontaminated seawater intake were 22, 28, and 27°C ,

respectively (Figs. 3e and 5a). These values match ambient temperatures of the surface mixed layers observed with CTD to within the uncertainty ($\pm 2^\circ\text{C}$). The alkenone temperatures recorded from suspended particles recovered from the upper 500 m varied markedly between stations. Alkenone temperatures were lower than surface values by more than 7°C with the largest difference evident at Station C. Below 1000 m, the alkenone temperature tended to converge to $\sim 19^\circ\text{C}$ implying either more uniform source inputs or mixing.

Table 1

Properties of suspended POC and core-top sediment (0–1 cm horizon) collected at three stations on the northwest Atlantic margin.

Station	Depth (m)	Volume pumped (L)	POC ($\mu\text{gC/L}$)	$\Delta^{14}\text{C}$ (‰)	Avg. $\Delta^{14}\text{C}$ (‰)	$\delta^{13}\text{C}$ (‰)	U_{37}^k	Alkenone T ($^{\circ}\text{C}$)	Avg. alkenone T ($^{\circ}\text{C}$)	$\text{C}_{37:2}+\text{C}_{37:3}$ conc. ($\mu\text{gC/g POC}$)	Long-chain FAMES (%)	Avg. long-chain FAMES (%)	
Station A (2000m)	0						0.763	22					
	40	326	15.5	38	33	-22.3	0.420	15	17	415	4	4	
	200	726	4.4	29		-20.6	0.382	14		500	3		
	400	745	2.4	11		-20.7	0.639	20		178	14		
	600	784	2.2	8		-20.8	0.613	19		147	6		
	800	883	1.9	17		-20.7	0.609	19		142	6		
	1000	732	1.9	5		-21.1	0.632	19		101	14		
	1200	1733	1.3	17	2 ± 16	-20.8	0.655	20	19 ± 1	147	11	11 ± 5	
	1400	1467	1.1	21		-20.8	0.682	20		170	15		
	1600	1419	1.4	-4		-20.7	0.633	19		156	7		
	1800	1454	1.4	-18		-20.6	0.611	19		86	21		
	1900	827	1.7	-17		-20.7	0.616	19		72	9		
	1927	920	1.7	-23		-20.5	0.619	19		44	11		
		Core-top			-248		-21.4	0.482	13			74	
Station B (3000m)	0			67		-20.6	0.978	28			14		
	250	781	2.1	36	52	-20.9	0.674	20	24	261	23	18	
	500	701	2.1	-60		-21.7	0.860	25		469	11		
	750	733	2.0	-77		-21.2	0.711	21		1138	39		
	1000	857	1.8	-40		-21.2	0.651	20		1203	23		
	1250	890	1.7	-67		-21.4	0.632	19		1080	12		
	1500	659	1.9	-35	-37 ± 35	-21.2	0.637	19	20 ± 2	1118	33	23 ± 13	
	1800	1487	0.91	1		-21.1	0.716	21		1124	27		
	2200	1318	1.1	-63		-21.3	0.600	19		98	13		
	2500	922	0.66	11		-21.4	0.685	21		290	47		
	2750	1706	1.1	20		-20.5	0.552	18		81	17		
	2930	1385	1.3	-59		-20.2	0.556	18		550	9		
		Core-top			-248		-21.5	0.551	15			79	
	Station C (3500m)	0						0.948	27				
200		1101	2.7	8	8	-20.3	0.428	15	21	531	9	9	
400		705	3.5	15		-20.0	0.492	16		439	8		
1000		764	1.9	15		-20.4	0.583	18		127	8		
1400		764	1.7	7		-20.8	0.657	20		154	15		
1800		794	1.6	-1		-20.8	0.651	20		55	12		
2200		888	1.6	-9	-11 ± 36	-20.6	0.575	18	18 ± 1	84	11	11 ± 3	
2600		883	0.72	27		-21.7	0.612	19		105	8		
3200		1116	1.0	-10		-20.3	0.559	18		58	14		
3400		1088	1.2	-64		-20.6	0.547	18		40	8		
3480	856	1.5	-77		ND	0.531	17		114	13			
	Core-top			-338		-22.2	0.573	16			57		

The water depths of Station A ($39^{\circ}45'N$, $69^{\circ}44'W$), Station B ($39^{\circ}27'N$, $68^{\circ}21'W$), and Station C ($38^{\circ}36'N$, $68^{\circ}54'W$) were 1957, 2970, and 3500 m, respectively. The average values within and below the upper 0–250 m are also listed for $\Delta^{14}\text{C}$, alkenone-temperature, and long-chain fatty acid index. ND means no data. The $\Delta^{14}\text{C}$ value for this sample was normalized using the $\delta^{13}\text{C}$ value of -20‰ .

3.5. Distribution of saturated fatty acids (FAs)

Short-chain (carbon number ≤ 22) FAs and long-chain (carbon number ≥ 24) FAs are predominantly produced by marine organisms and terrestrial higher plants, respectively (Killops and Killops, 2004). Because of their proximity to the continents, and also the selective preservation of long-chain FAs in the marine environment

(e.g., Mollenhauer and Eglinton, 2007), sediments on the margin typically contain higher proportions of long-chain FAs. In this sense, relative contribution of long-chain FAs can be used as a tracer of margin-derived resuspended sediment.

FA 16:0 (saturated fatty acid with 16 carbons) was the most dominant among the saturated FAs followed by 14:0 and 18:0 (Table 2). Long-chain even-numbered FAs ($24 \leq$

carbon number ≤ 30) made up a significant fraction (average = 15%) of total even-numbered saturated FAs (C14:0~C30:0) for all samples analyzed. An index defined as the percent contribution of long-chain FAs to the total FAs (Bourbonniere and Meyers, 1996) shows the contribution of resuspended sediment (Fig. 3f). The values for the core-top sediments ranged between 57% and 79%. The values for water column suspended particles from the surface and 40 m samples are low (4% at Station A and 14% at Station B), as expected if marine primary production was the predominant source of FAs. However, the values at greater depths were mostly higher than the values in

the surface waters at each station, also supporting the existence of OC from margin-derived resuspended sediment. Diagenetic modifications in the water column may also influence this ratio, but the significantly higher values for certain water depths at Station B (average = 23%) than at the other stations (average = 11% for both stations) imply that other processes must also be responsible. Moreover, elevated FA index values are generally consistent with low and irregular $\Delta^{14}\text{C}$ values, although maxima do not always coincide with $\Delta^{14}\text{C}$ minima, indicating that these relationships are not straightforward.

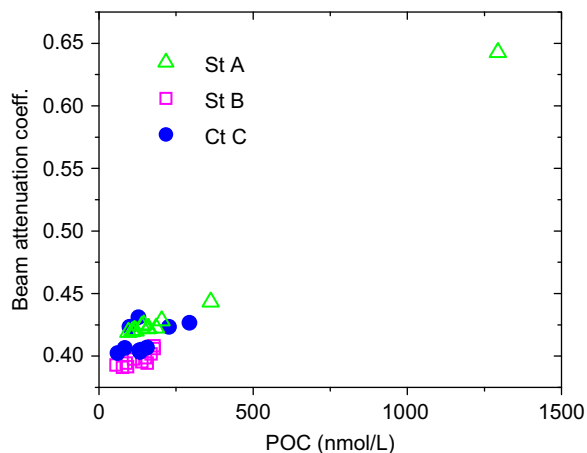


Fig. 4. Correlation between beam attenuation coefficients averaged over 40 m intervals and suspended POC concentrations in nmol/L at corresponding depths (see text for more detail).

4. Discussion

Suspended POC, operationally defined here as POC that is retained on a GF/F filter (nominal pore size = 0.7 μm), may derive from organic matter produced at various times and locations. In this study, we utilize $\Delta^{14}\text{C}$ values, alkenone-derived temperatures and FA chain-length index values as independent tracers of POC supply, on the basis that these parameters are affected by contributions from each source differently. Since alkenones are produced by phytoplankton, the alkenone-derived temperatures are those of the surface euphotic zone waters where these organisms grow. In this context, alkenone temperature may serve as an effective tracer of the provenance where the POC is ultimately derived if the surface waters exhibit significant temperature contrasts. On the NW Atlantic margin, waters nearer the coast and further northward (Scotian margin) are colder than waters basinward and to the south. The core-top sediments have a range of temperatures recorded in the sedimentary OC [8–9 °C in

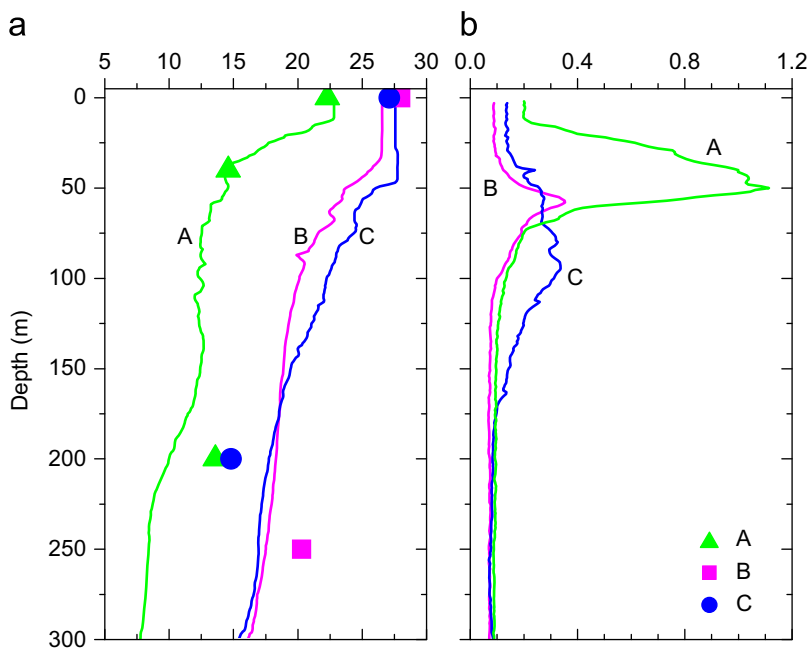


Fig. 5. (a) Alkenone-derived temperature recorded in suspended POC (triangles, squares, and circles for Station A, B, and C, respectively) and in-situ temperature (solid lines), and (b) fluorescence measured using a CTD-rosette system in the upper 300 m layer.

Table 2
Concentration of saturated fatty acids as fatty acid methyl esters (FAMES) in $\mu\text{gC/g}$ POC.

Station	C14:0	C16:0	C18:0	C20:0	C22:0	C24:0	C26:0	C28:0	C30:0	Total
Station A (2000 m)										
0										
40	1175	3340	296	105	70	119	54	32	16	5207
200	3601	9063	872	138	162	207	117	81	44	14,285
400	88	317	57	12	21	31	23	15	10	574
600	1776	5000	495	84	138	197	120	126	54	7990
800	579	2268	328	47	68	89	54	38	20	3492
1000	70	375	66	13	24	37	25	16	9	636
1200	393	1482	227	61	74	102	71	61	31	2501
1400	105	412	96	18	33	48	33	25	15	784
1600	714	2530	389	346	88	114	76	63	37	4357
1800	126	512	106	33	44	83	61	54	24	1044
1900	683	2951	538	91	123	169	123	85	46	4809
1927	162	613	136	26	35	52	31	25	14	1094
Station B (3000 m)										
0										
250	16	135	60	11	19	25	21	15	11	313
500	512	2235	441	78	125	171	123	93	51	3830
750	6	39	11	3	6	12	14	9	6	105
1000	749	2899	506	93	165	349	387	332	211	5691
1250	306	1719	274	55	83	111	99	77	38	2762
1500	0	37	1	1	2	4	6	7	4	63
1800	59	411	117	25	51	80	72	56	32	902
2200	92	419	135	18	33	46	30	19	12	803
2500	381	1803	579	96	192	560	740	891	472	5714
2750	53	279	70	11	23	35	28	17	10	527
2930	943	2876	570	83	123	197	143	92	43	5071
Station C (3500 m)										
0										
200	611	2582	387	78	109	157	106	76	50	4157
400	972	2231	217	53	74	117	87	71	40	3863
1000	657	1955	592	60	83	125	76	47	25	3620
1400	784	3603	648	120	229	302	194	241	205	6329
1800	268	1124	214	37	60	95	60	45	28	1930
2200	818	3193	600	85	115	268	141	119	60	5401
2600	848	3112	721	95	128	206	110	64	29	5312
3200	88	331	89	13	22	37	24	18	9	631
3400	379	1461	359	45	57	101	55	34	16	2507
3480	395	1387	363	58	85	159	102	62	29	2641

The concentrations should only be regarded as semi-quantitative because the recovery of the extraction process was not determined. The FAMES were identified by using a Supelco FAME standard.

the Emerald Basin off Nova Scotia and on the Laurentian Fan (Keigwin et al., 2003; Sachs, 2007); 13, 15, and 16 °C at Stations A, B, and C, respectively]. Therefore, resuspended sediment would carry various alkenone temperatures depending on its provenance. The annual average SST measured by satellites in the study area ranging between 16 and 20 °C (<http://nomad3.ncep.noaa.gov>) is higher than the alkenone temperatures recorded in the underlying sediments, implying lateral transport of OC from colder regions.

In using alkenone temperature as a tracer of POC provenance, several factors other than SST should also be considered. For example, the temporal aspects of alkenone production may be important. High vertical fluxes of alkenones were observed between August and October 2004 from a sediment trap deployed at 1000 m depth on a

mooring at Station B (Hwang and Eglinton, unpublished data). The turnover time of suspended POC is also important because of seasonal variations in SST that in turn influence alkenone temperatures. Moreover, while many studies have successfully related alkenone temperature in POC and sediments with the growth temperature of alkenone-producing organisms (e.g., Conte et al., 2001), factors other than temperature have been shown to influence U_{37}^k . In particular, nutrient availability and light limitation may influence alkenone unsaturation, resulting in lower or elevated apparent alkenone temperatures, respectively (Prah et al., 2006). High cell growth rate may also result in colder apparent alkenone temperatures (Epstein et al., 1998; Popp et al., 1998), and physiological response to environments may be different for different alkenone-producing strains (Popp et al., 1998). A certain

level of alteration of the alkenone temperature by abiotic degradation processes has been argued (e.g., Rontani et al., 2006). Despite these potential complications, the alkenone temperatures of suspended POC in surface waters closely match the observed SST. Hence, we suspect minor influences from non-temperature effects in this region.

Suspended POC $\Delta^{14}\text{C}$ will deviate from that of surface water DIC only as a consequence of inputs of pre-aged OC since POC turnover times are believed to be too short to affect $\Delta^{14}\text{C}$ values (Bacon and Anderson, 1982; Druffel et al., 1996). The most likely source of aged POC is resuspended sediment carrying terrestrial or marine organic matter. There are other potential sources of aged POC, such as anapleurotic incorporation of dissolved inorganic carbon by microorganisms (Rau et al., 1986), autotrophic incorporation of dissolved inorganic carbon by marine planktonic archaea (Pearson et al., 2001; Ingalls et al., 2006), and adsorption of dissolved organic carbon to particles (Williams and Druffel, 1987; Hwang et al., 2006). However, based on the magnitude of ^{14}C depletion, and on the covariance between $\Delta^{14}\text{C}$ and other properties, we suspect that incorporation of resuspended sediment is the primary cause of the observed low $\Delta^{14}\text{C}$ values, and focus our discussion on this process.

There are a few noticeable differences in suspended POC $\Delta^{14}\text{C}$ profiles between our study sites and those reported for other locations. In the north central Pacific, the $\Delta^{14}\text{C}$ values gradually decreased from surface to bottom and there was no observable difference between near-bottom $\Delta^{14}\text{C}$ values and those in the overlying water column (Druffel et al., 1992). However, at Station M, located near the rise of the California margin in the northeast Pacific (water depth = 4100 m; Druffel et al., 1998) and in the Sargasso Sea (water depth = 4450 m; Druffel et al., 1992) an abrupt decrease in $\Delta^{14}\text{C}$ was observed in near-bottom suspended particles. These features indicate the existence of BNLs maintained by resuspension and lateral transport of sediment containing pre-aged OC at both sites (Druffel et al., 1998, 2003; Sherrell et al., 1998). Another notable feature of the $\Delta^{14}\text{C}$ profiles is that the $\Delta^{14}\text{C}$ values in the subsurface layers are substantially lower than that of the surface water, and that $\Delta^{14}\text{C}$ values for samples below 500 m were near constant (except at Station B and within the BNLs). This observation suggests that suspended particle distributions in the upper water column are influenced by ^{14}C -depleted POC emanating from the shelf. Based on these observations, we discuss the potential sources of suspended POC in three different zones: (1) the upper water column, from surface to 500 m, (2) the meso- and bathy-pelagic zone from 1000 m to the upper boundary of BNLs, and (3) within the BNLs.

4.1. Upper water column: 0–500 m

The alkenone temperatures were the most variable in the upper 500 m of the water column at each station (Fig. 3e). At the time of sample collection (early summer), the water column was stratified, with surface mixed layer depths of between 10 m at Station A and 50 m at Station C

(Fig. 5a). Depths of the chlorophyll maximum determined by fluorometry – about 50, 58, and 94 m at Stations A, B, and C, respectively (Fig. 5b) – were deeper than the mixed layer depths. Fluorescence-weighted average seawater temperatures between the surface and 200 m, calculated as representative alkenone temperatures of freshly produced POC, were 15, 22, and 23 °C at Stations A, B, and C, respectively. Discrepancies may exist between the depths where the majority of POC and alkenones are produced. For example, it has been reported that alkenones are produced at depths shallower than that of the deep chlorophyll maximum (Prahl et al., 2005), suggesting that the estimated temperatures may represent a lower limit. The alkenone temperatures for suspended POC in the upper 300 m were similar to or slightly lower than the estimated temperatures at Stations A and B. However, at Station C, the temperature was lower than the estimated temperature by ~ 8 °C. This observation at Station C contrasts with previous studies where alkenone temperatures of suspended particles in the euphotic zone (Conte et al., 2001) and in the upper 100 m (Ternois et al., 1997) are strongly correlated with the in-situ temperatures. This different behavior may be due to the rapid increase in SST (more than 10 °C) from April to June, which complicates source assignments. However, the observation that the difference was significant only at Station C suggests allochthonous OC input at this station. The low $\Delta^{14}\text{C}$ value at Station C (8‰ at 200 m depth) and the highest variability of beam attenuation at this station (Fig. 2c) support the notion that over this depth range, there is a greater influence of resuspended sediment advected from colder source region at Station C.

The INLs observed in the transmissometry profiles are likely to have emanated from the shelf break. The density surface of the layer that exhibits anomalous temperature and salinity at 320 m at Station C intersects the margin at about 100 m, near the shelf break (Pickart, 2000). The allochthonous particles derived from the north and/or west would carry a lower apparent alkenone temperature signal.

Several processes may be responsible for lateral transport of resuspended sediment from the shelf. Detachment of benthic boundary layers (BBLs) from the shelf break and injection into slope waters along isopycnal surfaces has been documented on the New England shelf south of Cape Cod (Pickart, 2000). A similar phenomenon has been observed by transmissometry survey, and simulated by a regional model in the West Africa coast (Karakaş et al., 2006). These BBLs will entrain and transport resuspended sediment basinward. Cross-shelf transport of clouds of high particle concentration was also observed from transmissometer moorings along a transect on the New England shelf (Churchill et al., 1988). Submarine canyons may serve as conduits for transport of suspended particles to the continental slope (Biscaye and Anderson, 1994).

An alternative explanation may be the influence of warm-core rings (WCRs). Warm-core rings are produced from meandering of the Gulf Stream in the slope west of the New England Seamounts (Evans et al., 1985). These rings, which can reach several hundred kilometers wide

and persist up to several months, typically migrate to the west–southwest before being reabsorbed into the Gulf Stream. AVHRR satellite images (obtained from <http://fermi.jhuapl.edu/avhrr/>), show that a WCR, ~200 km wide, was spawned in mid- to late-May at around 67.5°N and 39.5°W. It migrated west–southwest slowly until the undulating north wall of the Gulf Stream engulfed it in late-June. The sampling of suspended POC was performed when this merge was occurring (Fig. 1b) and temperature profiles suggest that both Stations B and C were within the entrainment zone (i.e., periphery) of the WCR (Fig. 2a). Station B has more prominent trace of the thermostad layer between ~100 and ~300 m than Station C. The temperatures of surface waters at both stations were similar to that of the Gulf Stream. The profiles of temperature and salinity at Station C reveal that the upper 500 m layer was very dynamic. An interesting phenomenon involved in WCRs is the entrainment of shelf water as streamers that move basinward as a result of the clockwise rotation of the WCR, contributing to shelf water removal (Garfield III and Evans, 1987). Bishop and Joyce (1986) showed that particles inside of and in the entrainment zone of a WCR (WCR 82B) were controlled by different processes and had different origins. The water in the entrainment zone of the WCR was influenced by waters from the continental shelf (Bishop and Joyce, 1986). The entrained shelf water would contain POC produced over the Scotian shelf and hence carry a “cold” alkenone temperature signal. It therefore appears plausible that Station C was affected by a shelf water streamer more than the other stations.

4.2. Meso- and bathy-pelagic zone (between 1000 m and upper boundary of BNL)

Below ~1000 m depth, all alkenone temperatures converge to ~19°C (Fig. 3e). The alkenone temperatures carried by suspended POC are likely to exhibit a time-integrated signal given its estimated turnover time in the water column of months to several years (Bacon and Anderson, 1982; Druffel et al., 1996). Average alkenone temperatures of suspended POC were higher than the corresponding core-top values, the latter displaying a significant basinward increase (13, 15, and 16°C at Stations A, B, and C, respectively). The slight decrease in alkenone SST in suspended particles with increasing

sampling depth at Stations B and C may suggest an increased contribution of resuspended sediment in the lower water column.

Using a radiocarbon isotopic mass balance, we estimated the contribution from allochthonous OC under the assumption of two end-member mixing between freshly produced POC and an allochthonous source. The end-member for recent primary production should be tightly constrained and we adopted the value of suspended POC collected from the ship's uncontaminated seawater intake in the study area in 2005 and during another cruise in summer 2007 ($60 \pm 10\%$, $n = 4$). The end-member for resuspended sediment is hard to determine because the provenance of this POC is not well known. The $\Delta^{14}\text{C}$ values of core-top sediments represent a plausible allochthonous POC end-member, but these values vary spatially. In general, the $\Delta^{14}\text{C}$ values of core-top sediments in this region decrease with increasing bottom depth (Hwang and Eglinton, unpublished data). Two scenarios were considered for allochthonous POC: cross-shelf advection of resuspended sediment from the shelf, and along-slope advection of resuspended sediment from the upstream slope further north. The values of $-140 \pm 50\%$ and $-260 \pm 50\%$ were chosen for the shelf and slope sediment, respectively, considering the relationship between the bottom depth and $\Delta^{14}\text{C}$ value of core-top sediment. The uncertainty of $\pm 50\%$, although somewhat arbitrary, reflects the scatter of $\Delta^{14}\text{C}$ values. Allochthonous POC was estimated to account for 25–54% of suspended POC according to the former scenario and 15–33% according to the latter scenario, with the highest values at Station B (Table 3).

Station B is distinct from the other two stations in terms of lower and more erratic $\Delta^{14}\text{C}$ values and higher and more variable FA chain-length index values. The heterogeneous vertical distribution of biogeochemical properties may reflect sporadic particle clouds of high particle concentration. During the observations of particle concentrations by transmissometer moorings deployed along 71°W on the New England shelf and slope as part of the SEEP experiment, it was observed that the cross-shelf extent of turbid particle clouds was smaller than the ~30 km distance between the moorings (Churchill et al., 1988). An alternative explanation may be that Station B is east of Stations A and C, and closer to the New England Seamounts (Fig. 1c) where eddy kinetic energies in both the upper and the abyssal ocean are

Table 3

Estimates of the contribution of allochthonous POC to suspended POC in the meso- and bathy-pelagic zone excluding the bottom nepheloid layers.

Station	Observed $\Delta^{14}\text{C}$ (‰)	Autochthonous $\Delta^{14}\text{C}$ (‰)	Allochthonous			
			Shelf sediment $\Delta^{14}\text{C}$ (‰)	%	Slope sediment $\Delta^{14}\text{C}$ (‰)	%
A	11 ± 9 (600–1600 m)	60 ± 10	-140 ± 50	25 ± 9	-260 ± 50	15 ± 5
B	-47 ± 28 (750–2200 m)	60 ± 10	-140 ± 50	54 ± 20	-260 ± 50	33 ± 11
C	8 ± 14 (1000–2600 m)	60 ± 10	-140 ± 50	26 ± 11	-260 ± 50	16 ± 6

Depths of the samples used for averaging are shown in the parenthesis. Two scenarios were considered for allochthonous source of POC: shelf sediment and slope sediment. The uncertainties of the estimates are propagated uncertainties. See text for more details.

higher (Thompson and Schmitz Jr., 1989). It therefore seems plausible that the current regime and particle fields may differ at Station B as a consequence of these features.

POC concentrations at all stations reach a minimum at about 500–1000 mab (1400, 2500, and 2600 m depths at Stations A, B, and C, respectively). At these depths, $\Delta^{14}\text{C}$ values and alkenone temperatures were generally higher than at the adjacent depths. If samples deeper than 500 m (and above the BNL) only are considered, the $\Delta^{14}\text{C}$ values were the highest and alkenone temperatures were generally at a maximum at the depth of minimum POC. These depths thus appear to be least affected by allochthonous carbon, at least in terms of that material carrying low $\Delta^{14}\text{C}$ values and low alkenone temperatures.

4.3. Bottom nepheloid layers (BNLs)

Factors that influence the $\Delta^{14}\text{C}$ values of suspended POC in the BNLs include the $\Delta^{14}\text{C}$ values of surface sediment, strength of bottom currents that promote resuspension, the flux and quality of sinking POC reaching abyssal depths, and potential selective degradation of POC. The $\Delta^{14}\text{C}$ values in the BNLs decreased as the water depths of the stations increased, consistent with the trend towards decreasing $\Delta^{14}\text{C}$ values of surface sediments (0–1 cm horizon) with increasing bottom depth (–248‰, –248‰, and –338‰ at Stations A, B, and C, respectively). However, alkenone temperatures in the BNLs also decreased as the water depths of the stations increased, in the opposite direction of both SST and alkenone temperatures recorded in surface (0–1 cm) sediments from corresponding stations (Fig. 3e; Table 1). These observations suggest a significant influence from resuspended sediment POC supplied from the distal margin via BNLs. A transmissometry survey undertaken as part of the Line W program clearly showed that the thickness and beam attenuation signal of the BNLs increase with increasing bottom depth from the coast toward the Gulf Stream region (J. Toole, unpublished data, 2007).

It is surprising that FA index values do not increase significantly in the BNLs considering that the index values for core-top sediment were much higher than those in the overlying water column. The BNL is a biologically active layer supporting high levels of bacterial activities, and exhibiting high chlorophyll-*a* to POC ratios (Boetius et al., 2000). The FA index may also be affected by resuspension of fresh phytodetritus from the seafloor (Thomsen and van Weering, 1998). In both cases, biological reprocessing of POC may cause the anomalously low FA index values that were observed.

5. Conclusions

Resuspension of sediment and lateral transport of lithogenic material, responsible for hemipelagic sedimentation, has long been recognized as an important source of mineral to the continental margins and adjacent deep ocean basins (McCave and Tucholke, 1986; Rea and Hovan, 1995). Our results show that this process is also important for OC cycling, and low $\Delta^{14}\text{C}$ values of suspended POC and

sinking POC reported for various oceanic settings suggest that this phenomenon may be of widespread significance. Overall, our results show that the NW Atlantic continental slope system (and likely other margin systems) is complex in terms of particle sources and distribution. Clearly, larger scale and more intensive samplings at various times of the year will be necessary to better understand the OC cycle on this margin. High-resolution models such as ROMS (regional ocean modeling system; Karakaş et al., 2006) will be important for developing a more comprehensive picture of the processes involved. Nevertheless, several observations can be drawn from this initial survey of suspended POC distribution and composition. In particular, it is evident that particle dynamics and biogeochemical properties of suspended POC on the New England slope are influenced by intermediate-depth and near-bottom transport of resuspended sediment in addition to regional variations in surface ocean productivity. The origin of POC entrained in these layers and throughout slope waters in this region may also be influenced by warm-core rings from the Gulf Stream. Specific findings from this study include:

- Prominent intermediate nepheloid layers exist in the upper 1000 m in addition to distinct benthic nepheloid layers up to several hundred meters above the bottom, implying at least two modes of supply of suspended POC via lateral transport.
- Relatively low suspended POC $\Delta^{14}\text{C}$ values and alkenone-derived temperatures, and elevated particle concentrations at intermediate depths (below the surface mixed layer and shallower than 500 m) suggest the supply of resuspended sediment emanating from the shelf break as intermediate nepheloid layers.
- ^{14}C -based isotopic mass balance calculations suggest that remobilized sediment accounts for 15% to 54% of suspended POC below ~1000 m.
- Station B, centered on the 3000 m isobath, was distinct from the other stations in terms of low and irregular $\Delta^{14}\text{C}$ values, and higher relative abundances of terrestrially derived long-chain FAs. The greater influence of continental shelf-derived OC at this station may reflect its position within the core of the Deep Western Boundary Current, or its closer proximity to the New England seamounts. In either case, this observation indicates that input of resuspended sediment is spatially heterogeneous over a scale smaller than 150 km.

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