

Widespread influence of resuspended sediments on oceanic particulate organic carbon: Insights from radiocarbon and aluminum contents in sinking particles

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[1] Particulate organic carbon (POC) in the ocean often exhibits more depleted radiocarbon contents (lower $\Delta^{14}\text{C}$ values) than expected if its sole source were POC recently synthesized by primary production and export from the overlying surface waters. An examination of available $\Delta^{14}\text{C}$ data sets for sinking POC show that this phenomenon is both common and globally widespread. Also, a strong correlation is found to exist between $\Delta^{14}\text{C}$ values of organic carbon and aluminum content in sinking particles that is consistent over a range of oceanic settings. Together, these findings imply that aged organic carbon associated with lithogenic material from sediment resuspension is responsible for the observed low $\Delta^{14}\text{C}$ values as opposed to other processes such as incorporation of dissolved inorganic carbon or dissolved organic carbon into POC at depth. An estimate based on POC flux-weighted $\Delta^{14}\text{C}$ values shows that about 35% of sinking POC at the locations studied is derived from resuspended sediment. Our results suggest that resuspension of sediment and its subsequent lateral transport is an important component of the oceanic carbon cycle and should be considered in models of oceanic carbon export and burial.

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1. Introduction

[2] The photosynthetic fixation of carbon by phytoplankton in the surface ocean, and the subsequent export and burial of a fraction of this carbon in deep waters and underlying sediments is a fundamental component of the global carbon cycle, and drives many oceanic biogeochemical processes [Hedges, 1992]. Understanding the ocean's capacity to take up and sequester CO_2 via this "biological pump" [Volk and Hoffert, 1985] is of key importance in gauging responses to recent and future anthropogenically derived increases in atmospheric CO_2 . A key issue concerns the sinking flux of particulate organic carbon (POC) produced in surface waters, yet the mechanism(s) and efficiency of POC transfer to depth remain only partly understood [Martin *et al.*, 1987; Honjo *et al.*, 2008]. Multiple factors have been proposed to influence carbon export from the surface ocean, and progress is

being made in understanding the underlying processes, and in reconciling estimates based on different approaches [Dunne *et al.*, 2007]. However, considerable uncertainty remains in models of carbon export. The models are particularly unconstrained for regions overlying, or adjacent to, continental margins where carbon cycling is highly complex and dynamic [Inthorn *et al.*, 2006]; yet these systems are of global importance as regions of high primary productivity and carbon export [Hedges, 1992].

[3] One interesting geochemical characteristic of POC sinking from the surface ocean (which is true for suspended POC as well) is that its radiocarbon content (expressed herein as $\Delta^{14}\text{C}$ values) is generally lower than that expected if primary production in the overlying water column was the sole source of POC [Druffel and Williams, 1990; Druffel *et al.*, 1992]. These low $\Delta^{14}\text{C}$ values imply an additional source of aged (low $\Delta^{14}\text{C}$) carbon. Reconciling these lower than expected $\Delta^{14}\text{C}$ values of sinking POC may have important ramifications for models of carbon export, and hold clues concerning organic matter cycling in the ocean.

[4] Adsorption of dissolved organic carbon (DOC) to particles has been suggested as one potential source of this aged carbon [Druffel and Williams, 1990; Hwang *et al.*, 2006]. This could represent a significant removal process of DOC whose average ^{14}C age in the deep oceans is 4000–6500 years ($\Delta^{14}\text{C} = -394\text{‰}$ in the Sargasso Sea and -524‰

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in the North Central Pacific [Druffel et al., 1992; Beupré and Druffel, 2009]). Incorporation of dissolved inorganic carbon (DIC) by heterotrophic bacteria through an anapleurotic reaction, i.e., a direct uptake of bicarbonate ion through the tricarboxylic acid cycle [Rau et al., 1986], and by autotrophic archaea [Pearson et al., 2001; Ingalls et al., 2006] associated with sinking particles, also represents alternative sources of aged carbon. The contribution from bacterial and archaeal biomass to sinking POC below the euphotic zone is not well understood, but is expected to be spatially variable. Evidence for autotrophic incorporation of DIC stems from $\Delta^{14}\text{C}$ measurements of archaeal lipid biomarkers [Pearson et al., 2001; Ingalls et al., 2006] and from ^{13}C -labeling experiments [Wuchter et al., 2003]. Considering that crenarchaeota account for a large fraction of ocean picoplankton, especially in the deep ocean where their abundance is equivalent to or greater than bacteria [Karner et al., 2001], in situ incorporation of DIC to the organic matter pool in the deep ocean may be significant. Additional explanations of lower $\Delta^{14}\text{C}$ values of POC include input of aged terrestrial organic carbon via winds [Eglinton et al., 2002] or rivers [Kao and Liu, 1996; Masiello and Druffel, 2001; Goñi et al., 2005] to the surface waters, and selective preservation and long-term recycling of refractory organic components ultimately derived from primary production [Wang et al., 1998; Hwang and Druffel, 2003, 2006].

[5] Another potential source of aged POC directly supplied to deep waters is from sediment resuspension and lateral particle transport [Nakatsuka et al., 1997; Sherrell et al., 1998; Honda et al., 2000; Hwang et al., 2004, 2008]. Sediments deposited on continental margins are prone to resuspension by dynamic, and often vigorous bottom currents [Rea and Hovan, 1995]. Resuspension of sediment and subsequent lateral transport is evident from several observations, including material fluxes to sediment traps [Honjo et al., 1982; Biscaye et al., 1988; Freudenthal et al., 2001; Smith et al., 2001], bottom and intermediate nepheloid layers [McCave, 1983; McCave et al., 2001; Karakaş et al., 2006; Lam and Bishop, 2008], carbon supply and oxygen consumption in sediments [Jahnke, 1996], ^{14}C ages of suspended and sinking POC [Druffel et al., 1998a; Honda et al., 2000; Hwang et al., 2004, 2008, 2009a, 2009b], aluminum contents of suspended particles [Sherrell et al., 1998], the isotopic and molecular composition of organic matter in deep sea sediments [Benthien and Muller, 2000; Ohkouchi et al., 2002; Mollenhauer et al., 2006], and regional ocean modeling [Karakaş et al., 2006]. Sedimentation of aged POC on the continental margin has also been demonstrated by older ages of sedimentary organic matter in the bioturbated layer than expected from the sedimentation rate and mixing of surface sediment with deeper and older sediment [Anderson et al., 1988; Hwang et al., 2005]. If sediment resuspension was responsible for low $\Delta^{14}\text{C}$ values observed even in open ocean settings [Druffel et al., 1992], we would need to invoke large-scale, long-distance lateral POC transport. The existence of such a mechanism would have broad implications for our understanding of biogeochemical cycling of carbon and associated elements in the ocean. Although evidence exists for each of the above processes, the extent to which

they contribute to low $\Delta^{14}\text{C}$ values of sinking POC is not well understood.

[6] One way to distinguish the contribution of resuspended sediment from the other processes is to examine the ^{14}C and aluminum (Al) contents of a given sample. Al is an effective tracer of resuspended sediment, as it originates from weathered crust and is a major component of sediments delivered to continental margins. Eolian transport and sediment resuspension are two mechanisms by which Al can be transported to the open ocean and incorporated into sinking particles, in addition to riverine input that has local importance. Here, we use new and published $\Delta^{14}\text{C}$ values and Al concentrations to show that entrainment of resuspended sediment is the main source of aged carbon into sinking POC.

2. Data

[7] $\Delta^{14}\text{C}$ values used in this study were determined using accelerator mass spectrometry (AMS) techniques [Vogel et al., 1987] with the exception of a few early samples that were measured using beta counting techniques [Druffel et al., 1986, 1998b]. Samples were pretreated to remove inorganic carbon by either addition of H_3PO_4 [Druffel and Williams, 1990; Druffel et al., 1992; Anderson et al., 1994; Hwang et al., 2004], rinsing with HCl [Santschi et al., 1999; Honda et al., 2000; Ootosaka et al., 2008], or HCl fuming [Nakatsuka et al., 1997; Santschi et al., 1999; Hwang et al., 2008, 2009a]. $\Delta^{14}\text{C}$ values have been measured for sinking POC collected at various times, depths and locations (Figure 1a and Table 1). The time span of reported data at a given location ranges from a few years [Hwang et al., 2004] to a few days for a single sample collection [Druffel et al., 1992]. In some cases, samples spanning several months were combined to produce a single data point [Wakeham, 2006]. In Figure 1b, the error bars represent the standard deviation of the mean of the data points; single-point results do not have error bars. The $\Delta^{14}\text{C}$ values of sinking POC are normalized by subtraction of the $\Delta^{14}\text{C}$ values of DIC in the corresponding surface waters. Measured $\Delta^{14}\text{C}$ values of suspended POC collected from the surface waters are also used for this purpose under the assumption that suspended POC in surface water is dominated by in situ primary production. For example, the $\Delta^{14}\text{C}$ values of suspended POC collected at 25 m in the NE Pacific were $52 \pm 13\%$ and $59 \pm 14\%$ in September 1994 and June 1995, respectively, and the corresponding values for DIC were $65 \pm 9\%$ and $68 \pm 9\%$ [Druffel et al., 1998a; Masiello et al., 1998].

[8] With the exception of the NE Pacific samples (24 measurements), all Al data used in this study have been previously published. Al concentrations for NW Atlantic margin [Hwang et al., 2009a], NE Pacific (this study), Canada Basin [Hwang et al., 2008] were measured at Woods Hole Oceanographic Institution by inductively coupled plasma–emission spectroscopy (ICP-ES; Jobin-Yvon Horiba, ULTIMA2), with a detection limit of $22.5 \mu\text{g g}^{-1}$ and a standard deviation of 5.1% [Honjo et al., 1995]. Samples were treated following a modified high-temperature alkali fusion method using Alfa Aesar™ Spectroflux™ and sequentially dissolving with nitric acid [Ingamells, 1970].

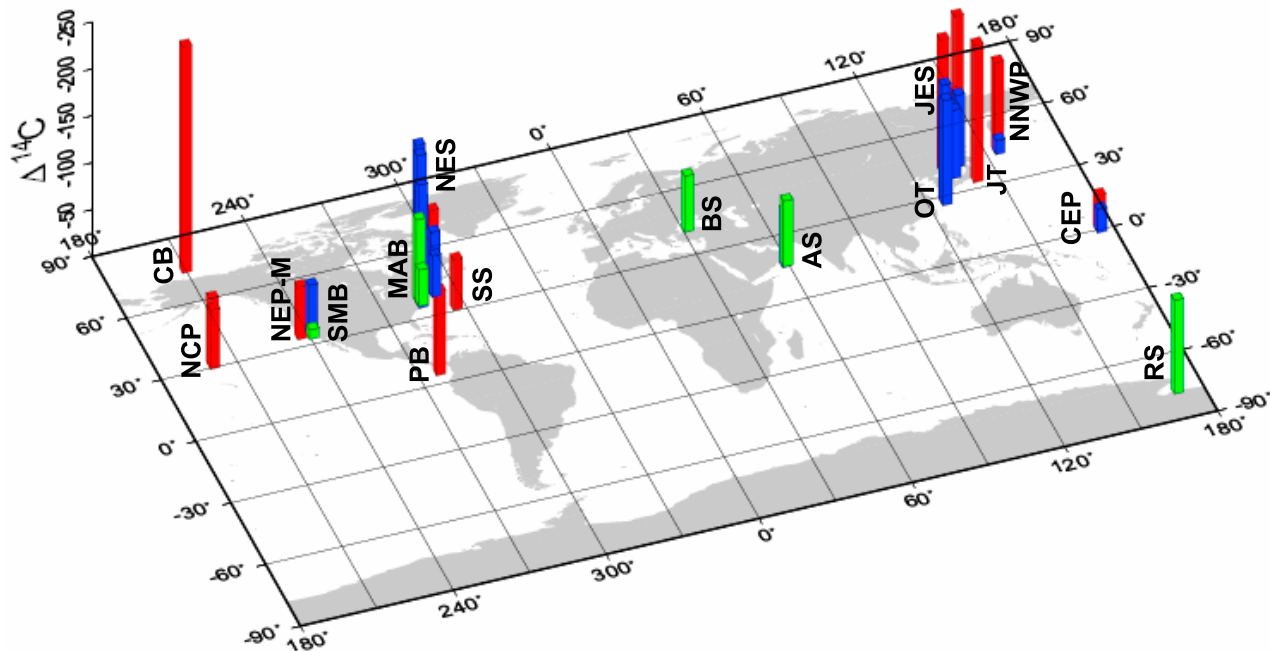
USGS MAG1 was used as reference material. Other previously reported results were obtained following a similar method with the exception of the Japan/East Sea samples that were treated with a mixture of HNO₃, HClO₄, then HF [Otosaka *et al.*, 2004]. The Okinawa Trough data were obtained by ICP-ES (JY138 ULTRACE) with a measurement error of less than 7% [Honda *et al.*, 2000]. The Japan/East Sea data were obtained by ICP-atomic emission spectroscopy

(AES) (SEIKO SPS7700) with an uncertainty smaller than 8% [Otosaka *et al.*, 2004].

3. Results

[9] Figure 1 shows the normalized $\Delta^{14}\text{C}$ values of sinking POC at various locations ranging from continental shelves to the open ocean. The sampling locations vary in terms of total

a)



b)

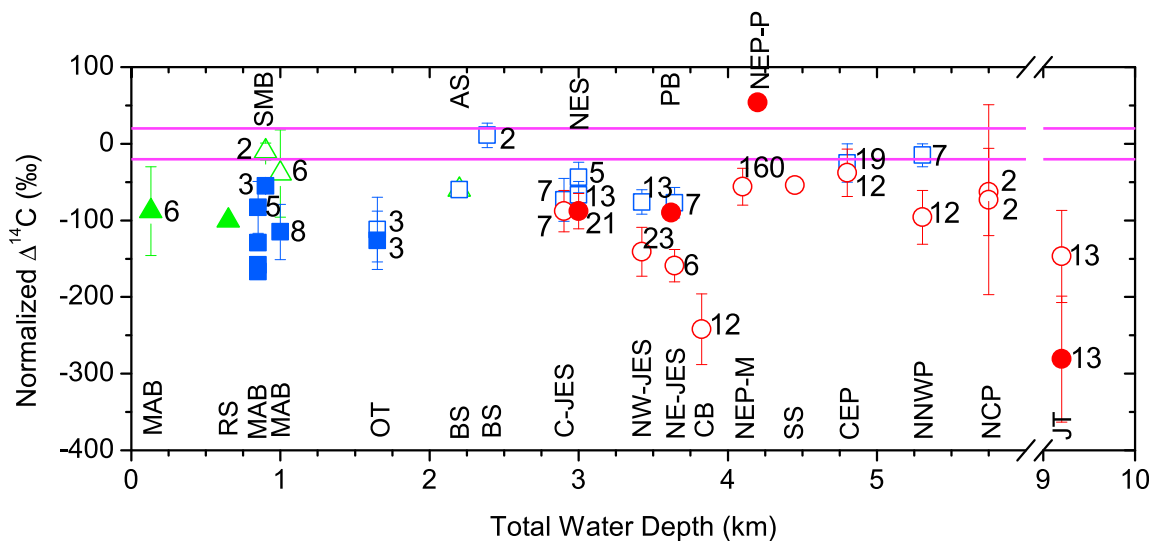


Figure 1

water depth and height above the seafloor. Only one point at Station P in the northeast Pacific was significantly higher than the corresponding DIC $\Delta^{14}\text{C}$ values (not plotted in Figure 1). This high value was interpreted by the authors as the contribution of terrestrial POC to that site [Druffel *et al.*, 1986]. Only a few locations on or proximal to continental margins (at 100 m in the Santa Monica Basin and at 1200 m in the Black Sea) exhibit normalized $\Delta^{14}\text{C}$ values that fall between +20‰ and -20‰, which conservatively represents the seasonal variability in DIC $\Delta^{14}\text{C}$ values.

[10] The majority of normalized $\Delta^{14}\text{C}$ values were depleted by more than 20‰. These low values were observed not only from near the seafloor (solid symbols in Figure 1b) and shallow margin environments, where local resuspension of sediment is expected, but also in open ocean settings such as the abyssal north central Pacific and the central Arctic Ocean (Canada Basin). The lowest normalized $\Delta^{14}\text{C}$ values were observed at the latter site [Hwang *et al.*, 2008] and in the Japan Trench [Nakatsuka *et al.*, 1997]. Other than these extreme values, the normalized $\Delta^{14}\text{C}$ values range between 0‰ and -200‰. The average normalized $\Delta^{14}\text{C}$ value for all sites where measured data have been reported is $-87 \pm 63\%$ (the uncertainty reflects the standard deviation of each average value representing each location and depth), and the average value weighted by POC flux is -95‰ (it is not easily doable to estimate the uncertainty of this value, but we believe that it is similar to or slightly larger than $\pm 63\%$ as determined above), corresponding to a conventional ^{14}C age of 800 years.

4. Discussion

[11] At the relatively few locations where both $\Delta^{14}\text{C}$ and Al contents are available (total 156 data points at 5 locations), the normalized $\Delta^{14}\text{C}$ and Al values show a strong negative quasi-linear correlation ($r^2 = 0.79$ when the Japan/East Sea and Canada Basin data are excluded (Figure 2); see below). This kind of correlation can be expected if the POC is a mixture of two end-members: freshly produced surface POC (normalized $\Delta^{14}\text{C} = 0\%$, Al = 0%) and aged POC tightly associated with clay and other aluminosilicates that are an

important component of lithogenic particles. The most likely source of the lithogenic particles is lateral supply of resuspended oceanic sediments (other potential sources such as eolian input are discussed below). $\Delta^{14}\text{C}$ values also exhibit similar relationships with other tracers of sediments such as excess Mn in the Japan/East Sea [Otosaka *et al.*, 2008] and Fe on the New England slope ($R^2 = 0.82$, $n = 39$ (S. J. Manganini and T. I. Eglinton, unpublished data, 2006)), further supporting our argument that the low normalized $\Delta^{14}\text{C}$ values are caused by the presence of resuspended sediment.

[12] Assuming the above is the case, the theoretical maximum Al content must be close to 8.2%, which represents the average Al content in the continental crust [Taylor, 1964]. For example, if the POC concentration of this end-member is 5% (equivalent to 12.5% as organic matter), then the Al content is 7.2% [= $8.2 \times (1 - 0.125)$]. If resuspended sediment contains other biogenic phases such as SiO_2 and CaCO_3 , the resultant Al content will become lower than this value. However, the contents of these biogenic minerals in resuspended sediment are expected to be small. For example, Al consistently accounts for 7–8% of total mineral mass for the clay size fraction ($<3 \mu\text{m}$) in coastal sediment on the Washington margin [Keil *et al.*, 1994b], and comprises 7.9% of resuspended sediment collected from the Minas Basin in the Bay of Fundy, Nova Scotia, Canada [Moran and Moore, 1991].

[13] The mixing curve is determined by both POC concentrations and normalized $\Delta^{14}\text{C}$ values of the end-members. A linear relationship between these two parameters is expected only when POC contents in the two end-members are equal. If the concentration of aged POC is smaller than freshly produced POC, the mixing line will become convex and deviate from a straight line. The data points from three different locations, NW Atlantic margin, NE Pacific, Okinawa Trough, appear to fall along a straight line (Figure 2). The solid line in Figure 2 represents an example mixing line between the two end-members with the same POC concentration but different normalized $\Delta^{14}\text{C}$ values: one end-member with normalized $\Delta^{14}\text{C} = 0\%$, POC = 5%, Al = 0% and the other with normalized $\Delta^{14}\text{C} = -182\%$, POC = 5%, and the resultant Al = 7.18%. The dotted line represents another example of two end-member mixing of

Figure 1. (a) A map showing the location of sinking POC samples and their average normalized $\Delta^{14}\text{C}$ values. The $\Delta^{14}\text{C}$ values are normalized to the corresponding values of dissolved inorganic carbon or those of suspended POC in the surface waters. Samples are categorized into three groups depending on the sampling depth: <500 m (green), $500\text{m} \ll 2000$ m (blue), and >2000 m (red). Except for one sample (54‰, at Station P in the northeast Pacific, not plotted), all samples exhibit negative average normalized $\Delta^{14}\text{C}$ values. (b) Normalized $\Delta^{14}\text{C}$ values (‰) of sinking POC plotted with respect to the total water depth. The symbols represent average values of each time series data set (numbers by the symbols represent the number of data points). The error bars represent the standard deviation of $\Delta^{14}\text{C}$ values. No error bars are provided when only a single datum is available. The horizontal bars represent the range of surface normalized $\Delta^{14}\text{C}$ values if in situ production were the only source of sinking POC considering the uncertainties and seasonal fluctuation in surface DIC $\Delta^{14}\text{C}$ values. Depending on the sampling depth, the data points are categorized into upper ocean (<500 m; triangles), mesopelagic zone ($500 \ll 2000$ m; squares), and bathypelagic zone (>2000 m; circles). Solid symbols represent the samples collected within 500 m from the bottom. Letters below or above the symbols represent the names of the sites. Abbreviations for the site names are as follows: AS, Arabian Sea; BS, Black Sea; CB, Canada Basin; CEP, central Equatorial Pacific; C-JES, central Japan/East Sea; JT, Japan Trench; MAB, Middle Atlantic Bight; NCP, north central Pacific; NE-JES, northeast Japan/East Sea; NEP-M, northeast Pacific-Station M; NEP-P, northeast Pacific-Station P; NES, New England slope; NNWP, north-northwest Pacific; NW-JES, northwest Japan/East Sea; OT, Okinawa Trough; PB, Panama Basin; RS, Ross Sea; SMB, Santa Monica Basin; SS, Sargasso Sea.

Table 1. $\Delta^{14}\text{C}$ Values of Sinking POC Normalized to the Corresponding Values of Dissolved Inorganic Carbon or Those of Suspended POC in the Surface Waters

Site	Latitude	Longitude	Sampling Year	Water Depth (m)	Surface Water $\Delta^{14}\text{C}$ (‰)	Trap Depth (m)	Normalized $\Delta^{14}\text{C}$ ‰ (Number of Data)	Average POC Flux ($\text{mg C m}^{-2} \text{d}^{-1}$)	Average AI (%) (Number of Data)
Canada Basin [Hwang et al., 2008]	75.0	-150.0	2004–2005	3824	25 [Jones et al., 1994]	3067	-242 ± 46 (12)	0.6	6.7 ± 0.7 (21)
New England slope [Hwang et al., 2009a]	39.5	-68.3	2004–2005	3000	67 ^a	968 1976 2938	-44 ± 20 (5) -67 ± 18 (13) -88 ± 23 (21)	16.6 9.3 10.0	2.0 ± 1.0 (11) 2.4 ± 1.1 (17) 3.4 ± 0.8 (21)
NE Pacific Station P [Druffel et al., 1986]	50.0	-145.0	1983	4200	74	3800	54 (1)	8.8	
North central Pacific [Druffel et al., 1992]	31.0	-159.0	1985, 1987	5750	121	4120, 4200 5120	-63 ± 57 (2) -73 ± 124 (2)	2.5 ^b 2.2 ^b	
Sargasso Sea [Druffel et al., 1992]	31.8	-63.5	1990	4450	120	3200	-54 (1)	1.6 ^b	
Panama Basin [Druffel et al., 1998b]	5.3	-82.0	1984	3620	88	3354	-91 (1)	8.1 ^b	
Santa Monica Basin [Druffel and Williams, 1990]	33.8	-118.8	1986–1987	900	96	100 850	-10 ± 11 (2) -55 ± 10 (3)	31.0 [Nelson et al., 1987] 24.7 [Nelson et al., 1987]	
Ross Sea [Wakeham, 2006]	-77.0	170.0	1997–1998	650	-100	230	-100 (1)	45.4 [Dunbar et al., 1998]	
Black Sea [Wakeham, 2006]	42.0	32.0	1985–1986	2200	70	250, 1000	-60 (1)	11.2 [Hay et al., 1990]	
Arabian Sea MS-1, MS-3 [Wakeham, 2006]	17.0	59.0	1994–1995	1500, 2900	70	500, 1500	-60 (1)	15.4 ^c	
Okinawa Trough [Honda et al., 2000]	27.4	126.7	1993	1650	105	1000 1500	-112 ± 42 (3) -126 ± 38 (3)	4.8 4.7	4.8 ± 0.7 (42) 5.1 ± 0.9 (3)
Northwest Pacific NNWP-02 [Nakatsuka et al., 1997]	44	155	1989–1990	5305	-44 ^d	1335 4785	-15 ± 15 (7) -96 ± 35 (12)	5.1 4.5	
Japan Trench, JT-06 [Nakatsuka et al., 1997]	34.2	142.0	1990–1991	9200	27 ^e	4789 8789	-147 ± 60 (13) -281 ± 82 (13)	3.4 3.3	
Central Equatorial Pacific 175E [Nakatsuka et al., 1997]	0.0	175.0	1992–1993	4800	100 ^f	1700 4220	-25 ± 25 (19) -38 ± 31 (12)	1.7 1.3	
Northeast Pacific Station M [Hwang et al., 2004]	34.8	-123.0	1993–1998	4100	60 [Masiello et al., 1998]	3450	-56 ± 24 (160)	3.6	2.8 ± 0.8 (24)
Middle Atlantic Bight [Santschi et al., 1999]	36.1	-74.7	1993	850	60 [Bauer et al., 2002]	825 770 650 700	-158 (1) -167 (1) -129 (1) -83 \pm 34 (5)	71.6 55.4 46.9 24.7	
Middle Atlantic Bight, M-10 [Anderson et al., 1994]	36.9	-74.6	1988–1989	1000	35 ^g	130 990	-39 ± 57 (6) -115 ± 36 (8)	12.0 65.0	
Middle Atlantic Bight, M-8 [Anderson et al., 1994]	36.9	-74.7	1988–1989	130	35 ^g	120	-88 ± 58 (6)	75.0	
Northwest Japan/East Sea [Otosaka et al., 2008]	41.2	132.4	2000–2002	3424	61 [Aramaki et al., 2007]	927 2746	-76 ± 16 (13) -141 ± 32 (23)	30.9 16.1	2.8 ± 0.9 (27) ^h 3.8 ± 1.3 (49) ^h
Northeast Japan/East Sea [Otosaka et al., 2008]	42.5	138.5	2000–2001	3642	56 [Aramaki et al., 2007]	1057 3043	-77 ± 20 (7) -159 ± 21 (6)	11.1 4.5	1.8 ± 0.2 (26) ^h 3.3 ± 0.8 (26) ^h

Table 1. (continued)

Site	Latitude	Longitude	Sampling Year	Water Depth (m)	Surface Water $\Delta^{14}\text{C}$ (‰)	Trap Depth (m)	Normalized $\Delta^{14}\text{C}$ ‰ (Number of Data)	Average POC Flux ($\text{mg C m}^{-2} \text{d}^{-1}$)	Average Al (%) (Number of Data)
Central Japan/East Sea [Otosaka et al., 2008]	38.0	135.0	2000–2001	2900	70	1175	-73 ± 28 (7)	27.1	2.7 ± 0.9 (13) ^b
					[Aramaki et al., 2007]	2100	-88 ± 27 (7)	19.8	3.6 ± 0.9 (13) ^b
Black Sea [Jones and Gagnon, 1994]	43.0	34.0	1988	2386	87.5	1200	11 ± 16 (2)	13.1 [Hay et al., 1990]	

^a $\Delta^{14}\text{C}$ value of suspended POC collected from the ship's clean seawater intake [Hwang et al., 2009b].

^bPOC fluxes at each sampling depth were estimated based on the data of François et al. [2002] and Martin et al.'s [1987] equation.

^cThe average value of POC flux at 808 m and 999 m at Station M-1, and at 746 m and 858 m at Station M-3 [Honjo et al., 1999].

^d $\Delta^{14}\text{C}$ value of suspended POC collected at 50 m in 1985 [Nakatsuka et al., 1997].

^e $\Delta^{14}\text{C}$ value of suspended POC collected at 200 m in 1987 [Nakatsuka et al., 1997].

^fAn assumed $\Delta^{14}\text{C}$ value based on the work by Levin and Hessheimer [2000].

^gThe highest $\Delta^{14}\text{C}$ value of sinking POC captured by a 130 m trap [Anderson et al., 1994].

^hJapan/East Sea aluminum data are from Otosaka et al. [2004].

fresh POC with organic matter of an older age and a lower POC concentration ($\Delta^{14}\text{C} = -274\text{‰}$, POC = 3%). In this case, a $\Delta^{14}\text{C}$ value of -274‰ represents the normalized value of -214‰ to the surface DIC $\Delta^{14}\text{C}$ value of $+60\text{‰}$ (regarding the choice of -214‰ , see discussion below). The choice for POC concentration of aged organic matter is somewhat arbitrary, lying between that of the average sinking POC (5%) and lower values for surface sediments. POC contents of sinking particles do not vary markedly in the deep ocean below mesopelagic/bathypelagic boundary [Armstrong et al., 2001], and averages $5.0 \pm 2.3\%$ for sinking particles intercepted at depths greater than 1000 m ($n = 2083$, data compiled by S. Honjo and R. Francois; http://www1.who.edu/datasys/SMP/Honjo/sed_traps.htm). This value decreases only slightly to $4.5 \pm 1.7\%$ ($n = 1425$) when data for samples deeper than 2000 m are used (data compiled by S. Honjo and R. Francois; http://www1.who.edu/datasys/SMP/Honjo/sed_traps.htm). POC contents of resuspended sediments are expected to have high spatial variability. They are likely to be generally higher than those of underlying sediments because fine-grained particles exhibit higher carbon loadings and low-density organic matter-mineral aggregates are likely to be preferentially resuspended relative to other components such as calcareous or siliceous biogenic debris [Thomsen and Gust, 2000]. For example, the OC content of the clay ($<3\mu\text{m}$) fraction (OC, 3–6%) in Washington margin sediments is higher than those of corresponding bulk sediment (1–3%) because of their higher mineral surface areas [Mayer, 1994; Keil et al., 1994a; Keil and Cowie, 1999; Coppola et al., 2007]. Similarly, the OC content of suspended particles collected within 1 m of the seafloor in the Gulf of Maine [Townsend et al., 1992], where sediment resuspension is prominent [Bothner et al., 1981; Pilskaln, 2006], was $4.0 \pm 1.6\%$ ($n = 19$); this is close to that of sinking POC but significantly higher than those of underlying sediments (1–2% [Charette et al., 2001]). If POC contents in resuspended sediments are comparable to those in sinking particles, a virtually linear correlation would be obtained between normalized $\Delta^{14}\text{C}$ values and Al content.

[14] Data from the deep Canada Basin exhibiting the highest Al concentrations fall below the linear mixing line

because of extremely low normalized $\Delta^{14}\text{C}$ values and/or low POC concentrations of the resuspended sediment end-member. The data from the shallow Canada Basin exhibit more scatter than at other locations, potentially because the trap drifted over widely different water depths from a few hundreds to thousands of meters [Macdonald et al., 2002], intercepting resuspended sediment that varies in provenance [Honjo et al., 2010].

[15] Dissolved Al scavenged to particles [Murray and Leinen, 1996] and eolian inputs constitute other potential

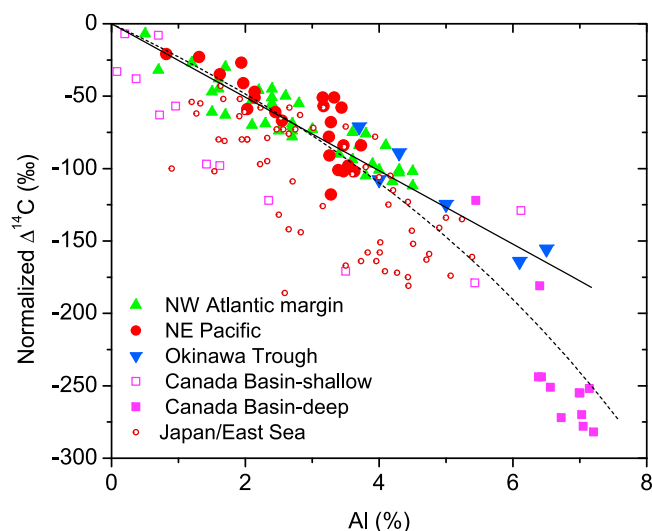


Figure 2. Correlation between normalized $\Delta^{14}\text{C}$ values of POC and aluminum contents in sinking particles. The solid line represents an example of two end-member mixing between recently produced organic matter ($\Delta^{14}\text{C} = 0\text{‰}$, POC = 5%) and aged OC associated with resuspended sediment ($\Delta^{14}\text{C} = -182\text{‰}$, POC = 5%). The dotted line represents a mixing of fresh organic matter with organic matter of an older age and a lower POC content ($\Delta^{14}\text{C} = -274\text{‰}$, POC = 3%, see text regarding the choice of these values). As the POC concentration deviates from the fresh POC value (5%) toward lower values, the line becomes more convex.

sources of particulate Al. Contributions from scavenged Al are locally important only when particulate Al contents are extremely small, such as in the Equatorial Pacific [Murray and Leinen, 1996]. Collier *et al.* [2000] used the ratio between Ti and Al of sinking particles in the Ross Sea to show that absorbed Al in the water is insignificant at that location. Eolian input can be important locally and seasonally. For example, locations proximal to continents and under the trajectory of major dust tracts, such as Okinawa Trough and Japan/East Sea, receive significant Al fluxes from atmospheric deposition, with annual average inputs exceeding $2 \text{ mg m}^{-2} \text{ d}^{-1}$ [Duce *et al.*, 1991; Measures and Vink, 2000]. Eolian input of Al may account for up to 7–15%, 12%, 20–39%, 20–50% of the observed Al flux in the NW Atlantic (New England slope), northeast Pacific off the coast of California, Okinawa Trough, and Japan/East Sea, respectively (estimated from the data by Measures and Vink [2000], Minakawa and Watanabe [1998], and Suzuki and Tsunogai [1988]). It is challenging to assess the influence of eolian supply of OC on sinking POC because there is no equivalent data on eolian supply of OC at the study sites. However, considering that eolian supply of OC comprises less than 1% of in situ production in surface waters, even on the continental margins proximal to the major sources of eolian dust, OC delivered by this mode of supply is likely to be insignificant. Studies of the biogeochemical and C isotopic properties of aerosol OC [e.g., Gustafsson *et al.*, 2009] and on the processes that influence the fate of eolian OC following deposition to the surface ocean will be necessary to further address this issue. The normalized $\Delta^{14}\text{C}$ value of sinking POC could either be lowered or raised depending on the $\Delta^{14}\text{C}$ value of eolian POC. This additional OC source is likely to cause deviation from, or higher scatter in relation to the mixing line, such as is observed for data from the Japan/East Sea [Otosaka *et al.*, 2008]. However, for the seasonally ice-covered Arctic Ocean regions including portions of the central Canada Basin [Hwang *et al.*, 2008], this source is expected to be negligible. Even when ice started to melt and release dust accumulated on the ice (suggested by an abrupt increase in normalized $\Delta^{14}\text{C}$), Al content virtually remained unchanged [Hwang *et al.*, 2008].

[16] The y intercept of the trend line (i.e., when Al content is zero) should indicate the influence of processes other than the incorporation of resuspended sediment and potentially, eolian dust. Excluding the Canada Basin and Japan/East Sea data, the y intercept of the linear regression is $-8 \pm 5\%$, which is not significantly different from 0‰ considering the uncertainty of $\Delta^{14}\text{C}$ measurements and the normalization method. This implies that incorporation of DIC and DOC into sinking POC via microbial or physicochemical processes may be smaller than previously suggested [Rau *et al.*, 1986; Druffel and Williams, 1990; Pearson *et al.*, 2001; Hwang *et al.*, 2006; Ingalls *et al.*, 2006]. The y intercept also enables upper limits to be placed on the contribution from each potential source. To provide estimates for the upper limits, a 20‰ decrease in normalized $\Delta^{14}\text{C}$ (a conservative uncertainty for seasonal variability of DIC $\Delta^{14}\text{C}$), was used for the overall effect of these other sources. In a model case where the $\Delta^{14}\text{C}$ value for freshly produced POC is +60‰, that for deep water DIC is between -80 and -220‰, and that for deep

DOC is between -400 and -550‰ [Druffel *et al.*, 1992; Beupr e and Druffel, 2009], the upper limits for the contribution from DIC and DOC to sinking POC are between 7 and 14%, and between 3 and 4%, respectively. The contribution of each individual process is likely smaller than these values considering that incorporation of both DIC and DOC may occur simultaneously. However, it should also be noted that isotopic heterogeneity of DOC [Loh *et al.*, 2004] may cause uncertainty in this estimation. For example, if bacteria preferentially uptake young DOC components [Cherrier *et al.*, 1999], this might imply that estimates of DOC incorporation onto POC are too low.

[17] Acknowledging that the data coverage is currently very sparse and that additional measurements are needed, we derive a tentative estimate of the global contribution of resuspended sediment to sinking POC using the average POC flux-weighted decrease in normalized $\Delta^{14}\text{C}$ (95‰). The assignment of a global average $\Delta^{14}\text{C}$ value for the resuspended sediment end-member represents a major source of uncertainty, and ideally, this should be determined for each specific location. Here, we used ^{14}C measurements on core top sediment (0–1 cm) at 24 locations on the NW Atlantic margin that range in water depth from 280 and 3865 m in order to derive an average $\Delta^{14}\text{C}$ value ($-214 \pm 59\%$) that we adopted as representative for resuspended sediment. Based on this value, the 95‰ difference in $\Delta^{14}\text{C}$ between flux-weighted POC and average surface water DIC (60‰) implies that 35% ($\pm 13\%$ propagated error) of sinking POC is derived from resuspended sediment. This may represent a minimum value because the sedimentary component that is prone to resuspension is composed of fresher, fluffy organic matter-rich aggregates and other detrital materials residing at or near the sediment-water interface that would likely have higher $\Delta^{14}\text{C}$ values [Wang *et al.*, 1996]. This inference is supported by the observation that NW Atlantic margin, NE Pacific, Okinawa Trough data are well represented by the solid mixing line in Figure 2, with an end-member $\Delta^{14}\text{C}$ value of -182‰ (corresponding to the unnormalized $\Delta^{14}\text{C}$ value of -122‰), significantly higher than -214‰. If this is the case, we estimate that the contribution of resuspended sediment could be greater than 50%. In either case, the results imply that the current estimate of export production based on direct POC flux measurements is biased toward higher values.

[18] Our findings show that resuspension of sediment and subsequent lateral advection is widespread in the ocean, especially over, or proximal to, the continental margins. Lithogenic particles originating from the continent are transported to the continental margin and further into the deep ocean interior via hydrodynamic processes. This phenomenon, termed “hemipelagic sedimentation” [Rea and Hovan, 1995], is important for the supply of minerals to the ocean basins. Our observation that lithogenic particles are tightly associated with aged POC implies that the role of hemipelagic sedimentation in POC cycling should be reevaluated and better quantified, especially with respect to carbon export from the continental shelves to the interior ocean. Export of DIC alone has been described in the context of the continental shelf pump hypothesis [Tsunogai *et al.*, 1999]. It seems clear that we must also consider the continental shelf pump in terms of POC (and likely DOC) export.

[19] Another important implication of our results concerns the role of lithogenic particles in vertical transport of POC [Dunne *et al.*, 2007]. Calcium carbonate has been found to be the major ballast mineral of POC in open ocean settings [François *et al.*, 2002; Klaas and Archer, 2002], whereas lithogenic particles have hitherto been considered to play only a minor role, primarily because lithogenic fluxes in the deep sea are small [François *et al.*, 2002]. However, this paradigm may not hold for margin-proximal sites with abundant sources and higher fluxes of lithogenic particles, and where normalized $\Delta^{14}\text{C}$ values are low. The high POC fluxes that typify margin settings may be supported to a significant degree by laterally supplied lithogenic particles and associated OC. Finally, the biogeochemical character of OC entrained with resuspended sediment likely differs from that of freshly produced POC because it has experienced more extensive degradation and because of its intimate association with the mineral surfaces [Keil *et al.*, 1994a; Mayer, 1994; Keil and Cowie, 1999; Coppola *et al.*, 2007]. These differences may influence its fate during transport and sedimentation, and the biogeochemical properties of resuspended sedimentary OC may be further altered by water column processes. Incorporation of these factors into our understanding of the oceanic carbon cycle will require models that accommodate lateral POC transport as well as vertical settling of POC to the deep ocean, and will also need to account for the potentially different OC loading and reactivity of these inputs.

5. Summary and Conclusions

[20] Numerous prior studies have provided evidence for the resuspension and subsequent lateral transport of sediments in a range of marine environments, however, the ubiquity and overall significance of this process in terms of organic matter cycling in the oceans has remained unclear. In this study, new insights have been gained from bringing together disparate observations and data sets on the contents of Al and ^{14}C within sinking particles intercepted by sediment traps. The relationship between Al content and normalized $\Delta^{14}\text{C}$ is widespread and the overall slope of the relationship is remarkably constant, indicating that this phenomenon is global and consistent over a wide range of oceanic settings. The near-zero y intercept for the mixing lines implies that aged organic carbon associated with lithogenic material from sediment resuspension is responsible for the observed low normalized $\Delta^{14}\text{C}$ values (as opposed to other processes such as incorporation of dissolved inorganic carbon or dissolved organic carbon into POC at depth). Using a compilation of data sets of $\Delta^{14}\text{C}$ and POC flux, we estimate that 35% of sinking POC may derive from resuspended sediments globally; this proportion may be even higher depending on the choice of isotopic end-member for this calculation.

[21] Our results suggest that sediment remobilization and redistribution is an important component of the oceanic carbon cycle and should be considered in models of oceanic carbon export and burial. Given that particles intercepted by sediment traps have mostly been assumed to originate from the overlying water column, these findings imply that the

vertical component of the flux is overestimated whereas continental margin export is underestimated. The role of the continental margins in exporting POC to the interior ocean should be carefully reevaluated and better quantified. In addition, the influence of sediment remobilization processes on the biogeochemical properties of resuspended POC and its consequences for the bathypelagic food web and for OC burial in deep ocean sediments requires further examination.

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