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Seasonal and decadal shifts in particulate organic matter processing and sedimentation in the Bering Strait Shelf region

Lee W. Cooper *[,1], Catherine Lalande [2], Rebecca Pirtle-Levy [3], I.L. Larsen [4], Jacqueline M. Grebmeier [1]

Marine Biogeochemistry and Ecology Group, Department of Ecology and Evolutionary Biology, University of Tennessee, 10515 Research Drive, Suite 100, Knoxville, TN 37932, USA

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ABSTRACT

We present data on the quality and quantity of particulate organic material deposited to the benthos in the Chukchi Sea. This analysis is undertaken by using ^{7}Be, a short-lived radiotracer, which is associated with particle deposition, the stable carbon isotopic composition of organic material and its C/N ratio in the water column and within the sediments, and the inventories of chlorophyll *a* present in surface sediments. Using previously published data, we show that sedimentation processes in the regional Bering Strait ecosystem may have shifted in the past decade. Surface sediments collected in 2004 adjacent to the Russian coastline in the Chukchi Sea are less refractory in terms of carbon isotope ratios and C/N ratios than was observed for surface sediments at similar locations in 1995 and 1988. Based upon sediment ^{7}Be and chlorophyll *a* inventories, short-term sedimentation on the shelf occurs immediately north of Bering Strait, and within and downstream of Barrow and Herald Canyons. Seasonal differences (i.e., ice-covered versus open-water conditions) in the quality of particulate organic carbon reaching the benthos appear to be small in the most productive waters, such as Barrow Canyon. However, in less productive waters, C/N ratios and δ^{13}C values show seasonal variations. Once on the bottom, δ^{13}C values in the organic fractions of the sediments are less negative than observed in settling material in the water column, which is commonly thought to result from biological processing within the sediments.

1. Introduction

The Pacific-influenced waters that pass northward over the shallow continental shelves of the northern Bering and Chukchi Seas are utilized and influenced by underlying rich benthic faunal communities as well as modified as a result of biological processes within the water column (Grebmeier et al., 2006b). Past studies have shown that organic sedimentation is highly variable seasonally in this region, with maximal organic fluxes shortly after the dissolution of seasonal ice cover (Cooper et al., 2002). Organic sedimentation is also regionally variable with high localized deposition from waters with high productivity and nutrient burdens (Grebmeier et al., 1988). While the large retreat of seasonal Arctic sea-ice coverage in this decade (e.g., Serreze et al., 2003; Stroeve et al., 2005) has stimulated interest in how this productive ecosystem will respond to possibly irreversible changes in the seasonal sea-ice regime, understanding ecosystem response as a whole has been hampered because ecosystem and oceanographic studies cannot be carried out routinely on both sides of the political boundary shared by the United States and Russia in the Amerasian Arctic. The Russian–American Long-term Census of Marine Life (RUSALCA) is the first multidisciplinary collaboration between US and Russian scientists in more than a decade in the Chukchi Sea. This bilaterally supported field sampling program in 2004 was undertaken at the same time as several biologically oriented research programs in US waters including the Western Arctic Shelf–Basin Interactions (SBI) and the Bering Strait Environmental Observatory (BSEO). Therefore, the combination of data from the separate sources provided an unusual opportunity for ecosystem evaluation across the international boundary.

This focused sampling over much of the Chukchi Sea in 2004 followed a significant reduction in summer sea-ice extent over the past decade, so the data collected in 2004 are potentially useful for shelf-wide evaluation of Chukchi Sea ecosystem status relative to past trans-boundary evaluations (Walsh et al., 1989; Grebmeier, 1993; Naidu et al., 2000; Khim et al., 2003). In one particular

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* Corresponding author.
E-mail addresses: cooper@cbl.umces.edu (L.W. Cooper), catherine.lalande.1@ulaval.ca (C. Lalande), rspirtle@ncsu.edu (R. Pirtle-Levy), jgrebmei@cbl.umces.edu (J.M. Grebmeier).

[1] Current address: Chesapeake Biological Laboratory, University of Maryland Center for Environmental Science, 1 Williams St., PO Box 38, Solomons, MD 20688, USA.

[2] Current address: Québec-Océan, Université Laval, Québec, Canada G1V 0A6.

[3] Current address: Marine, Earth and Atmospheric Sciences Department, North Carolina State University, Raleigh, NC 27695-8208, USA.

[4] Deceased, 9 June 2008.

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follow-up to our 2004 sample analyses, we investigated how recent seasonal sea-ice retreat and/or other hydrographic changes might have influenced the timing and dynamics of organic carbon sedimentation and benthic biogeochemical processes in sediments last sampled in 1995 and prior to that in 1988 during past joint Russian–US research.

The biogeochemical indicators we chose to use in this study of organic sedimentation have proven useful in previous work on this highly productive continental shelf (e.g., Grebmeier and Cooper, 1995; Cooper et al., 2002, 2005) in defining locations with recent sedimentation of organic matter that influence the productivity of the underlying benthic ecosystem. Specifically, the distributions of the short-lived radionuclide ^{7}Be ($t_{1/2} = 53\,\mathrm{d}$) in surface sediments were evaluated to identify where particles accumulated on the shallow continental shelf following ice retreat, as well as in deeper slope waters where it was detectable. We determined the inventories of the radionuclide in the snow on the sea-ice surface prior to melt, estimated its flux rates through the water column using short-term drifting sediment traps and then measured inventories that reached the sea floor following a transition to almost completely ice-free conditions by July–August 2004. The distribution of ^{7}Be reflects deposition of particulate materials in snow on the sea-ice surface and sedimentation during ice melt, at least early in the summer (Grebmeier and Cooper, 1995; Cooper et al., 2002, 2005), providing an indication of the fate of particles released from receding ice cover. Because of the short half-life of this cosmogenic isotope and its atmospheric source, sediment and water-column inventories decline below detectable levels by the end of each sea-ice covered period and the retreat of sea ice initiates a new annual cycle of contributions of the radioisotope to the marine system.

We also used surface inventories of chlorophyll *a* on the sediments as a marker to follow sedimentation of freshly produced particulate organic matter before and following ice retreat as well as during open-water periods. Active chlorophyll does not have a precise half-life in surface sediments such as a short-lived radionuclide, but it is directly tied to biological activity in the water column, unlike ^{7}Be, which may be deposited to the sediments through attachment to minerals and other non-biogenic particles released from sea ice (Cooper et al., 2005). The persistence of chlorophyll *a* and its pattern of distribution spatially and vertically in sediments is becoming better understood on the Chukchi shelf (Clough et al., 2005), and more results from the SBI program have been recently summarized elsewhere (Pirtle-Levy et al., 2009).

In-sediment processing of organic carbon deposited to the sea floor was evaluated by measuring elemental C/N ratios and the stable carbon isotope composition of organic materials in surface sediments and by comparing these data to C/N ratios and δ^{13}C values of particles collected into drifting sediment traps. These analyses were also used as the basis for a retrospective comparison of the surface sediment composition in 2004 (data presented for the first time here) with surface sediment composition sampled in 1988 (Naidu et al., 1993, 1995; Cooper et al., 1998; Khim et al., 2003).

2. Methods

2.1. Sediment and drifting sediment trap sediments

Surface sediment samples (0–1 cm) for ^{7}Be, chlorophyll *a*, and the C/N ratios and the δ^{13}C values of the organic fraction were collected on cruises of the USCGC *Healy* (15 May–23 June 2004 and 16 July–24 August 2004; SBI Project), R/V *Professor Khromov* (8–24 August 2004; RUSALCA Project) and the CCGS *Sir Wilfrid Laurier* (8–22 July 2004; Bering Strait Environmental Observatory Project) using a multi- or single-HAPS benthic corer (133 cm^2; Kanneworff and Nicolaisen, 1973) or from a van Veen grab (0.1 m^2). Sampling followed tests to determine the conditions under which disturbance of surface sediments by the van Veen grab affects results relative to coring. These tests are documented in detail elsewhere (Cooper et al., 1998; Pirtle-Levy, 2006), but, briefly, we compared surface sediment parameter values (^{137}Cs and sediment chlorophyll) collected with both cores and grabs at the same locations on the Bering and Chukchi continental shelves. Using these paired sets of data, we analyzed the results to determine if there was any systematic difference between the results from grabs and cores, or if bioturbation was such a dominant sediment process that the less-disturbed surface sediments collected with cores were nevertheless equally well-mixed as a result of bioturbation. We determined that there was in most cases no significant difference in surface chlorophyll *a* inventories or of activities of the anthropogenic clay particle-associated radionuclide ^{137}Cs ($t_{1/2} = 30.2\,\mathrm{y}$) for paired surface sediment sampling undertaken using both devices (cores and grabs) at the same sites in waters shallower than 200–500 m (Cooper et al., 1998; Pirtle-Levy, 2006). In other words, because of apparent high rates of bioturbation on these productive shelves, sediment chlorophyll *a* inventories from the surface of grab samples from shallow depths were not significantly different from chlorophyll *a* inventories in surface sediments collected using coring devices that disturb surface sediments to a lesser extent.

Samples for sediment trap ^{7}Be, chlorophyll *a*, C/N ratios, and δ^{13}C samples were collected at nine stations from drifting sediment traps (KC Denmark, Silkeborg) deployed at five depths (30, 40, 50, 60 and 100 m) for periods ranging from 11–20 h during the two cruises on *Healy* in 2004 mentioned above (more information at http://www.eol.ucar.edu/projects/sbi/). The first cruise was almost entirely under ice cover (May–June 2004) and second one was under largely open-water conditions (July–August 2004). More details on deployments, including locations and duration of collection are reported in Lalande et al. (2007a, b).

2.2. Snowpack and precipitation collections

Samples for ^{7}Be measurements in snowpack on the sea-ice surface were collected only during the first *Healy* spring cruise using a 625-cm^2 quadrat when snow was present on the sea-ice surface.

Precipitation was collected in duplicate using open precipitation collectors mounted on the ship's flying bridge during the two *Healy* cruises to determine the steady-state atmospheric flux (both wet and dry precipitation) of the radionuclide during the sampling period. At the end of the cruises, both snow samples from the sea-ice surface and the duplicate precipitation samples were heated to reduce volumes in order to fit within calibrated containers. Dilute hydrochloric acid was used as a rinse during the transfer of samples to keep ^{7}Be in solution.

2.3. Analysis

All samples for ^{7}Be were measured using a Canberra GR4020/S reverse electrode closed-end coaxial detector at the University of Tennessee. Due to low activities, we pooled individual GF/F filters from each sediment trap depth (30, 40, 50, 60 and 100 m) into a single 15-cm^3 Petri dish for gamma spectroscopy and we thus report data for ^{7}Be collected at each sediment trap station collectively (all other sediment trap data was separated by depth). Activities reported are based upon the proportions of water in each trap that were mixed well and then filtered relative to the total volume (and surface area) of all traps that were deployed at each station. The total volume of water in all traps deployed was 36 L, and these traps intercepted 0.0814 m^2 of planar ocean area so the fraction of ocean area intercepted (0.0814 m^2) was calculated to be proportional to the fraction of 36 L filtered.

Surface sediment determinations of ^{7}Be were made on samples packed into 90-cm^3 cans or 500-mL Marinelli beakers. The beakers were used during the *Professor Khromov* (RUSALCA) cruise in Russian waters, where sediment collections were solely made out of the top of the van Veen grab before it was opened. Because quantification of the surface area of sediments collected from a van Veen grab is problematic, all sediments were dried after gamma spectroscopy and weighed to establish activities of ^{7}Be on a dry weight basis for inter-comparison of the 2004 data. In addition, to facilitate comparisons with previously published ^{7}Be inventory data from the Chukchi Sea (Cooper et al., 2005) inventories were also determined on a square meter basis, for those sediments collected on the *Healy* cruise by sediment coring rather than by a van Veen grab. Corrections for efficiency and calibrations for all samples were made prior to counting with a mixed gamma standard traceable to the National Institute for Standards and Technology. Background corrections and control samples were analyzed prior to counting to verify detector performance. All samples were analyzed within two half-lives of the date of collection and sediment trap and sediment data reported have been decay-corrected to the date of collection.

Surface sediment chlorophyll *a* inventories were measured using a Turner Designs fluorometer without acidification using a standardized method that includes a 12-h dark incubation in 90% acetone at 4 °C (Cooper et al., 2002). Surface sediment inventories reported are the mean of two independent determinations. The sediment trap inventories of chlorophyll *a* also were measured and have been reported elsewhere (Lalande et al., 2007a).

$^{13}C/^{12}C$ ratios were measured on both the organic carbon fraction of sediment trap material collected on Whatman GF/F filters and the organic fraction of surface sediments. De-carbonation was achieved by exposing the GF/F filers to fuming concentrated HCl for 24 h in a dessicator (Lalande et al., 2007a). Sediments were de-carbonated using 1N HCl (2 mL g^{-1} of sediment) at 105 °C for 12 h (Grebmeier et al., 1988). Samples were combusted off-line and cryogenically purified prior to analysis using a VG Instruments SIRA Series II dual inlet stable isotope mass spectrometer at the University of Tennessee. Precision of the $\delta^{13}C$ values reported, based upon replicate analyses of an internal sediment standard, was ± 0.2 per mille. C/N ratios were also determined with de-carbonated sediments on an elemental analyzer (Exeter Analytical CEC 440HA) at the Marine Science Institute Analytical Laboratory, University of California, Santa Barbara.

2.4. Retrospective comparisons

Since no samples of the types used in this study had been collected in nearly a decade in the Russian waters that were sampled in 2004, a retrospective analysis was included in our study to compare results obtained with previous work. The sediment parameters that were available to compare were data on the stable carbon isotope composition of organic carbon and C/N ratios of the organic fraction of surface sediments. These prior data were the result of collections on the 189th cruise of the R/V *Alpha Helix* (HX 189) in the Russian sector of the Chukchi Sea and the East Siberian Sea in August–September 1995 and on the Third Joint US–USSR Bering and Chukchi Seas Expedition (BERPAC) in 1988 (Nagel, 1992). These previously published data (Cooper et al., 1998; Khim et al., 2003; Grebmeier et al., 2006b) were collected and processed using the same methods as our sampling in 2004. In particular, the 1995 samples were collected, as in 2004, using an identical van Veen grab, and prepared using the same methods and analyzed with the same stable isotope mass spectrometer, internal standards and sample preparation protocols (Cooper et al., 1998; Khim et al., 2003; Grebmeier et al., 2006b).

3. Results

Inventories of ^{7}Be collected in snow ranged from not detectable to as high as 68 Bq m^{-2} (Table 1). Deposition of ^{7}Be measured in duplicate (side-by-side) precipitation collectors was higher (25–42 Bq m^{-2}) than most steady-state inventories present in the snow on the sea-ice surface (Table 1). Inventories of ^{7}Be that reached the sea floor by the time of the July–August cruise ranged from undetectable to 48 Bq m^{-2} (Table 2). No ^{7}Be was detected in sediment trap material collected under sea-ice cover or in surface sediments collected under ice cover during May–June 2004. However, fluxes of ^{7}Be in the upper 100 m under largely open-water conditions in July–August 2004 ranged from 1 to 5 Bq m^{-2} d^{-1}, albeit with significant counting errors due to low activity (Table 3).

On a geographical basis, recent sedimentation as indicated by ^{7}Be distributions on the sea floor in July–August 2004, was focused on the Chukchi outer shelf, on the Russian Chukchi Shelf immediately north of Bering Strait, and in deeper deposition zones in Barrow Canyon (Fig. 1) with activities on a dry weight basis of up to 10.6 Bq kg^{-1} (Fig. 1). The radioisotope was only detected one time in seven stations along the EHS and WHS transects lines from shelf to basin in the Chukchi Sea, but in two other transect lines, one in the Barrow Submarine Canyon, and along another transect line immediately to the east in the Beaufort Sea, the radioisotope was consistently detected in surface sediments as deep as 2000 m (Table 2).

Another indicator of recent sedimentation, chlorophyll *a*, varied seasonally with significant increases in mean inventories observed at 18 stations occupied both before and after ice dissolution, primarily on the Chukchi outer continental shelf following sea-ice retreat (two-tailed *t*-test, $p<0.009$; Figs. 2 and 3; see Pirtle-Levy, 2006 for additional details on seasonal differences). For both organic materials settling in sediment traps, as well as surface sediments, higher C/N ratios are significantly correlated with more negative $\delta^{13}C$ values (Fig. 4).

Most of these raw data reported here and used in our analysis are freely available in a public data archive (http://www.eol.ucar.edu/projects/sbi).

As outlined in Section 2.4, our retrospective comparison of these 2004 results presented here was made with previously published data on the carbon isotope composition and C/N ratios of the organic fraction of sediments collected in 1988 and 1995. Both the 1988 (BERPAC) and 1995 (*Alpha Helix* 189) cruises occupied stations on the Russian Chukchi Shelf that are close to or adjacent to those occupied in 2004 (Fig. 5; Table 4). The more recently measured $\delta^{13}C$ values and C/N ratios have unambiguously and systematically shifted (Figs. 5 and 6) for several sampling locations adjacent to Chukotka and in Russian waters, although elsewhere not enough sampling resolution is available to verify any shift. Where sampling was close enough to compare, the change represents a decline of ~1 per mille for $\delta^{13}C$ values and a decline observed in C/N ratios was from ~6 to ~5. These changes indicate that surface sediment samples collected in 2004 near the Russian coastline were consistently less refractory than samples collected at nearby stations in 1988 and 1995.

Table 1
Inventories of ^{7}Be in snow on the sea-ice surface during the early season cruise of the USCGC *Healy*, May–June 2004, and on-going inventories to the sea surface as measured in shipboard precipitation collectors, May–June 2004 and July–August 2004.

Location	Activity, ^{7}Be (Bq m^{-2})$\pm 1\sigma$ at date of collection	Date of collection	Activity, ^{7}Be (Bq m^{-2}) corrected to 5 August 2004
67.50°N, 168.91°W	17.81 ± 3.06	18 May 2004	6.34
67.50°N, 168.91°W	15.94 ± 2.95	18 May 2004	5.67
72.01°N, 159.85°W	17.38 ± 3.01	24 May 2004	6.69
72.01°N, 159.85°W	15.82 ± 2.94	24 May 2004	6.09
72.08°N, 159.64°W	2.91 ± 6.26	26 May 2004	1.15
72.08°N, 159.64°W	5.55 ± 2.05	26 May 2004	2.19
72.70°N, 158.81°W	18.91 ± 3.21	30 May 2004	7.87
72.73°N, 158.46°W	67.87 ± 3.99	31 May 2004	28.63
72.73°N, 158.46°W	14.40 ± 2.44	31 May 2004	6.07
72.90°N, 158.26°W	Not detected	2 June 2004	Not detected
72.90°N, 158.26°W	10.02 ± 2.13	2 June 2004	4.34
73.13°N, 157.79°W	9.33 ± 1.99	4 June 2004	4.15
72.12°N, 154.68°W	7.54 ± 3.94	13 June 2004	3.77
72.28°N, 154.61°W	4.01 ± 1.42	15 June 2004	2.06
Precipitation inventory	26.86 ± 2.15	18 May–21 June 2004	–
Precipitation inventory	24.65 ± 3.21	18 May–21 June 2004	–
Precipitation inventory	38.57 ± 2.57	18 July–24 August 2004	–
Precipitation inventory	41.93 ± 2.59	18 July–24 August 2004	–

Inventories of ^{7}Be in snow on the sea-ice surface were decay corrected to 5 August 2004, the median date of July–August cruise to allow for comparisons with ^{7}Be inventories observed in surface sediments during the July–August cruise.

Table 2
Inventories of ^{7}Be in surface sediments collected from 0 to 1-cm increment of HAPS corer on USCGC *Healy* cruise HLY 04–03 in July–August 2004 in Chukchi Sea.

Station name, coordinates	^{7}Be (Bq m^{-2})$\pm 1\sigma$	Date of collection	Water depth (m)
BC2, 71.44°N, 159.27°W	1.76 ± 0.82	23 July 2004	122
BC3.2, 72.35°N, 156.92°W	22.12 ± 10.26	24 July 2004	126
BC4, 71.95°N, 155.89°W	$14.83 \pm 8,76$	26 July 2004	326
BC5, 71.93°N, 154,72°W	6.61 ± 9.51	27 July 2004	960
BC6, 72.18°N, 153.92°W	15.21 ± 15.21	26 July 2004	1914
EB1, 71.29°N, 152,54°W	30.34 ± 9.16	29 July 2004	52
EB2, 71.54°N, 152.44°W	47.87 ± 10.96	30 July 2004	118
EB3, 72.18°N, 152.81°W	7.83 ± 10.42	30 July 2994	156
EB6, 71.96°N, 152.14°W	12.67 ± 5.87	4 August 2004	2124
EB4, 71.65°N, 152.37°W	11.99 ± 7.36	6 August 2004	578
EHS1, 72.36°N, 159.17°W	n.d.	10 August 2004	49
EHS4, 72.65°N, 158.50°W	n.d.	11 August 2004	91
EHS5, 72.71°N, 158.45°W	24.79 ± 17.53	12 August 2004	219
EHS6, 72.83°N, 158.23°W	n.d.	13 August 2004	398
EHS7, 72.90°N, 158.30°W	n.d.	14 August 2004	1077
EHS9, 73.05°N, 157,96°W	n.d.	15 August 2004	1938
WHS8, 73.90°N, 157,85°W	n.d.	19 August 2004	3760
WHS6, 73.48°N, 159.61°W	n.d.	19 August 2004	2110

Data are decay-corrected to date of collection.
n.d. = Not detected.

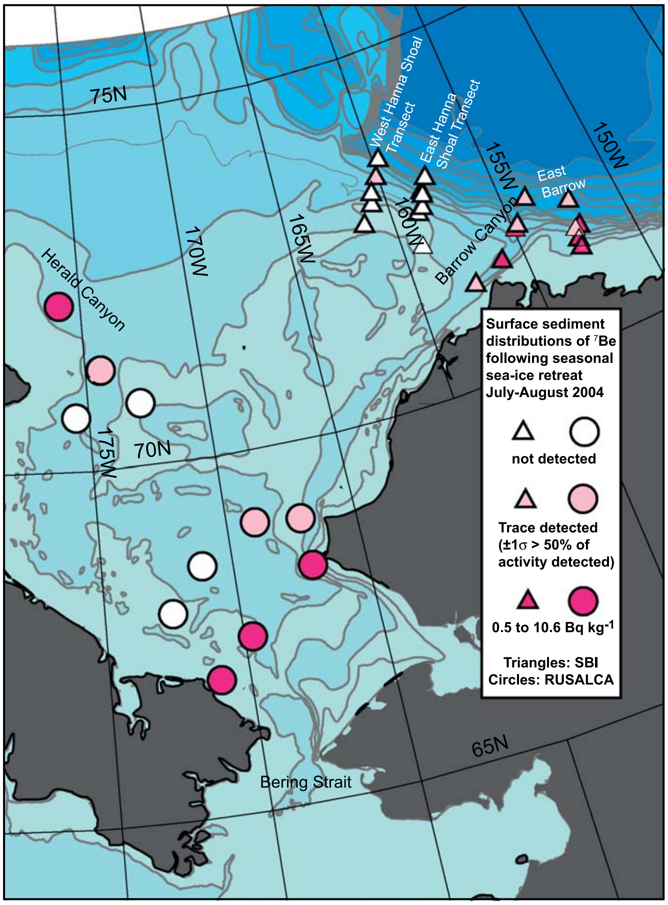
4. Discussion

The distributions of chlorophyll *a* and ^{7}Be in surface sediments following sea-ice retreat bear similarities to results obtained in field studies undertaken in the Chukchi Sea in 2002 (Cooper et al., 2005). As in this previous work, ^{7}Be was largely undetectable in sediments under sea-ice cover. The radioisotope was also not detected in any sediment trap collections made under ice cover during the May–June *Healy* cruise. However, following ice retreat it was detected in the surface sediments of the Herald and Barrow Submarine Canyons and particularly downstream of Barrow Canyon, as well as in surface sediments immediately north of Bering Strait (Fig. 1) where currents slow down and particulate matter tends to settle (Grebmeier et al., 2006b). This geographical pattern is similar to the 2002 sampling, and suggests that particle sedimentation zones indicated by ^{7}Be and chlorophyll *a* likely persist inter-annually. Total inventories of ^{7}Be in surface sediments where it was detectable during the open-water period were similar in magnitude to the decay-corrected inventories that had been present in snow on the sea-ice surface prior to melt (Tables 1 and 2). This indicates that a significant fraction of the radionuclide present in snow and ice on the sea surface prior to melt reached surface sediments within one-to-three months following ice retreat. The ^{7}Be fluxes from the sediment traps (Table 3), while somewhat problematic because of low activities and proportionally high counting errors, appeared to be consistent with the snow inventories, precipitation fluxes divided by the duration of sampling, and bottom sediment steady-state inventories. Although ^{7}Be deposition is not always tightly linked with other sedimentation indicators including surface sediment chlorophyll *a*

Table 3
Fluxes of ^{7}Be calculated from sediment trap collections during two cruises of the USCGC *Healy* in May–June 2004 under ice cover and July–August 2004 under largely open-water conditions.

Cruise	Ice conditions	Station name	^{7}Be (Bq m^{-2} d^{-1})	$\pm 1\sigma$
May-June	Ice cover	BC4	n.d.	–
	Ice cover	BC5	n.d.	–
	Ice cover	EHS4	n.d.	–
	Ice cover	EHS5	n.d.	–
	Ice cover	EHS6	n.d.	–
July-August	Open water	BC4	1.26	1.15
	Open water	BC6	3.87	1.81
	Open water	EHS5	5.22	3.71
	Open water	EHS6	3.18	0.97
	Open water	EHS9	1.42	1.23

Due to low activities, filters were combined from collections made at five separate depths (30, 40, 50, 60, 100 m). Separate depth analyses were made of other variables (e.g. δ^{13}C, C/N ratios, POC, chlorophyll) and are discussed in Lalande et al (2007a, b).



Surface sediment distributions of ^{7}Be following seasonal sea-ice retreat July-August 2004

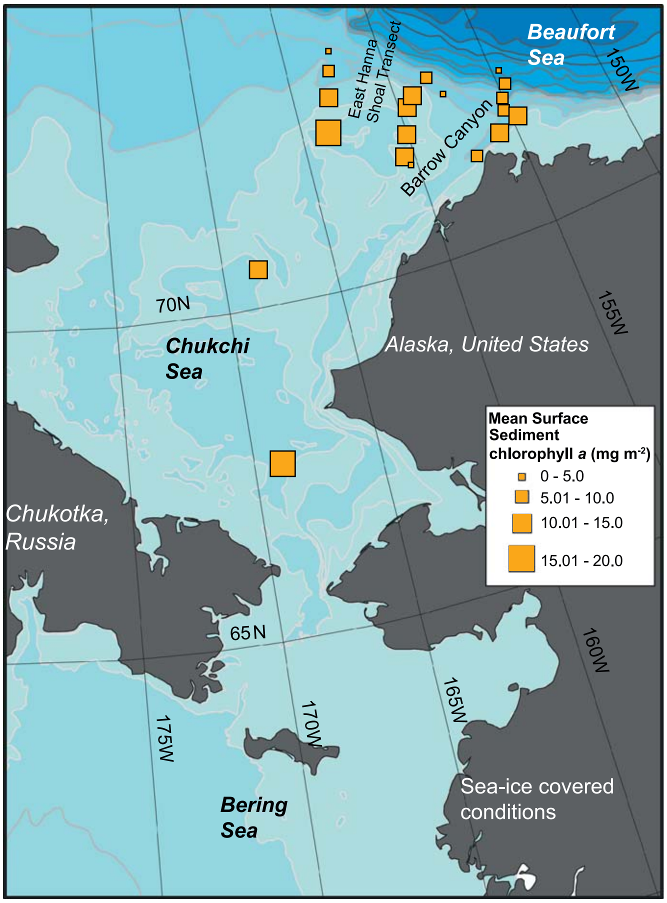
△ ○ not detected

△ ○ Trace detected (±1σ > 50% of activity detected)

▲ ● 0.5 to 10.6 Bq kg^{-1}

Triangles: SBI
Circles: RUSALCA

Fig. 1. Distributions of ^{7}Be in surface sediments on the Chukchi Shelf, July–September 2004, with circles representing samples collected on R/V *Professor Khromov* (August–September) and triangles representing samples collected on USCGC *Healy* (July–August). Following the convention of Cooper et al. (2005), ^{7}Be inventories were simply categorized as undetected (white symbols), trace inventories (shaded pink symbols) where counting errors exceeded 50% of the activities detected, and detectable inventories (dark pink symbols), where activities ranged from 0.5 to 10.6 Bq kg^{-1} with counting errors smaller than 50% of activities detected.



Mean Surface Sediment chlorophyll *a* (mg m^{-2})

■	0 - 5.0
■	5.01 - 10.0
■	10.01 - 15.0
■	15.01 - 20.0

Fig. 2. Inventories of chlorophyll *a* present in surface sediments at the end of the ice-covered season, May–June 2004. Sampling was from USCGC *Healy*.

inventories (Cooper et al., 2005), its detection in slope sediments downstream of Barrow Canyon is consistent with other indications of down-canyon and offshore transport that have been observed for plankton (Ashjian et al., 2005), particulate organic carbon fluxes derived from ^{234}Th (Moran et al., 2005) and benthic biomass and sediment oxygen demand (Grebmeier et al., 2006b).

In previous work in this study area in 2002, sediment chlorophyll *a* inventories dramatically increased from 1 to 10 mg m^{-2} under ice-covered conditions to more than 30 mg m^{-2}, in open-water conditions, particularly east and downstream of Barrow Canyon (Cooper et al., 2005), but in 2004 the increases in chlorophyll *a* inventories following ice retreat were more modest (Figs. 2 and 3). However, for stations occupied under both ice-covered and open-water conditions, there was a significant increase in sediment chlorophyll *a* inventories between the ice-covered and the open-water periods (two-tailed *t*-test; $p < 0.009$, $n = 18$). One potential explanation for this difference between 2002 and 2004 is that the ice-covered sampling occurred roughly 2 weeks later in 2004 when sea-ice cover had already begun to degrade, so the smaller increase in sediment chlorophyll *a* inventories may simply reflect early season deposition of chlorophyll *a*, while ice cover was still present following snowmelt (e.g., Peinert et al., 2001; Fortier et al., 2002; Sakshaug, 2004). It is also worth noting that water-column chlorophyll *a* was higher under ice-covered conditions in 2004 than in 2002 (Lalande et al., 2007b; Codispoti et al., 2009), which would explain the subsequent higher surface sediment chlorophyll *a* values and the reduced difference between ice-covered and open-water conditions in 2004 compared to 2002.

C/N ratios and the carbon isotope composition of the organic carbon fraction of sediments have been previously used in this region as key indicators of the quality of organic carbon deposited to the benthos (Grebmeier et al., 1988; Grebmeier and McRoy, 1989; Feder et al., 2007). Results from both sediment trap

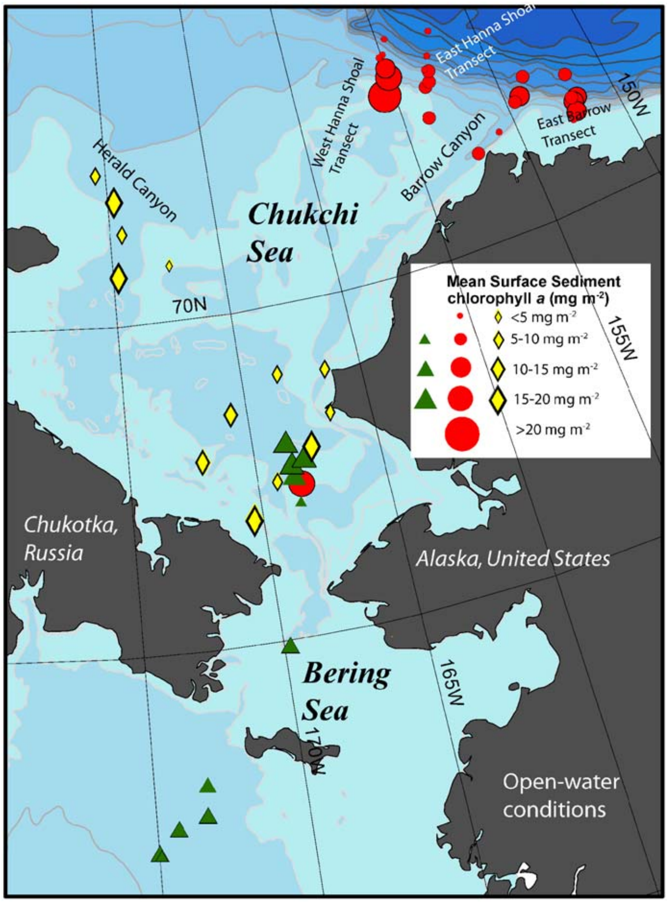


Fig. 3. Inventories of chlorophyll *a* present in surface sediments under largely open-water conditions, July–August 2004. Sampling was from USCGC *Healy* (circles: July–August), CCGS *Sir Wilfrid Laurier* (triangles: July) and R/V *Professor Khromov* (diamonds: August).

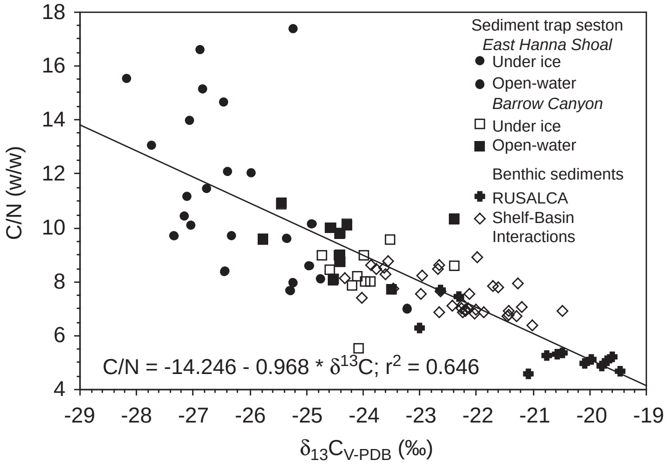


Fig. 4. Carbon/nitrogen (C/N) ratios versus δ^{13}C values of sediment trap material and sediments collected in 2004 during RUSALCA (R/V *Professor Khromov*) and Shelf-Basin Interactions (USCGC *Healy*) projects. Two transects were used for sediment trap deployments, East Hanna Shoal (EHS) and Barrow Canyon (BC). More information concerning depths and locations for the sediment trap deployments is reported in Lalande et al. (2007a, b).

sampling, as well as sediment sampling indicate that higher C/N ratios are significantly correlated with more negative δ^{13}C values (Fig. 4; see Lalande et al., 2007b for additional discussion of variation in these trap data by depth). This pattern reflects upon the refractory nature of the organic carbon sampled; more labile organic carbon is present in regions such as Barrow Canyon that have been identified as being more biologically productive (Hill and Cota, 2005) and on the Russian Chukchi Shelf (Walsh et al., 1989, 2005). More refractory organic carbon (more negative δ^{13}C values and higher C/N ratios) is also observed under ice cover in waters over East Hanna Shoal (Fig. 4) that are considered less productive (Hill and Cota, 2005). By contrast, there was less difference in the quality of sinking particles in Barrow Canyon under ice-covered versus open-water conditions (although the differences remain significant; Lalande et al., 2007b), possibly reflecting higher biological productivity and more labile organic carbon sedimenting to the sea floor throughout the productive season.

Several additional insights are apparent from the combined sediment trap and surface sediment data presented here. First, regional differences in the quality of organic carbon deposited to the benthos are significant, reflecting terrestrial or otherwise more refractory carbon versus more labile marine organic carbon sources in addition to overall biological productivity (Naidu et al., 2000; Cooper et al., 2002). Simultaneously, seasonal differences in the quality of carbon that are observed in less productive shelf waters such as East Hanna Shoal are larger than the same indicators in sinking particles or sediments than occurs in productive sites such as Barrow Canyon and the Russian sector of the Chukchi Sea. The similar organic carbon isotope composition between ice-covered and open-water conditions in sinking particles in Barrow Canyon (Fig. 4) is different than observed in the Northeast Water and North Water polynyas, where differences in the carbon isotope composition of ice algae and particulate organic matter under open-water conditions exceeded 8 per mille (Northeast Water; Hobson et al., 1995) and 4 per mille (North Water; Hobson et al., 2002). The lack of a difference in the δ^{13}C values for particulate organic matter under ice cover (expected to be less negative) versus open-water production (expected to be more negative) may be muted because of the very high primary production rates that have been observed in Barrow Canyon (Hill and Cota, 2005). Since under these productive waters there is no significant difference in the δ^{13}C values of sinking organic matter (Lalande et al., 2007b), whether from under-ice production that is dominated by ice algae (Gradinger, 2009) or from later open-water production, our results suggest that accurate estimates of the extent of contributions of ice algae versus open-water production to the underlying sediments and food webs could be subject to overestimation in highly productive areas where discrimination against ^{13}C during photosynthesis decreases. This possibility also would explain in part the greater isotopic differences between ice-covered and open water particulate matter observed in the Northeast Water polynya relative to the North Water polynya (Hobson et al., 1995, 2002) because the Northeast Water polynya is less biologically productive than the North Water (Deming et al., 2002; Klein et al., 2002).

Another process that is apparent in our sampled sediments is bacterial mineralization that tends to actively shift δ^{13}C values of organic material over time to less negative values (Lovvorn et al., 2005). The organic carbon fraction in our benthic sediments are systematically less depleted in δ^{13}C values than particles collected from the water column (Dunton et al., 2005; Lalande et al., 2007b). However, it is worth noting that the trap particles under ice on Hanna Shoal have even higher (more refractory) C/N ratios than any sediments that were collected while still retaining more negative δ^{13}C values (~ -27 to -28 per mille) than most sediments. While the higher C/N ratios probably reflect regeneration in the water column, it is clear that they also reflect differences in the temporal integration capabilities of the

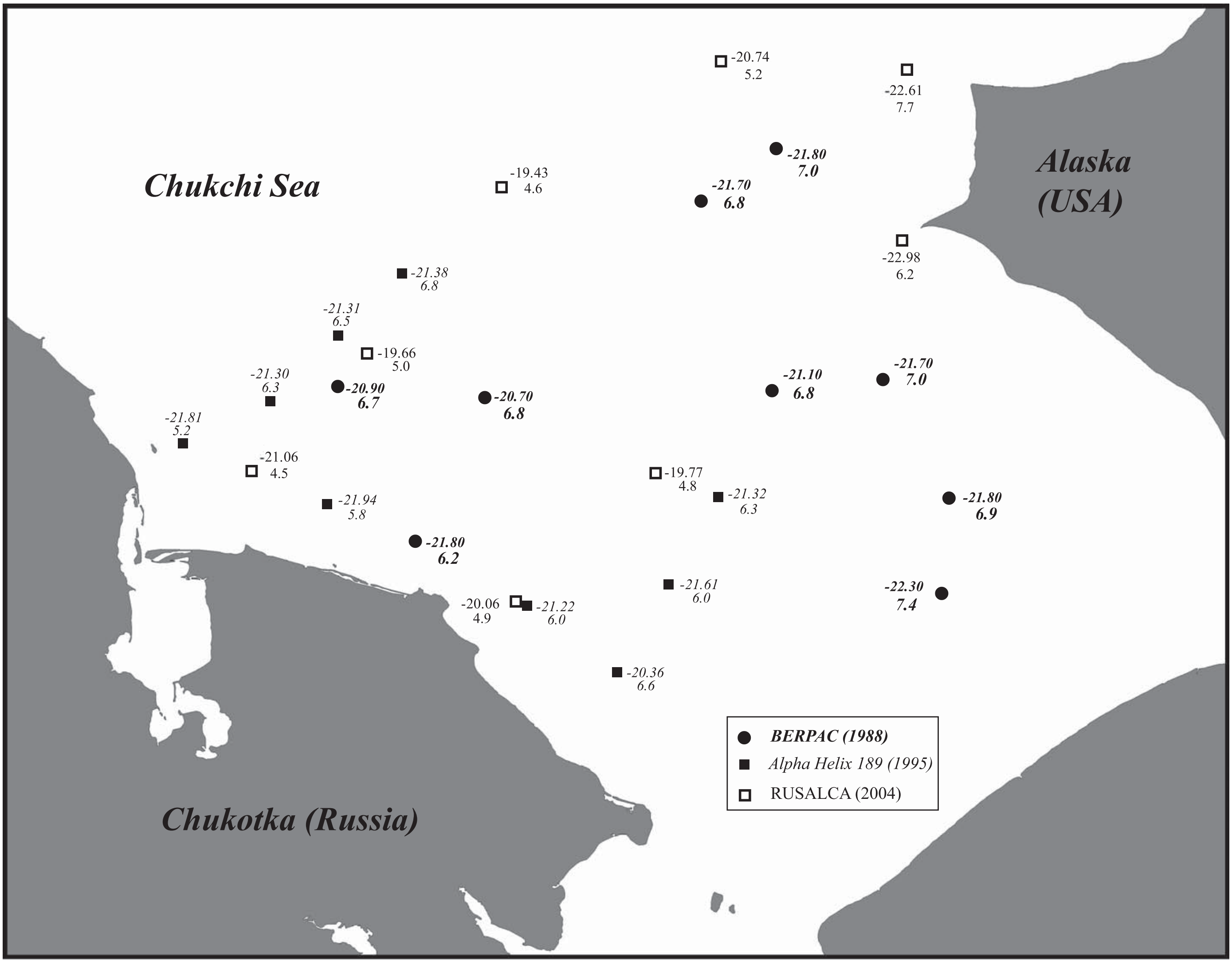


Fig. 5. Subset of sampling locations where the 1988 BERPAC (filled circles) and R/V *Alpha Helix* cruise 189 (filled squares: August–September 1995) overlapped with sampling locations on R/V *Professor Khromov* cruise (open squares: August 2004). Data shown for each station are δ^{13}C values, with C/N ratios (weight/weight) immediately below; italics and bold are used to separate individual cruises.

short-term trap deployments versus the longer-term integration provided by sediments.

In addition to seasonal differences in sedimentation, one of the objectives of this study was to compare sedimentation patterns with past work in order to determine if there have been any perceivable impacts of the recent retreat of seasonal sea ice in the Amerasian Arctic on organic sedimentation to the shallow seafloor. The 189th cruise of the R/V *Alpha Helix* (HX 189) in the Russian sector of the Chukchi Sea and the East Siberian Sea in August–September 1995 overlapped some of the sampling sites occupied during the RUSALCA cruise aboard the R/V *Professor Khromov* in August 2004 (Fig. 5; see also Table 4). Another cruise that sampled the sediments in this area (Fig. 5; Table 4) was the Third Joint US–USSR Bering and Chukchi Seas Expedition (BERPAC) in 1988 (Nagel, 1992). Previously published data from these two cruises (Cooper et al., 1998; Khim et al., 2003; Grebmeier et al., 2006b) show that the δ^{13}C values and C/N ratios of organic materials in 2004 were both significantly lower than in a subset of surface sediment samples collected in 1988 and 1995 where there was close overlap in sampling with our 2004 sampling. The 1995 samples in particular were collected, as in 2004, using an identical van Veen grab, and prepared using the same methods and analyzed with the same stable isotope mass spectrometer, internal standards and sample preparation protocols (Cooper et al., 1998; Khim et al., 2003; Grebmeier et al., 2006a). The identical methodology indicates the shift in organic carbon quality is real and not a sampling artifact.

It is known that benthic community productivity has declined in some areas of the Bering Strait region, potentially reflecting changes in particle deposition to the sediments (Grebmeier et al., 2006b), There are also indications of fluctuations of water flow through Bering Strait into the Chukchi Sea (Woodgate et al., 2006), so it is possible that changes we observe in biogeochemical markers in the sediments reflect some of the same regional changes being observed ecologically and in Bering Strait flow. Ultimately, a change in the timing, quantity and quality of organic matter reaching the benthos, as indicated by short-term markers such as chlorophyll *a* inventories in surface sediments and δ^{13}C values and C/N ratios in organic sediment fractions, could be expected to impact benthic biological communities, which have among the highest biomass of any soft-bottom marine communities worldwide (Highsmith and Coyle, 1990; Grebmeier and Barry, 1991, 2007; Grebmeier et al., 1995, 2006b; Ambrose et al., 2001).

Table 4
Data used in comparison of C/N (w/w) ratios and $\delta^{13}C$ values of sediment organic carbon from three separate cruises in the southern Chukchi Sea, 1988 (Joint Soviet–US expedition to the Bering and Chukchi Seas, "BERPAC"), 1995 (*Alpha Helix* cruise 189), and 2004 (Joint Russian–US RUSALCA program).

Data source	Station	Date of collection	Latitude (°N)	Longitude (°W)	$\delta^{13}C_{V-PDB}$	C/N (w/w)
BERPAC	45	9 August 1988	67.733	−172.833	−20.90	6.44
BERPAC	49	10 August 1988	68.467	−169.133	−21.70	6.82
BERPAC	50	10 August 1988	68.662	−168.333	−21.80	7.22
BERPAC	54	11 August 1988	67.763	−167.315	−21.70	6.85
BERPAC	55	11 August 1988	67.735	−168.440	−21.10	6.59
BERPAC	57	12 August 1988	67.710	−171.345	−20.70	6.58
BERPAC	59	12 August 1988	67.153	−172.000	−21.80	6.39
BERPAC	64	13 August 1988	67.297	−166.710	−21.80	6.80
BERPAC	67	14 August 1988	66.933	−166.833	−22.30	7.64
HX 189	66	31 August 1995	67.666	−173.502	−21.3	6.32
HX 189	67	31 August 1995	67.483	−174.351	−21.87	5.17
HX 189	68	31 August 1995	67.416	−173.998	−23.54	3.53
HX 189	69	31 August 1995	67.317	−173.600	−23.59	No data
HX 189	70	31 August 1995	67.283	−172.880	−21.94	5.84
HX 189	72	1 September 1995	66.917	−171.803	−21.22	6.06
HX 189	73	1 September 1995	66.666	−170.001	−20.36	6.60
HX 189	74	1 September 1995	67.000	−169.499	−21.61	6.03
HX 189	76	1 September 1995	67.333	−168.998	−21.32	6.28
RUSALCA	7	11 August 2004	66.937	−170.997	−20.06	6.25
RUSALCA	9	12 August 2004	67.432	−169.621	−19.78	7.66
RUSALCA	13	13 August 2004	68.297	−167.051	−22.99	4.65
RUSALCA	14	14 August 2004	68.950	−166.912	−22.61	5.03
RUSALCA	16	15 August 2004	69.006	−168.895	−20.75	4.52
RUSALCA	19	15 August 2004	68.524	−171.214	−19.43	5.16
RUSALCA	21	16 August 2004	67.870	−172.551	−19.67	5.33
RUSALCA	23	18 August 2004	67.397	−173.653	−21.07	5.05
RUSALCA	26	20 August 2004	70.760	−175.534	−19.58	5.24
RUSALCA	68	21 August 2004	72.317	−175.984	−20.45	6.25
RUSALCA	74	20 August 2004	71.902	−175.486	−19.95	7.66
RUSALCA	75	21 August 2004	71.396	−174.912	−20.55	5.23

BERPAC data from Naidu et al. (1993); *Alpha Helix* cruise 189 from Cooper et al. (1998), Khim et al. (2003) and Grebmeier et al. (2006b).

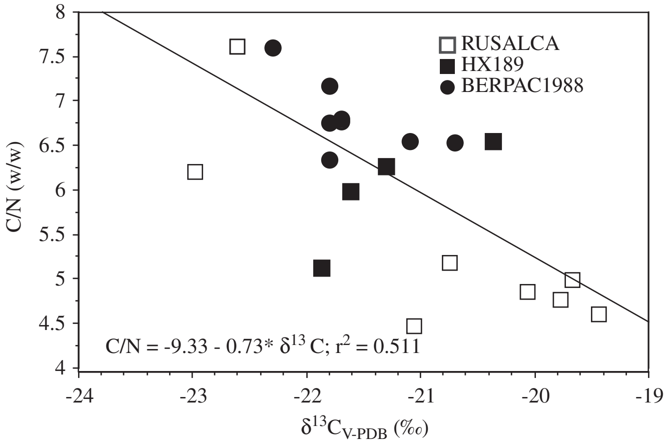


Fig. 6. Carbon/nitrogen (C/N) weight/weight ratios versus $\delta^{13}C$ values for a subset of sediments collected in 2004 during RUSALCA (R/V *Professor Khromov*—open squares) and in 1988 (BERPAC—filled circles) and in 1995 (R/V *Alpha Helix* cruise 189—filled squares) where sampling locations closely coincided. The full cruise transects sampled in 1988 and 1995 are shown in Nagel (1992) and Khim et al. (2003), respectively.

Moreover, it should be recognized that detecting change in sediment characteristics such as we report here is also potentially significant because changes in sediment chemistry are not likely to be as unambiguous as physical oceanographic processes or the ecosystem shifts in benthic communities that have been detected (e.g., Grebmeier et al., 2006a). Bioturbation mixes sediments to depths of 25–35 cm on the Chukchi shelf over multidecadal time scales (Grebmeier and McRoy, 1989; Cooper et al., 1998), so any changes in the quality of organic matter are averaged out over a number of years. Large-scale syntheses of sedimentary characteristics (e.g., Naidu et al., 2000; Cooper et al., 2002; Dunton et al., 2005; Grebmeier et al., 2006b) also imply a fairly static sedimentation regime, with terrestrial, refractory carbon deposited near river mouths, particularly on the Alaskan coastal margin and less refractory organic carbon deposited on the Russian side of the Bering and Chukchi Seas that are influenced by productive Anadyr water.

Despite this, the more recently measured $\delta^{13}C$ values and C/N ratios have unambiguously and systematically shifted (Figs. 5 and 6), and samples were consistently less refractory than samples collected at nearby stations in 1988 and 1995. Although additional sampling is probably necessary to verify this apparent shift, it seems reasonable to hypothesize that it is due to significant changes in hydrography over the past decade such as possible increases in freshwater flow through Bering Strait (Woodgate et al., 2006). These increases in flow of Alaska Coastal Current northward through Bering Strait could have the effect of restricting and ultimately shifting the influence of more productive and saline Anadyr waters on the western side of Bering Strait to a more localized area of the Russian Chukchi Sea adjacent to Chukotka. This hypothetical explanation is also consistent with decadal-scale declines in benthic biomass and sediment oxygen respiration observed in areas of formerly higher productivity in the US sector of the Bering Strait region (Grebmeier et al., 2006a). Clearly additional longer-term observations of the sediment and biological system in the Bering Strait region are needed to document the potential impacts of shifting hydrographic and sea-ice regimes upon biogeochemical processes.

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