2013

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JOURNAL OF GEOPHYSICAL RESEARCH: OCEANS, VOL. 118, 4017–4035, doi:10.1002/jgrc.20286, 2013

Distribution and sources of organic matter in surface marine sediments across the North American Arctic margin

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Received 18 February 2013; revised 22 May 2013; accepted 19 June 2013; published 5 September 2013.

[1] As part of the International Polar Year research program, we conducted a survey of surface marine sediments from box cores along a section extending from the Bering Sea to Davis Strait via the Canadian Archipelago. We used bulk elemental and isotopic compositions, together with biomarkers and principal components analysis, to elucidate the distribution of marine and terrestrial organic matter in different regions of the North American Arctic margin. Marked regional contrasts were observed in organic carbon loadings, with the highest values ($\geq$1 mg C m^{-2} sediment) found in sites along Barrow Canyon and the Chukchi and Bering shelves, all of which were characterized by sediments with low oxygen exposure, as inferred from thin layers (<2 cm) of Mn oxihydroxides. We found strong regional differences in inorganic carbon concentrations, with sites from the Canadian Archipelago and Lancaster Sound displaying elevated values (2–7 wt %) and highly depleted ^{14}C compositions consistent with inputs from bedrock carbonates. Organic carbon:nitrogen ratios, stable carbon isotopes, and terrigenous organic biomarkers (lignin phenols and cutin acids) all indicate marked regional differences in the proportions of marine and terrigenous organic matter present in surface sediments. Regions such as Barrow Canyon and the Mackenzie River shelf were characterized by the highest contributions of land-derived organic matter, with compositional characteristics that suggested distinct sources and provenance. In contrast, sediments from the Canadian Archipelago and Davis Strait had the smallest contributions of terrigenous organic matter and the lowest organic carbon loadings indicative of a high degree of post-depositional oxidation.

Citation: Goñi, M. A., A. E. O'Connor, Z. Z. Kuzyk, M. B. Yunker, C. Gobeil, and R. W. Macdonald (2013), Distribution and sources of organic matter in surface marine sediments across the North American Arctic margin, *J. Geophys. Res. Oceans*, *118*, 4017–4035, doi:10.1002/jgrc.20286.

1. Introduction

[2] The Arctic Ocean, which is characterized by broad shelves and large inputs of water, sediment, and organic matter from land [*Stein and Macdonald*, 2004], is undergoing rapid, unprecedented changes associated with climate warming [*Intergovernmental Panel on Climate Change*, 2007]. The changes in the cryosphere include loss of multiyear pack ice, permafrost thawing, and loss of snow cover (http://www.climatewatch.noaa.gov/article/2012/2012-arctic-report-card), all of which have the potential to dramatically affect the biogeochemistry of the Arctic Ocean and its adjacent seas [e.g., *Richter-Menge et al*., 2006; *Serreze et al*., 2007, 2009; *Arrigo et al*., 2008; *Moline et al*., 2008; *Lavoie et al*., 2009; *Zhang et al*., 2010; *Walsh et al*., 2011]. For example, reductions in ice cover increase light penetration and lead to higher photosynthetically active radiation (PAR) at depth, which in the absence of nutrient limitation can result in elevated rates of primary production and higher carbon stocks and fluxes [e.g., *McLaughlin and Carmack*, 2010; *Forest et al*., 2011; *Arrigo et al*., 2012]. Increased ice-free conditions also can increase the supply of nutrients from deeper water masses via wind-driven mixing and upwelling [e.g., *Mundy et al*., 2009; *Pickart et al*., 2009, 2011], potentially enhancing primary productivity and carbon fluxes [e.g., *Moran et al*., 2005; *Lalande et al*., 2007; *Forest et al*., 2010], and possibly leading to shifts in pelagic ecosystems toward "sub-Arctic"-like conditions [e.g.,

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2169-9275/13/10.1002/jgrc.20286

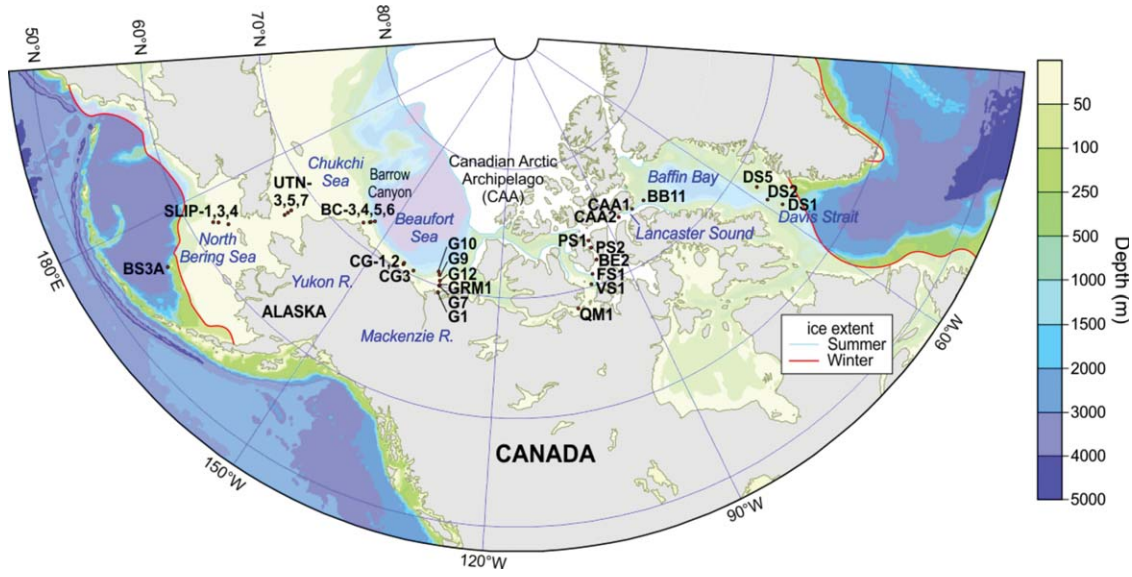

Figure 1. Chart of the North American Arctic margin showing locations of stations, major bathymetry, and names of major oceans/canyons/rivers.

Anderson and Kaltin, 2001; *Zhang et al*., 2010; *Walsh et al*., 2011].

[3] The seaward transfer of land-derived inorganic sediments and organic material is also expected to be strongly affected by climate change. Extended periods of open water allow storm-induced waves during late summer and fall to build over longer fetches. Combined with permafrost thawing and sea level rise, sea ice retreat may result in accelerated rates of coastal erosion over many regions of the Arctic [e.g., *Rachold et al*., 2000, 2004; *Jorgenson and Brown*, 2005; *Lantuit et al*., 2012]. Much of the sediment (and associated organic matter) mobilized from the near shore by wave action is delivered to the interior basins through resuspension and transport to submarine canyons and other bathymetric features that facilitate lateral export during downwelling or dense water formation conditions [e.g., *Weingartner et al*., 1998; *Williams et al*., 2008; *Vogt and Knies*, 2009]. In contrast, sediment entrainment and transport by ice [e.g., *Eicken*, 2004; *Dethleff*, 2005], which is an important process in regions such as the Barents Sea, may decrease as the extent and duration of ice-free periods increase. Overall, reduced sea ice and degradation of permafrost are expected to enhance the fluxes of land-derived lithogenic materials as well as nutrients and organic matter, the latter of which appears to be reactive and contributes significantly to community metabolism in marine systems [e.g., *Amon*, 2004; *Davis and Benner*, 2005; *Goñi et al*., 2005; *Holmes et al*., 2008; *Alling et al*., 2010; *Letscher et al*., 2011; *Sanchez-Garcia et al*., 2011]. Such increases in allochthonous organic matter inputs can have significant impacts on the higher trophic levels of the pelagic and benthic food webs [e.g., *Carmack and Wassmann*, 2006; *Dunton et al*., 2006; *Grebmeier et al*., 2006; *Moline et al*., 2008].

[4] Documenting the magnitude and rates of biogeochemical changes in the Arctic Ocean requires a comprehensive understanding of conditions prior to anthropogenic impacts on climate. Sediments provide valuable records of past biogeochemical conditions that can be used to assess recent changes. Specifically, past work on various locations has demonstrated the value of organic compositional data for determining sources of carbon—i.e., autochthonous versus allochthonous—in the Arctic Ocean [e.g., *Peulve et al*., 1996; *Yunker et al*., 1995, 2005, 2011; *Goñi et al*., 2005; *Knies et al*., 2007; *Belicka et al*., 2009; *Vonk et al*., 2012, and references therein]. Given the ongoing changes in the cryosphere and their likely effects on these carbon pools, it seems clear to us that organic biomarker studies will provide useful data to further our understanding of change in Arctic biogeochemistry—past, present, and future.

[5] Although multiple studies have investigated organic carbon provenance and distribution in the Arctic Ocean, most have focused on a limited region or a single shelf, and each has applied its own set of analytical tools (see previous citations). Collectively, these studies are building an overarching image of the Arctic's organic cycle, but major gaps still remain because of the difficulties in applying these differing data sets across Arctic regions. Because the sources and inputs of carbon vary widely among Arctic shelves and basins, none of them serves as a reliable proxy for the Arctic in general or for each other [e.g., *Carmack et al*., 2006; *Macdonald et al*., 2010]. Accordingly, an important step to enhance the interpretation of biomarker data from the Arctic Ocean is to collect and analyze sediments from a variety of Arctic margin settings using the same techniques. Here, we provide a comparative survey of biomarker data for sediments collected along a section extending from the Bering Sea to Davis Strait via the Canadian Arctic Archipelago (Figure 1). This study was made possible by the International Polar Year (IPY) Program, which also afforded the opportunity to develop a better comparative understanding of the oceanographic domains and sediment accumulation processes along these various shelf margins domains [*Carmack et al*., 2010; *Kuzyk et al*., 2013; *Macdonald and Gobeil*, 2012]. Using the biomarker data together with bulk isotopic analyses, we evaluate the relative importance of terrigenous and marine carbon in these sediments and infer processes involved in the supply

Table 1. Location and Characteristics of Sediment Samples[a]

Sample Code	Latitude (N)	Longitude (W)	Water Depth (m)	Date of Collection	Core Length (cm)	Horizon Analyzed (cm)	Mn-Layer Thickness (cm)	Surface Mixed Layer (cm)	Sediment Accum. Rate (cm/y)
Bering Sea Slope									
BS3A	56.476	172.780	790	12 Jul 2007	13	0–0.5	n.a.[b]	n.a.	n.a.
Bering Sea Shelf									
SLIP1	62.014	175.056	80	13 Jul 2007	22	0–0.5	0.75	20	n.a.
SLIP3	62.393	174.570	73	13 Jul 2007	20	0–0.5	0.75	15	n.a.
SLIP4	63.029	173.458	73	14 Jul 2007	22	0–0.5	0.75	20	n.a.
Chukchi Sea Shelf									
UTN3	67.334	169.000	50	17 Jul 2007	11	0–0.5	0.25	10	n.a.
UTN5	67.670	168.958	51	17 Jul 2007	21	0–0.5	0.25	20	n.a.
UTN7	68.000	168.933	58	17 Jul 2007	24	0–0.5	0.25	30	n.a.
Barrow Canyon									
BC3	71.578	156.018	186	19 Jul 2007	16	0–0.5	0.75	19	n.a.
BC4	71.930	154.887	599	19 Jul 2007	24	0–0.5	1.75	4	0.24
BC5	72.000	154.708	1015	20 Jul 2007	22	0–0.5	3.50	2	0.06
BC6	72.233	154.037	2125	20 Jul 2007	27	0–0.5	1.75	6	0.11
Beaufort Sea slope									
CG1	70.562	142.858	204	24 Jul 2007	25	0–0.5	5.5	2	0.20
CG2	70.700	142.833	619	24 Jul 2007	25	0–0.5	15	1	0.13
CG3	70.461	140.160	566	25 Jul 2007	24	0–0.5	19	2	0.10
Mackenzie Shelf									
G1	69.542	133.292	5	31 Jul 1987	Grab	0–2	n.m.[c]	n.m.	n.m.
G7	69.892	133.412	25	3 Aug 1987	Grab	0–2	n.m.	n.m.	n.m.
GRM1	70.017	133.433	30	3 Aug 1987	Grab	0–2	n.m.	n.m.	n.m.
G12	70.737	134.165	61	5 Aug 1987	Grab	0–2	n.m.	n.m.	n.m.
G9	70.947	134.595	210	5 Aug 1987	Grab	0–2	n.m.	2[d]	~0.1[d]
G10	70.947	134.595	210	5 Aug 1987	Grab	0–2	n.m.	2[d]	~0.1[d]
Canadian Archipelago									
QM1	68.667	103.004	113	17 Aug 2008	25	0–0.5	3.5	1	0.18
VS1	70.249	98.929	210	16 Aug 2008	25	0–0.5	6.5	1	0.05
FS1	70.999	97.998	141	15 Aug 2008	17	0–0.5	15	2	0.09
BE2	71.973	95.998	431	15 Aug 2008	24	0–0.5	15	1	0.09
PS1	73.597	96.295	245	14 Aug 2008	26	0–0.5	4.5	1	0.08
PS2	73.000	96.218	340	14 Aug 2008	27	0–0.5	11	1	0.18
Lancaster Sound									
CAA2	73.935	86.108	368	11 Aug 2008	8	0–0.5	1.5	1	0.11
CAA1	73.927	81.814	630	11 Aug 2008	17	0–0.5	4.5	4	0.15
BB11	73.933	77.930	850	11 Aug 2008	26	0–0.5	3.5	1	0.11
Davis Strait									
DS5	68.037	57.537	340	8 Aug 2008	19	0–0.5	1.25	2	0.06
DS2	66.758	58.545	780	9 Aug 2008	18	0–0.5	19	0	0.07
DS1	65.488	57.771	575	9 Aug 2008	25	0–0.5	5.5	2	0.08

[a]All data from *Macdonald and Gobeil* [2012] and *Kuzyck et al*. [2013].
[b]n.a., not applicable.
[c]n.m., not measured.
[d]From *Yunker et al*. [2011].

and cycling of organic matter along the North American Arctic margin. Our intention is to use the results of this study to develop insights into the biogeochemistry of this region that is poised to undergo substantial change in the coming decades.

2. Sample Collection and Analysis

[6] Surface sediments for this study were collected from a variety of locations and water depths along the North American Arctic margin (Figure 1 and Table 1). Details on the characteristics of each location and of the coring and subsampling methods have been discussed previously [e.g., *Goñi et al*., 2000; *Kuzyk et al*., 2013]. Briefly, in 2007 sediments were collected from the CCGS Wilfrid Laurier using a Pouliot box corer at stations located in the Bering Sea slope (BS3), Bering Sea shelf (SLIP1, SLIP3, and SLIP3), Chukchi Sea shelf (UTN3, UTN5, and UTN7), Barrow Canyon (BC3, BC4, BC5, and BC6), and the Beaufort Sea slope (CG1, CG2, and CG3). In 2008, the core collection was completed from the CCGS Louis St. Laurent on an east to west transect from Davis Strait (DS2, DS1, and DS5) to Lancaster Sound (CAA2, CAA1, and BB11) and then through the Canadian Arctic Archipelago (QM1, VS1, FS1, PS1, PS2, and BE2). Once recovered, each box core was subsampled at 0.5 cm intervals near the surface and greater intervals at depth, with sediments from individual horizons placed in sealed containers and stored frozen until

analyses. For this study, where we examine the distribution of organic biomarkers along the entire ~8000 km section, only the top horizon from each core is considered. To this sample set, we have added sediments from a transect across the shelf off the Mackenzie River delta (G1, G7, GRM1, G12, G9, and G10) in the Beaufort Sea, which were collected in 1987 using Smith McIntyre and Ponar (G1 only) grab samplers. The top 2 cm of each grab was subsampled and frozen for analysis. Some of the samples from the Mackenzie shelf transect were located directly below the marginal ice zone and its impact on organic matter composition was considered in previous publications [*Goñi et al*., 2000; *Yunker et al*., 1995]

2.1. Analytical Methods

[7] In the laboratory, sediment samples were freeze dried and homogenized by grinding with a mortar and pestle. A split of unground sample was combusted to remove organic matter and then measured for mineral surface area using a five-point BET method and a Micromeritics TriStar 3000 surface area analyzer [e.g., *Tesi et al*., 2012]. Elemental and isotopic analyses were conducted on ground samples according to established techniques [e.g., *Goñi et al*., 2000, 2005, 2009; *Hastings et al*., 2012]. Total carbon and nitrogen contents were determined by high temperature combustion of untreated samples using a NC2500 ThermoQuest Elemental Analyzer. Organic carbon contents (%OC) were determined on splits of each sample that were subjected to multiple acid treatments, including exposure to concentrated HCl fumes followed by additions of 10% aqueous HCl. These treatments were done on preweighed samples in silver boats, which after oven drying were wrapped in tin boats and analyzed by high temperature combustion using the same elemental analyzer. Inorganic carbon contents were determined as the difference between total carbon and organic carbon contents. We observed a significant linear correlation between organic carbon and total nitrogen contents in the analyzed samples, which exhibited an intercept statistically indistinguishable from zero (Figure 2). From this we infer that nitrogen in these margin sediments is predominantly organic in origin [e.g., *Goñi et al*., 2003; *Macdonald et al*., 2004]. Previous work has shown significant inorganic nitrogen in illite-rich sediments from the deep basin in the central Arctic Ocean [*Schubert and Calvert*, 2001] and in sediments from the Laptev Sea and Yermak Plateau [*Stein and Macdonald*, 2004; *Winkelmann and Knies*, 2005]. Given no evidence for significant amounts of bound inorganic N in our Arctic margin surface sediments, we assume that total organic carbon:nitrogen ratios are indicative of the C:N in sedimentary organic matter.

[8] Carbonates were removed from sediment subsamples prior to stable isotopic analysis of organic carbon ($\delta^{13}C_{org}$) by repeatedly treating with aliquots of 10% HCl, thoroughly mixing with the sediment matrix, and then evaporating to dryness. The $\delta^{13}C_{org}$ analyses were done by high temperature combustion followed by isotope ratio mass spectrometry using a Carlo Erba 1500 Elemental Analyzer coupled to a ThermoQuest Delta Plus XP Mass Spectrometer. The $^{13}C/^{12}C$ compositional data are reported relative to the PDB standard (VPDB) using the δ (‰) notation. Radiocarbon analyses of organic and inorganic carbon ($\Delta^{14}C_{org}$ and $\Delta^{14}C_{inor}$, respectively) from selected sediment samples were performed at the National Ocean Sciences Accelerator Mass Spectrometry facility at Woods Hole Oceanographic Institution following established procedures [*McNichol et al*., 1992; *Vogel et al*., 1987]. Carbon dioxide from either combustion of acid-treated samples or from the acidification of sedimentary carbonates was converted to graphite, pressed into targets, and analyzed by accelerator mass spectrometry. Radiocarbon data are reported using the $\Delta^{14}C$ notation and as the fraction modern and conventional radiocarbon age for each sample [e.g., *Stuiver et al*., 1986, and references therein].

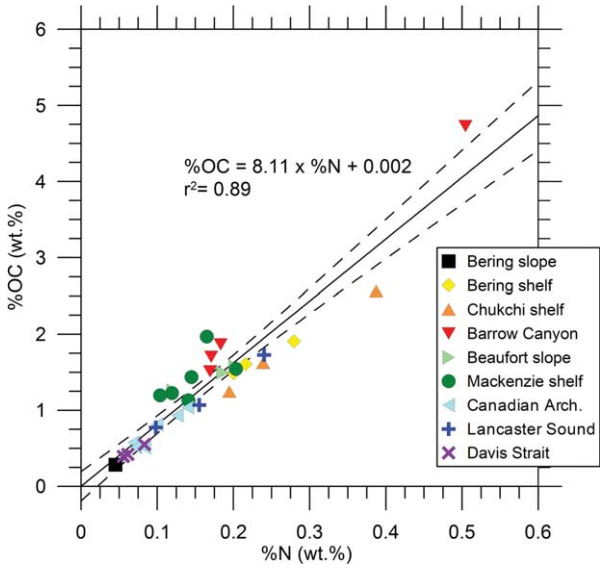

Figure 2. Relationship between weight percent content of nitrogen (%N) and organic carbon (%OC) for surface sediments analyzed. Linear fit of the data is shown along with the 95% confidence intervals.

[9] Biomarker analysis was done by the alkaline CuO oxidation technique using either direct heating in stainless steel pressurized vessels [e.g., *Goñi and Hedges*, 1992] or microwave-controlled heating in pressurized Teflon vessels [*Goñi and Montgomery*, 2000]. The microwave-based technique was developed as a faster, more convenient approach to perform CuO oxidations and produces comparable concentration and compositional results to the direct heating technique. After oxidation and extraction, over 100 individual CuO products were quantified using gas-chromatography mass spectrometry [e.g., *Goñi et al*., 2009; *Hatten et al*., 2012]. We used multilevel, external calibrations made up of commercially available standards to determine the responses of different structure-specific ions and to quantify individual product yields. For this study, we report the combined yields of individual CuO products as different biomarker classes including lignin-derived vanillyl, syringyl, and cinnamyl phenols, all of which are characteristically derived from different terrestrial vascular plant sources [e.g., *Hedges and Mann*, 1979]. Other biomarker classes reported include cutin-derived hydroxy fatty acids, which are characteristic of soft aerial (leaves, needles) and subaerial (roots) tissues of vascular plants [e.g., *Goñi and Hedges*, 1990; *Crow et al*., 2009a], as well as several product classes derived from nonvascular

plant sources, including p-hydroxybenzenes, benzoic acids, amino acid-derived products, dicarboxylic acids, and fatty acid products [e.g., *Goñi and Hedges*, 1995]. The yields of individual compounds that make up these product categories were used in multivariate analyses (see below) and are presented in supporting information.

[10] Ancillary measurements of Ca and Mg contents (C. Gobeil, unpublished data, 2009), radionuclide compositions [*Kuzyk et al*., 2013], and a number of redox-sensitive element distributions including Mn [*Macdonald and Gobeil*, 2012] were performed on sediments from the box cores collected in the 2007 and 2008 campaigns. The authors used ^{210}Pb and ^{137}Cs profiles to infer sediment focusing and scavenging at core sites along the transect [*Kuzyk et al*., 2013], and down-core Mn profiles to evaluate sediment redox conditions at each site [*Macdonald and Gobeil*, 2012]. We utilize both of these data sets to help interpret our measurements in the context of supply and preservation of sedimentary organic matter. Because the Mackenzie shelf samples were all from surface grabs, this ancillary information is not available for this region of the margin.

2.2. Principal Components Analysis (PCA)

[11] Each individual CuO oxidation product was evaluated for potential interferences, closeness to the limit of detection, and the percentage of undetectable values before inclusion in the final PCA data set containing 39 variables (supporting information). Bulk and isotope data were not included in the PCA because they have a different basis of quantification (external standard versus internal standard for concentration data) with different variable distributions (binomial for percent versus likely log normal for concentrations), which the PCA model could interpret as differences in composition. A PCA model was developed for the 26 Arctic margin samples from 2007 and 2008 and the six Mackenzie shelf sediment samples. For this sediment data set, the few undetectable values (29 total or 2.3% of the entire data set) were replaced by a random number between zero and the limit of detection by multiplying the value of the detection limit by a random number between 0 and 1 (generated by a library function in the spreadsheet program). Samples were normalized to the concentration total before PCA to remove artifacts related to concentration differences between samples. The centered log ratio transformation (division by the geometric mean of the concentration-normalized sample followed by log transformation) was then applied to this compositional data set to produce a data set that was unaffected by negative bias or closure [*Yunker et al*., 2005, 2011] and where the average concentration and concentration total were identical for every sample. Data were then autoscaled by subtracting the variable mean and dividing by the variable standard deviation before PCA to give every variable equal weight.

3.. Results

3.1. Sediment Compositions

[12] Mineral surface area (SA) values ranged from ~4 to over 40 $m^2\ g^{-1}$ (Table 2) indicating a wide contrast in the distribution of fine-grained sediments. For example, based on previous studies [e.g., *Goñi et al*., 2006], mud-rich sediments are characterized by SA values of >30 $m^2\ g^{-1}$, whereas sandy sediments have SA values <10 $m^2\ g^{-1}$. There were no clear trends with water depth and/or longitude but in general we observed the lowest surface areas (e.g., coarsest sediments) in regions with low fluvial inputs, including the Canadian Archipelago and Davis Strait (e.g., FS1, PS2, CAA2, CAA1, and DS5) and the deep slope of the Bering Sea (BS3) or in sites with shallow water depths where winnowing of fines may occur (e.g., UTN3). Surface area values above 30 $m^2\ g^{-1}$ were found in the slope region of the Beaufort Sea (CG2 and CG3) and at the eastern entrance of Lancaster Sound (BB11).

[13] Organic carbon content (%OC) throughout most stations along the Arctic margin ranged from 1 to 2 wt % (Table 2), with lower values measured at BS3, the Davis Strait stations (DS5, DS2, and DS1) and five of the six Canadian Archipelago and Lancaster Sound stations. One of the Chukchi Sea shelf sites displayed higher %OC content (2.6 wt %), whereas the shallowest station within Barrow Canyon (BC3) stood out because of its significantly elevated %OC values (4.7 wt %). Generally, the %OC values measured in surface sediments along Bering, Chukchi, and Beaufort margins are comparable to those found by others (e.g., see regional summaries in *Stein and Macdonald* [2004]). The Barrow Canyon region has been shown to be highly productive [e.g., *Cooper et al*., 2009; *Christensen et al*., 2008; *Grebmeier et al*., 2006], although we are not aware of previous studies showing such elevated %OC contents as the one we measured in BC3. Finally, because sediment organic data from the Canadian Archipelago are rare and of limited coverage [e.g., *Vare et al*., 2009], we have few data with which to compare our results from this region.

[14] Accompanying these spatial patterns in %OC, there were marked regional contrasts in inorganic carbon contents (%IC) of surface sediments analyzed in this study (Table 2). Stations from the Bering and Chukchi Seas displayed very low %IC contents (<0.2 wt %), whereas intermediate values (0.3–2 wt %) characterized most of the stations along Barrow Canyon, Beaufort slope, Mackenzie shelf, and Davis Strait. Elevated %IC contents (~2–7wt %) were found in most of the Canadian Archipelago and Lancaster Sound stations, indicating that 30–60% of the sediment mass at some of these locations (VS1, FS1, BE2, PS1, PS2, and CAA2) may be made up of $CaCO_3$. With the exception of a few data from the easternmost part of the study area [e.g., *Andrews et al*., 1991], we were unable to find published literature on the %IC of surface sediments from the margin regions of the North American Arctic with which to compare our results.

3.2. Bulk Organic Matter Compositions

[15] Sedimentary organic matter along the North American Arctic margin displayed molar carbon:nitrogen ratios (C:N) ratios that ranged from 7 to 14 (Table 2). The lowest C:N ratios (<8) were found in the Chukchi Sea stations whereas most stations in the Bering Sea, Canadian Archipelago, Lancaster Sound, and Davis Strait displayed intermediate values between 8 and 10. The highest C:N ratios (10–14) were found in stations from Barrow Canyon, one of the Beaufort slope locations and several stations from the Mackenzie shelf. The moderately elevated C:N values of these latter stations are consistent with a higher

Table 2. Bulk Compositions of Surface Sediments

Sample	SA (m^2/g)	%OC (wt %)	%IC (wt %)	%Ca (wt %)	$C:N_{org}$ (molar)	$\delta^{13}C_{org}$ (‰)	$\Delta^{14}C_{org}$ (‰)	OC F_{mod}	OC Age	$\Delta^{14}C_{inor}$ (*‰*)	IC F_{mod}
Bering Sea Slope											
BS3A	4.9	0.28	0.01	1.98	7.4	−21.6	n.m.[b]	n.m.	n.m.	n.m.	n.m.
Bering Sea Shelf											
SLIP1	29.4	1.61	0.02	1.47	8.7	−21.9	−201.5	0.80	1750	n.m.	n.m.
SLIP3	30.5	1.49	0.09	1.87	8.7	−21.8	n.m.	n.m.	n.m.	n.m.	n.m.
SLIP4	26.6	1.91	0.07	1.81	8.0	−21.7	n.m.	n.m.	n.m.	n.m.	n.m.
Chukchi Sea Shelf											
UTN3	13.4	1.25	0.15	1.69	7.5	−20.6	n.m.	n.m.	n.m.	n.m.	n.m.
UTN5	18.3	1.62	0.08	1.76	7.9	−21.2	n.m.	n.m.	n.m.	n.m.	n.m.
UTN7	21.2	2.56	0.15	1.61	7.7	−21.1	−171.1	0.83	1450	n.m.	n.m.
Barrow Canyon											
BC3	22.7	4.73	0.52	1.98	10.9	−22.4	n.m.	n.m.	n.m.	n.m.	n.m.
BC4	23.9	1.87	0.76	2.25	11.9	−24.6	n.m.	n.m.	n.m.	n.m.	n.m.
BC5	28.6	1.51	0.53	1.53	10.4	−24.0	n.m.	n.m.	n.m.	n.m.	n.m.
BC6	28.4	1.70	0.64	1.85	11.6	−25.1	−658.6	0.34	8580	−980.2	0.02
Beaufort Sea Slope											
CG1	25.9	1.26	0.83	2.50	12.2	−23.6	−520.6	0.48	5850	−990.7	0.01
CG2	34.9	1.50	0.41	1.21	9.4	−24.0	n.m.	n.m.	n.m.	n.m.	n.m.
CG3	37.1	1.58	0.37	1.06	9.1	−24.5	n.m.	n.m.	n.m.	n.m.	n.m.
Mackenzie Shelf[a]											
G1	19.2	1.13	1.88	n.m.	9.4	−25.9	n.m.	n.m.	n.m.	n.m.	n.m.
G7	23.1	1.97	0.28	n.m.	13.9	−25.5	−691.3	0.31	9390	n.m.	n.m.
GRM1	26.6	1.44	0.63	n.m.	11.6	−25.7	−720.8	0.28	10200	n.m.	n.m.
G12	27.3	1.54	0.50	n.m.	8.9	−24.4	−590.9	0.41	7130	n.m.	n.m.
G9	28.6	1.23	0.45	n.m.	12.0	−25.1	n.m.	n.m.	n.m.	n.m.	n.m.
G10	29.0	1.20	0.44	n.m.	13.5	−25.5	−876.8	0.12	16750	n.m.	n.m.
Canadian Archipelago											
QM1	26.5	0.81	1.44	3.53	9.5	−21.7	−127.9	0.88	1040	−994.1	0.01
VS1	24.5	0.93	3.10	5.86	8.6	−21.0	n.m.	n.m.	n.m.	n.m.	n.m.
FS1	15.1	0.58	3.96	7.35	9.7	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.
BE2	21.9	0.51	4.88	8.68	7.2	−21.1	n.m.	n.m.	n.m.	n.m.	n.m.
PS1	19.3	1.04	3.74	7.02	8.6	−20.5	−301.8	0.70	2830	−997.0	0.00
PS2	13.2	0.52	5.12	9.24	8.4	−21.5	n.m.	n.m.	n.m.	n.m.	n.m.
Lancaster Sound											
CAA2	9.0	0.77	6.96	12.39	9.2	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.
CAA1	14.4	1.07	2.12	5.97	8.0	−21.6	n.m.	n.m.	n.m.	n.m.	n.m.
BB11	41.2	1.73	1.29	3.27	8.4	−22.0	−181.4	0.82	1550	−981.8	0.02
Davis Strait											
DS5	3.7	0.39	0.10	2.66	8.3	−20.4	−161.7	0.84	1360	n.m.	n.m.
DS2	18.8	0.42	1.27	4.34	8.0	−20.4	n.m.	n.m.	n.m.	n.m.	n.m.
DS1	19.6	0.55	1.03	2.61	7.7	−20.1	−150.0	0.86	1250	−990.6	0.01

[a]Mackenzie Shelf data from *Goñi et al*. [2003].
[b]n.m., not measured.

contribution from terrigenous OM sources derived from vascular plants relative to the other stations with lower C:N values. The range in C:N values measured in this study is comparable to previous studies of surface sediments from specific Arctic regions [e.g., *Naidu et al*., 2000; *Stein and Macdonald*, 2004; *Cooper et al*., 2009; *Iken et al*., 2010].

[16] The stable isotopic composition of organic carbon ($\delta^{13}C_{org}$) from surface sediments ranged from −20 to −26‰ (Table 2). As was the case with C:N values, there was marked variability in $\delta^{13}C_{org}$ among stations and regions. For example, stations along the Bering Sea, Chukchi shelf, Canadian Archipelago, Lancaster Sound, and Davis Strait had relatively enriched $\delta^{13}C_{org}$ values (> -22‰). In contrast, surface sediments from Barrow Canyon, Beaufort slope, and Mackenzie shelf displayed relatively depleted $\delta^{13}C_{org}$ values (<-24‰), suggesting enhanced contributions from ^{13}C-depleted, terrigenous OM sources. The $\delta^{13}C_{org}$ compositions determined in this study overlap with those from previous studies of specific areas of the North American Arctic margin [e.g., *Naidu et al*., 2000; *Stein and Macdonald*, 2004; *Cooper et al*., 2009; *Magen et al*., 2010; *Griffith et al*., 2012; *Connelly et al*., 2012].

[17] To further investigate the provenance of carbon in the study area, we determined the radiocarbon compositions of both organic and inorganic components ($\Delta^{14}C_{org}$ and $\Delta^{14}C_{inor}$, respectively) in surface sediments from selected stations (Table 2). In the case of $\Delta^{14}C_{org}$, moderately depleted values (−128 to −300‰) were measured in stations from the Bering shelf, Chukchi shelf, Canadian Archipelago, Lancaster Sound, and Davis Strait. In contrast, stations from Barrow Canyon, Beaufort slope, and Mackenzie shelf were characterized by significantly depleted

Table 3. Sediment-Normalized Yields (μg/g Sediment) of CuO Oxidation-Derived Biomarker Classes[a]

Sample	VP	SP	CP	CA	PB	BA	AA	DA	FA
Bering Sea Slope									
BS3A	4.41	3.74	*0.02*	0.58	7.70	4.78	97.9	7.76	16.22
Bering Sea Shelf									
SLIP1	25.7	15.1	5.38	11.8	42.0	25.8	606	36.3	106
SLIP3	20.4	12.4	5.49	10.6	27.8	19.9	489	33.6	95.1
SLIP4	35.5	20.1	7.03	12.5	96.7	50.2	1025	70.7	94.4
Chukchi Sea Shelf									
UTN3	22.0	14.3	5.52	9.70	37.2	20.0	506	30.4	91.0
UTN5	29.7	19.2	7.41	13.7	65.5	32.1	788	33.0	92.2
UTN7	52.2	31.6	11.4	17.4	178	88.4	1365	88.86	233
Barrow Canyon									
BC3	60.6	28.6	17.9	32.6	185	70.7	2076	373	658
BC4	122	78.3	23.5	45.2	118	59.3	581	66.7	94.6
BC5	59.5	39.4	13.3	26.5	61.7	34.6	485	39.1	61.0
BC6	87.9	57.7	19.0	38.6	81.1	40.6	326	33.0	29.2
Beaufort Sea Slope									
CG1	7.96	6.44	3.70	1.25	11.5	12.8	190	13.0	71.3
CG2	10.2	6.86	3.65	3.45	18.1	21.0	236	20.0	41.3
CG3	9.13	5.31	3.07	3.18	20.3	22.6	277	26.3	64.6
Mackenzie Shelf									
G1	65.4	38.2	10.3	9.19	27.5	27.9	78.3	43.3	34.2
G7	32.3	26.2	6.81	7.21	21.7	21.8	87.6	54.7	186
GRM1	33.7	25.6	5.94	6.76	17.7	23.6	69.8	36.1	36.9
G12	15.2	32.1	4.52	1.93	35.6	26.7	102	47.6	72.2
G9	14.6	17.7	4.62	2.06	13.8	15.4	60.6	30.3	49.7
G10	19.1	17.7	4.95	3.73	12.7	12.5	52.2	27.8	29.9
Canadian Archipelago									
QM1	1.23	1.82	*1.22*	*0.19*	16.1	8.86	289	11.8	33.9
VS1	*0.35*	*0.91*	*0.02*	*0.06*	9.79	9.94	260	10.6	11.5
FS1	*0.25*	*0.03*	*0.02*	*0.06*	6.39	5.31	165	7.28	29.7
BE2	1.15	*0.03*	*0.02*	1.09	8.66	7.29	215	14.0	32.7
PS1	0.59	*0.92*	*0.02*	*0.49*	14.1	16.5	311	20.7	62.1
PS2	*0.25*	*0.79*	*0.02*	*0.22*	7.84	3.34	117	14.4	29.8
Lancaster Sound									
CAA2	*0.38*	*1.04*	*0.02*	*0.10*	13.1	7.00	169	14.6	20.3
CAA1	6.56	5.82	*1.34*	*1.29*	36.2	30.9	321	33.3	65.1
BB11	3.23	1.40	*1.41*	*1.92*	33.6	46.1	473	36.9	40.5
Davis Strait									
DS5	2.72	*0.03*	*0.02*	0.92	10.9	8.37	187	13.4	27.3
DS2	*0.19*	*0.51*	*0.02*	*0.06*	3.78	4.67	115	4.37	8.88
DS1	*0.30*	*0.68*	*0.02*	0.42	10.3	7.45	157	9.46	15.1

[a]CuO product classes: VP, vanillyl phenols; SP, syringyl phenols; CP, cinnamyl phenols; CA, cutin acids; PB, p-hydroxybenzenes; BA, benzoic acids; AA, amino acid-derived products; DA, dicarboxylic acids; FA, fatty acids. Numbers in italic identify biomarker classes for which at least one individual product had yields that were at or below detection limit (<10 ng/g sediment).

$^{14}C_{org}$, values that ranged from −520 to −870‰. These compositions indicate that organic carbon in surface sediments from the central region of the North American Arctic margin (Barrow Canyon to Mackenzie shelf) has much lower relative contributions from modern sources ($0.1 < f_{mod} < 0.5$) and is, in a bulk sense, significantly older ($^{14}C_{age}$ 5850 to 16,750 ybp) than its counterparts in the western and eastern regions (f_{mod} of 0.7–0.9 and $^{14}C_{age}$ of 1040–2830 ypb, respectively; Table 2). We also measured the radiocarbon signature of inorganic carbon in selected stations and found that $\Delta^{14}C_{inor}$ signatures were highly depleted (<−980‰) in all the samples analyzed. These values are close to the limit of radiocarbon dating, indicating a negligible contribution of modern sources ($f_{mod} < 0.02$) and are consistent with very old ages ($^{14}C_{age} > 31{,}000$ ybp) for particulate inorganic carbon in surface sediments throughout the study area (Table 2). The ranges in $\Delta^{14}C_{org}$ compositions observed in our study are within the range of those measured in previous studies of sediments and water columns from specific areas of the Arctic Ocean [e.g., *Drenzek et al*., 2007; *Griffith et al*., 2012; *Vonk et al*., 2012].

3.3. Biomarker Yields

[18] Surface sediments from all stations yielded a variety of terrigenous and nonterrigenous biomarkers that suggest marked spatial differences in contributions from land-derived organic matter within the study area (Table 3). For example, yields of lignin-derived vanillyl phenols ranged from below detection limits (<0.5 μg/g sed) in several stations from the Canadian Archipelago and Davis Strait to ~90 μg/g sed in one of the Barrow Canyon stations (BC4). Similar distributions were observed for other terrigenous biomarkers such as syringyl phenols, cinnamyl phenols,

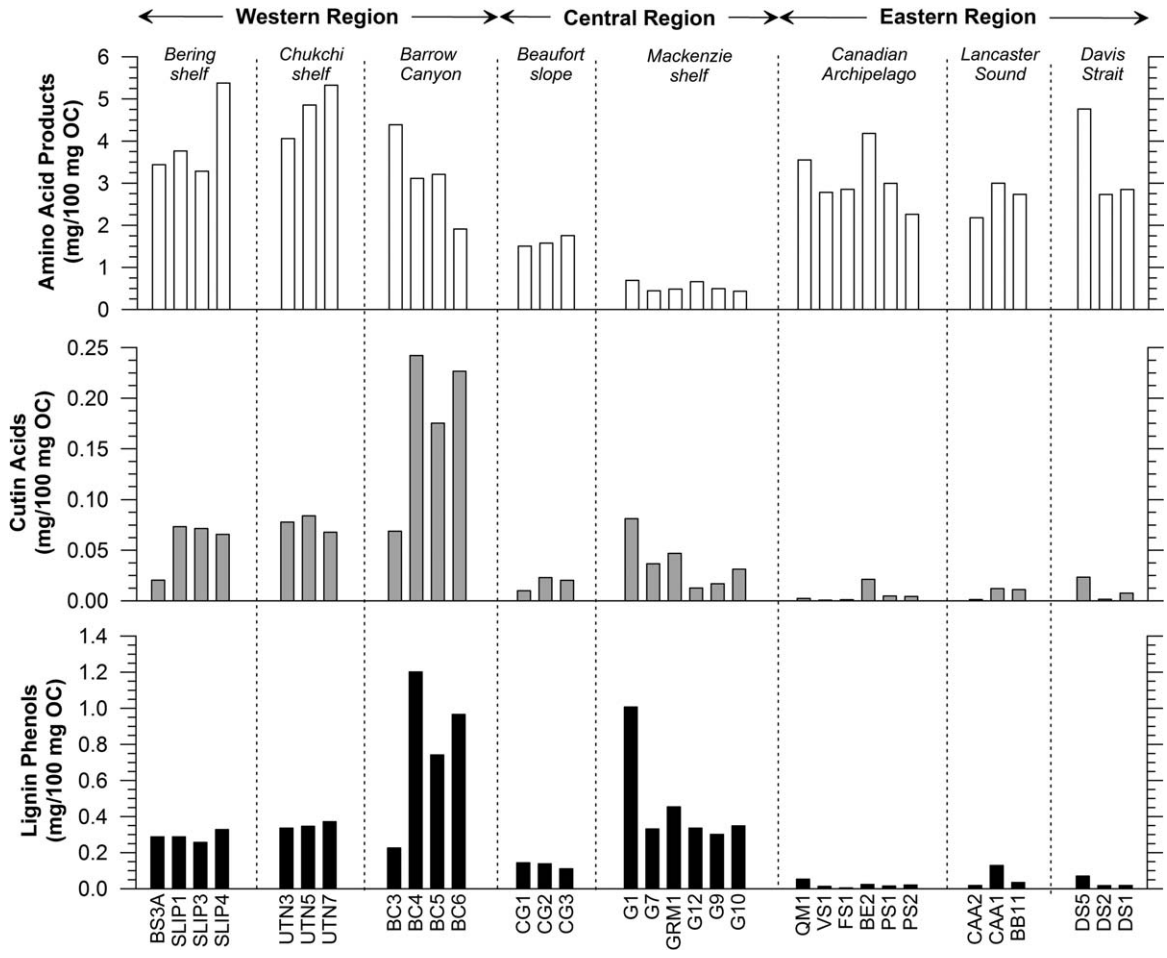

Figure 3. Organic carbon-normalized yields of selected biomarker classes from surface sediments along North American Arctic margin.

and cutin acids. For all of these compounds, which are uniquely synthesized by vascular land plants, sediment yields were highest for stations along Barrow Canyon, followed by those from the Mackenzie shelf, Chukchi shelf, Bering shelf, and Beaufort slope. The yields of these terrigenous biomarkers were much lower in stations from the Canadian Archipelago, Lancaster Sound, and Davis Strait.

[19] In contrast to lignin-derived and cutin-derived products, the yields of nonterrigenous biomarkers such as amino acid-derived products, *p*-hydroxybenzenes, and benzoic acids were comparable between stations in the eastern and western regions of the study area (Table 3). For example, the yields of amino acid products were highest (400 to over 1000 μg/g sed) in stations from Bering and Chukchi shelves but were also relatively high (>110 μg/g sed) in the Canadian Archipelago as well as Lancaster Sound and Davis Strait. The lowest yields of amino acid products (50–100 μg/g sed) were found in sediments from the Mackenzie shelf. Overall, yields of *p*-hydroxybenzene products were generally higher (40–180 μg/g sed) among stations from the western region of the margin (Bering shelf, Chukchi shelf, and Barrow Canyon) relative to eastern sites. Benzoic acid products had a more mixed distribution, with elevated yields (>50 μg/g sed) measured in stations from the Bering shelf, Chukchi shelf, Barrow Canyon, Mackenzie shelf, and Lancaster Sound.

[20] Because biomarker yields (i.e., μg/g sediment) can vary due to dilution by mineral matter, it is useful to normalize them to the organic carbon content of each sample (Figure 3). For example, OC-normalized yields of amino acid products ranged from <1 to over 5 mg/100 mg OC and displayed clear geographical contrasts. The highest values were measured in the western regions of the study area, followed by the eastern regions while the values in the central Beaufort Sea region were the lowest. Because amino acid products have a predominant nonvascular plant source, e.g., phytoplankton, zooplankton, bacteria [e.g., *Goñi and Hedges*, 1995], their distributions reflect relatively greater contributions from marine organic matter sources (versus terrestrial sources) to surface sediments from the western and eastern regions of the study area.

[21] The OC-normalized yields of cutin acids and lignin phenols, both land-derived vascular plant biomarkers, ranged from ~0 to over 0.2 mg/100 mg OC and 0.05 to over 1.0 mg/100 mg OC, respectively (Figure 3). Both of these compound classes displayed regional distributions that were consistent with very low contributions of OM from terrestrial land plant sources in the eastern region of the study area. Surface sediments from the central and western regions were characterized by intermediate cutin acid and lignin phenol yields that suggest moderate contributions from land-plant sources. The major exceptions were samples from

Barrow Canyon, especially those from the deeper parts (BC4, BC5, and BC6) and the near-shore (G1, 5 m depth) sample from the Mackenzie shelf, all of which were characterized by terrigenous biomarker yields that were 2–3 times greater than those from adjacent locations. These patterns indicate that contributions from terrestrial plant sources to the OM in surface sediments are much higher at these locations than at all other stations along the margin. Furthermore, the differences in the distributional patterns of cutin acids and lignin phenols observed among surface sediments from the Mackenzie shelf and Barrow Canyon (Figure 3) suggest that there are contrasts in the composition and provenance of the land-derived OM in these areas.

3.4. Multivariate Comparison of Sediment Composition Using PCA

[22] The multivariate method of PCA used in this study, which is based on the relative proportions of CuO oxidation products in each sediment sample, provides a robust overview comparison of sediment compositions in different parts of the North American Arctic margin [*Yunker et al*., 1995, 2005, 2011]. PCA operates with no a priori assumptions about the data structure and readily illuminates the major underlying trends of a large data set by grouping variables that covary and samples that have similar compositions with respect to the variables being examined. PCA decomposes a data set into a series of orthonormal rank 1 matrices (a matrix that cannot be expressed in a simpler form) or PCs, where each PC is the outer product of a scores vector (t1, t2, etc.) for the samples and a loadings vector (p1, p2, etc.) for the variables [*Geladi and Kowalski*, 1986; *Meglen*, 1992]. Variable loadings near zero indicate essentially no contribution to a PC, while the contribution to a PC increases as the absolute magnitude of the variable loading (and distance from axis centre) increases.

[23] Because samples were normalized prior to multivariate analysis, the PCA model separates samples based on the relative proportions of each variable in each individual sample. In the plot of individual PCA variables (Figure 4a), vanillyl (VP), cinnamyl (CP) and syringyl (SP) phenols, and cutin acids (CA) project on the left side, while benzoic acids (BA), dicarboxylic acids (DA), amino acid products (AA) and most *p*-hydroxybenzenes (PB), and fatty acids (FA) project on the right side. These relationships can be seen more clearly in Figure 4b, where ellipses show the mean ± one standard error for the variables in each of the CuO product families (33 of the 39 variables in the PCA model), and the size of the ellipse provides a measure of how characteristic a family is a as source indicator. In particular, the VP, CA, DA, BA, and AA families project in a relatively small range in the first PC, producing ellipses that are narrow in the *x* axis direction, and these families will have good source discrimination for the largest source of variation in samples (PC1 captured 51.3% of the overall variation in the data set).

[24] The PB and FA families show wider ellipses in PC1 because one variable in the group projects on the other side of the *y* axis (Pn: *p*-hydroxyacetophenone and C10FA, respectively, or one out of three and seven variables). In the case of PB, the separation of Pn relative to Pl and Pd is consistent with their distinct biochemical sources. The latter two compounds are major oxidation products of the amino acid tyrosine [*Goñi and Hedges*, 1995] and thus plot in the same region as AA products. In contrast, Pn has been shown to be a product of nonwoody lignin [*Benner and Kaiser*, 2011] and its plotting in the same region as the other terrestrial indicators (VP, CA, SP, and CP) is a reflection of its land-derived source. Most of the remaining variables in these two groups have good source discrimination. The CP family has only one variable, while SP has two variables (Sl: syringealdehyde and Sn: acetosyringone) projecting on the left side of the *y* axis (Figures 4a and 4b).

[25] Accordingly, in the corresponding samples plot (Figure 4c), samples from the Barrow Canyon, Mackenzie shelf, Chukchi shelf, Bering shelf, and Beaufort slope, which had high proportions of vanillyl, syringyl and cinnamyl phenols, and cutin acids, projected on the left side, while samples from Davis Strait, Lancaster Sound, the Canadian Archipelago, and Bering Sea slope, which had high proportions of the amino, dicarboxylic and fatty acid products and most *p*-hydroxybenzenes, projected on the right side. In the second PC, which captured 12.2% of the overall variance, Mackenzie shelf samples projecting in the top part of the samples plot had higher proportions of most dicarboxylic acids, and a few fatty acids, while the other western Arctic samples projecting in the lower part of the plot had higher proportions of amino acid products (particularly 2-carboxypyrrole and *p*-hydroxyphenylglyoxylic acid, Cp and Pg, respectively) and two minor vanillyl products (5-carboxyvanillic acid and vanillylglyoxalic acid, 5cV and Vg, respectively) and to a lesser extent the *p*-hydroxybenzenes (Figures 4a–4c).

[26] The first PC of the PCA model thus ranked samples according to the relative input from terrestrial vascular plant sources, with the sediments from Barrow Canyon (BC4–6) and the near-shore (G1) sample from the Mackenzie shelf having the highest amount of plant input and the sediments from the Canadian Archipelago and Davis Strait having the least amount of plant input (Figure 4c). The main effect of the second PC was to separate the Mackenzie shelf samples from all other western Arctic samples, which are from locations further offshore and away from a major river (Figure 4c).

[27] On a proportional basis, amino acid derivatives dominated in most of the samples. Barrow Canyon and Mackenzie shelf samples had the highest proportions of lignin products, cutin acids, *p*-hydroxybenzenes, and benzoic acids, with notably higher proportions of fatty and dicarboxylic acids in the Mackenzie shelf and Beaufort slope samples (Figure 4d). The sample projections in the PCA model exhibit the largest (positive) correlation between t1 and %IC or $\%CaCO_3$ ($r^2 = 0.62$, $p < 0.0001$) with weaker (positive) correlations between t1 and the isotopes and the other bulk parameters ($r^2 < 0.30$, $p < 0.001$ in most cases).

4. Discussion

[28] Our results are consistent with the presence of a spatially variable mixture of autochthonous and allochthonous sources of OM in surface sediments along the North American margin of the Arctic Ocean. Such variability is apparent in the results from previous studies [e.g., *Peulve et al*., 1996; *Yunker et al*., 2005; *Goñi et al*., 2005; *Belicka et al*., 2009; *Vonk et al*., 2012, and references therein]. However, unlike other efforts that relied on different analytical tools,

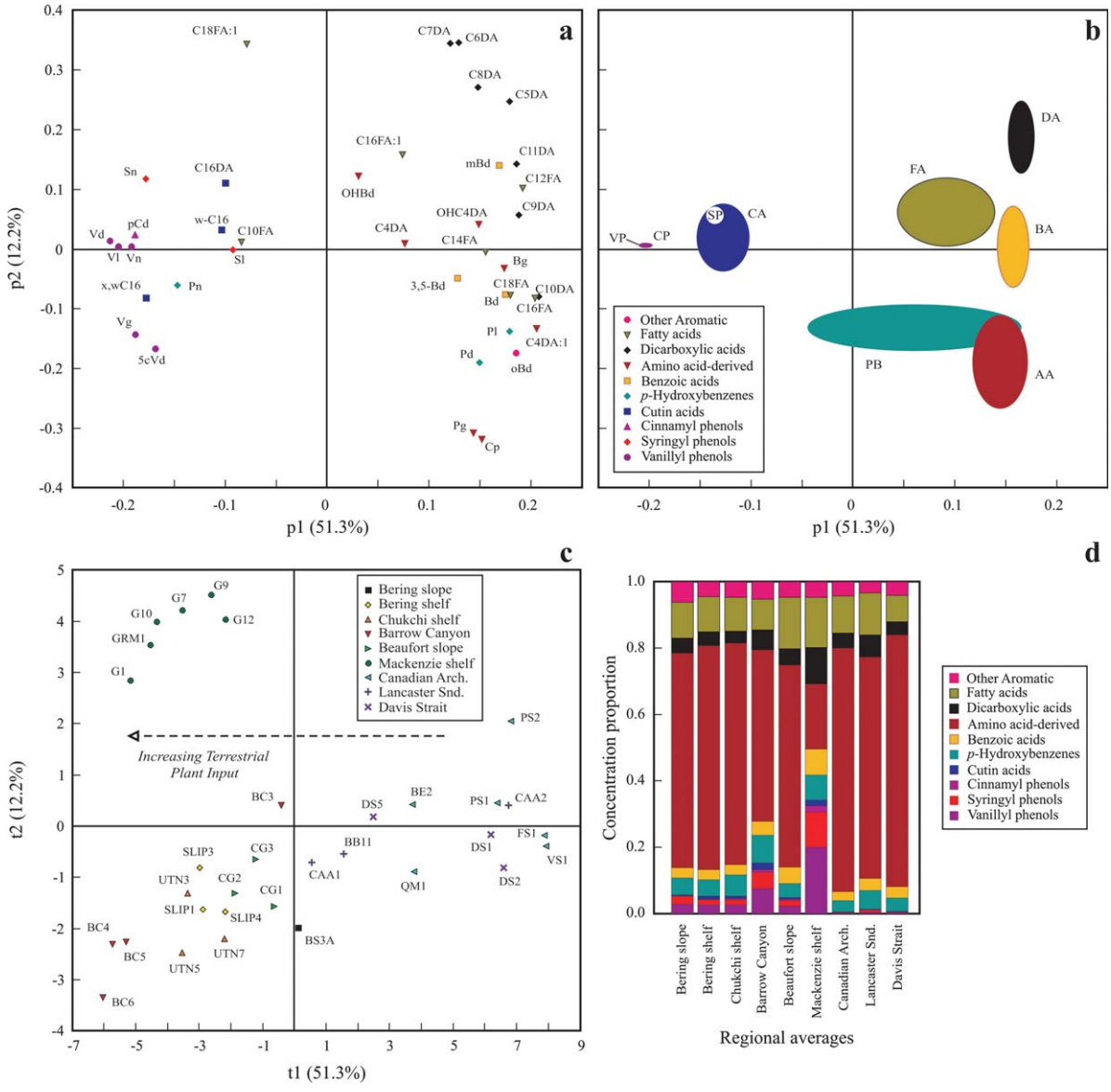

Figure 4. Results from principal component analysis (PCA) of individual biomarker yields**.** The PCA model based on 39 CuO oxidation variables (supporting information) separates the 32 surface sediment samples spanning the margins of the North American Arctic according to the relative input from terrestrial plant sources. The separate figures show (a) variable projections in the first two PCs, (b) ellipses showing the mean ± one standard error (for $n > 2$) for the variables in each of the CuO product families, (c) sample projections for the sediment samples, and (d) regional averages of the concentration proportions of each CuO oxidation family.

our study provides a comprehensive and analytically consistent characterization of sedimentary organic matter across a broad range of locations that span the whole North American Arctic margin. This data set allows us to investigate first-order controls on the concentration and composition of OM present in the shelf and slope sediments of this high-latitude ocean margin and provides a baseline against which future compositions may be compared when investigating climate-induced changes.

4.1. Controls in Distribution of OC in Surface Sediment

[29] Although the amount of OC preserved in margin sediments is affected by a variety of factors, including surface productivity, organic matter fluxes to the benthos, and exposure to effective oxidants (i.e., oxygen), the relationship between mineral surface area and OC content [e.g., *Mayer*, 1994; *Keil et al*., 1994] provides a means to evaluate the overall loadings of organic matter under different depositional conditions [see reviews by *Hedges and Keil*, 1995; *Blair and Aller*, 2012]. In this study, it is evident that the various regions along the North American Arctic margin display a wide range of OC loadings, from 0.25 to over 1.0 mg C m^{-2} of sediment (Figure 5). In particular, BC3 from Barrow Canyon displayed exceptionally high OC loadings (>2.0 mg C m^{-2} sediment), similar to those observed in sediments from regions of high productivity

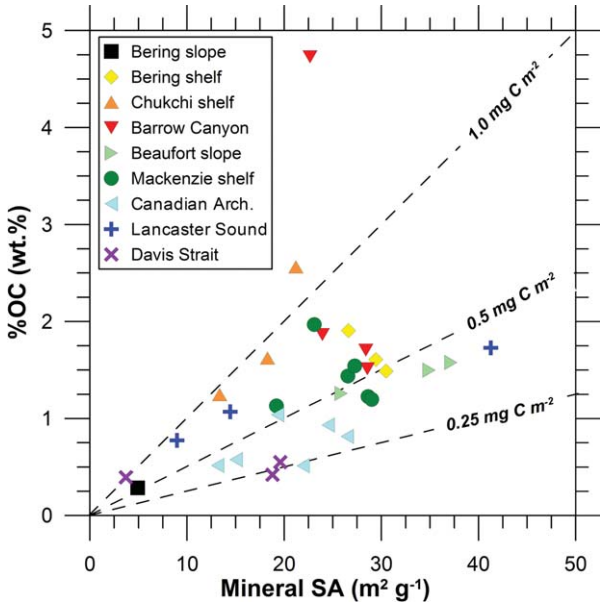

Figure 5. Relationship between mineral surface area (SA) and weight percent organic carbon (%OC) contents of surface sediments from the North American Arctic margin. Dashed lines showing various levels of carbon loadings (mg C m^{-2} of sediment) are plotted in the graph for reference.

and low dissolved-oxygen concentrations [e.g., *Hedges and Keil*, 1995]. The head of Barrow Canyon region is recognized as a productivity "hot spot" partly due to dynamic physical mixing [*Shroyer*, 2012] and upwelling of offshore, high-nutrient upper halocline waters [*Christensen et al*., 2008; *Cooper et al*., 2009; *Mathis et al*., 2009]. Annual primary production in this area has been estimated at 430 g C m^{-2} yr^{-1} (see Table 1 in *Grebmeier et al*., [2006 and references therein]), which is probably the second-highest estimate for anywhere in the Arctic (behind only the southern Chukchi Sea, which is estimated at 470 g C m^{-2} yr^{-1}). The high OC:SA values from surface sediments in the BC3 core may, therefore, reflect elevated OC fluxes to the benthos from the highly productive overlying water column at this location.

[30] Organic carbon loadings in sediments from other regions with high primary production, i.e., the Chukchi shelf and Lancaster Sound, tend to lie along the 1 mg C m^{-2} line, while remaining stations exhibit lower OC:SA values (0.25–1.0 mg C m^{-2} sediment), which are "typical" of most margin sediments. Within each area, e.g., Beaufort slope, Mackenzie shelf, Canada Basin, Lancaster Sound, and Davis Strait, stations from deeper water depths tend to have lower OC loadings (0.25–0.5 mg C m^{-2}) than those from shallower locations. However, over the whole data set there is no significant relationship between OC:SA and water depth, sediment accumulation rate, and/or surface mixed layer.

[31] The total amount of organic matter present in surface sediments is determined partly by the supply of organic carbon and partly by metabolic processing during transport and within the seabed. Here, the marine carbon supply is controlled by local productivity, which is regionally variable, the depth of the water column, which affects the vertical flux [e.g., *O'Brien et al*., 2013; *Honjo et al*., 2010], and sediment transport processes, which can focus particle fluxes in locations such as the Barrow Canyon and enhance delivery of fresh material to the abyss [*Macdonald and Gobeil*, 2012]. Metabolism, on the other hand, depends on exposure to efficient oxidants, mainly oxygen and metal oxides [e.g., *Hartnett et al*., 1998; *Hedges et al*., 1999; *Meile and van Cappellen*, 2005]. A plot of the thickness of the Mn-rich layer at the sediment surface, here used as a proxy for in situ redox conditions and the effective penetration depth of oxygen into the sediment column at each site, versus OC:SA ratios (Figure 6) shows that the highest OC loadings tend to occur in stations with the thinnest Mn layers (i.e., shallowest oxygen penetration). These samples include those from Barrow Canyon, the Chukchi and Bering shelves, and selected locations from Lancaster Sound (CAA2) and Davis Strait (DS5), all of which have Mn-layer thicknesses of less than 2 cm. As exposure to efficient oxidants increases in surface sediments (as indicated by greater Mn-layer thicknesses), there is a rapid decline in OC:SA values, so that samples with Mn-layer thicknesses >5 cm all have OC loadings of less than 0.5 mg C m^{-2} (Figure 6). Most of the samples in this latter category are from the Canadian Archipelago, but also include deep-water (>500 m) samples from the Beaufort slope (CG2 and CG3) and Davis Strait (DS1 and DS2). In addition to variable exposure to oxidants, contrasts in other factors, such as the magnitude of organic matter fluxes, the degree of alteration during transport, and the composition of mineral and organic matrices, can also contribute to differences in OC loadings [e.g., *Keil et al*., 2004; *Goñi et al*., 2005, 2008; *Tesi et al*., 2010; *Connelly et al*., 2012; *Ransom et al*., 1998; *Mead and Goñi*, 2008]. The variability observed in Figures 5 and 6 suggests that some of these other factors are likely important. Certainly, in the Canadian Archipelago, the influence of low fluxes on the low organic matter loadings can be inferred from the very low inventories of ^{210}Pb in these sediments [*Kuzyk et al*., 2013].

4.2. Controls in Distribution of IC in Surface Sediment

[32] The concentration of inorganic carbon in surface sediments displays a linear relationship with Ca contents (Figure 7), indicating $CaCO_3$ minerals (i.e., calcite, aragonite) are responsible for sedimentary carbonate distributions. Ca contents in the Canadian Archipelago and Lancaster Sound, which are below the 1:1 relationship expected if all the sedimentary carbonate was made up of $CaCO_3$, indicate that dolomite ($CaMg(CO_3)_2$) may be another important carbonate source in this region. Adding Mg concentrations to the Ca contents in these samples shifts their position to the right in Figure 7, resulting in compositions that are much closer to a 1:1 relationship with carbonate content. The presence of dolomite in these sediments is consistent with surveys that show much of the exposed bedrock in the Canadian Archipelago includes dolomitic carbonates [e.g., *Hamilton et al*., 2001; *Turner and Kamber*, 2012] and that erosion processes [*Bischof and Darby*, 1999; *Stokes et al*., 2005; *Niessen et al*., 2010] result in widely distributed terrestrial glacial sediments that have high calcite and dolomite contents [*Andrews et al*., 1991].

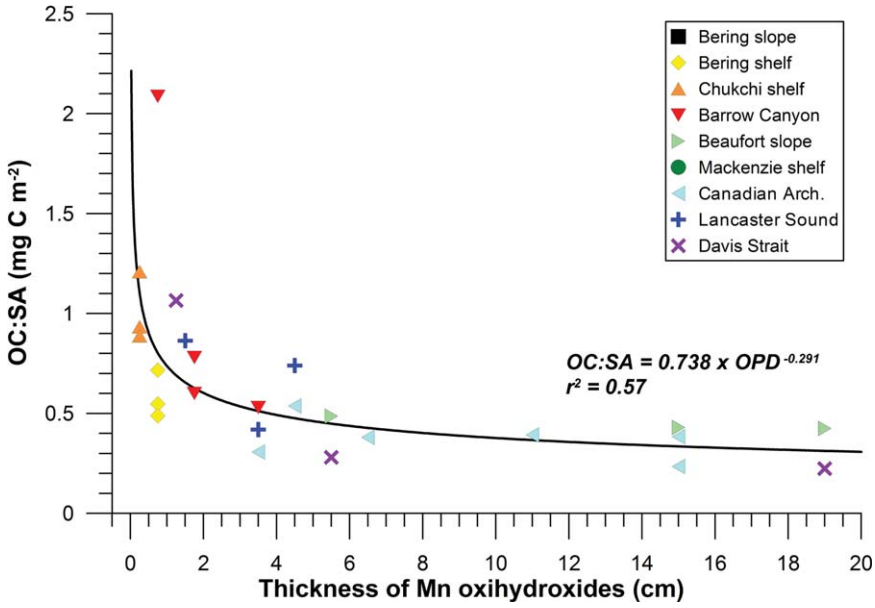

Figure 6. Relationship between the thickness of the layer containing Mn oxihydroxydes at each coring site and the surface area-normalized organic carbon loading (OC:SA) of the top horizon from each core along the North American Arctic margin.

[33] The central Canadian Archipelago, similar to Hudson Bay, is still undergoing significant isostatic rebound associated with ice sheet unloading [*Sella et al*., 2007]. Resuspension and lateral transport of shallow water, largely glacigenic deposits, which have been brought up into the wave base as a consequence of rebound and relative sea-level fall, may therefore be an important source of such materials, as proposed in Hudson Bay [*Kuzyk et al*., 2008, 2009]. The highly depleted ^{14}C compositions of IC ($\Delta^{14}C < -980‰$; Table 2) corroborate the notion that the carbonate is derived, for the most part, from detrital sources and not from modern biogenic production by biota forming calcite or aragonite hard parts. Large contributions of land-derived glacial material to the Canadian Archipelago margin sediments are consistent with the low organic carbon contents and loadings in these sediments and with their low proportions of terrestrial vascular plant material as interpreted from the principal component analysis of the biomarker data (Figure 4).

[34] Allochthonous detrital sources of IC appear to be important not only in Canadian Archipelago/Lancaster Sound sites but also, to a lesser extent, in Barrow Canyon and the Beaufort slope. The Mackenzie shelf sediments display %IC contents of 0.3–2 wt %, consistent with previous studies indicating inputs of detrital carbonate from dolomitic deposits in the Mackenzie River watershed [*Pelletier*, 1987, 1986]. In Figure 7, sediments from the Bering and Chukchi Seas (and one sample from Davis Strait) display Ca contents that are higher than expected based on their low IC contents. It is likely that the Ca in these samples does not originate from carbonate minerals but rather is associated with clay minerals and other noncarbonate phases.

4.3. Composition and Sources of OM in Surface Sediments

[35] Because of the heterogeneity in potential sources of OM to the Arctic Ocean, it is important to apply a combination of bulk and biomarker tracers to investigate its provenance in sedimentary samples [e.g., see *Belicka et al*. 2009; *Yunker et al*., 1995, 2005]. Figure 8a shows an overall positive relationship between the $\delta^{13}C$ and N:C signatures of OM in surface sediments from the study area, which suggests, along with the PCA model (Figure 4), that most sedimentary compositions can be explained as

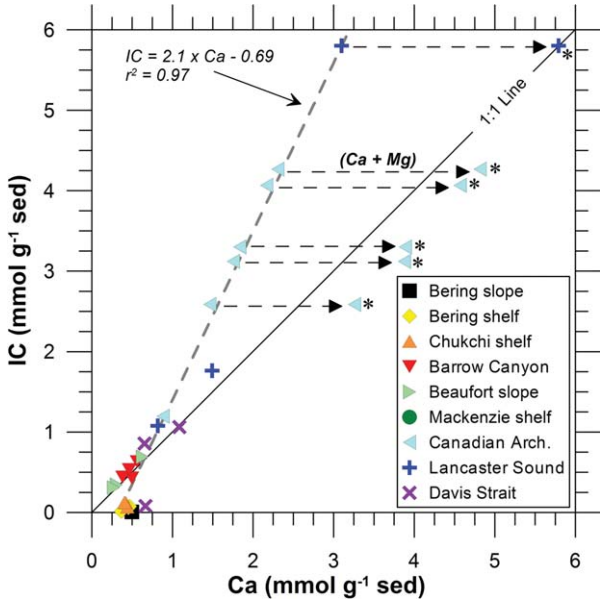

Figure 7. Relationship between calcium (Ca) and inorganic carbon (IC) contents in surface sediments from the North American Arctic margin. A linear fit of the data is indicated with the dotted line. The 1:1 molar Ca:IC line is provided for reference. The symbols marked with * indicate the combined concentration of calcium + magnesium for selected samples (see text for details).

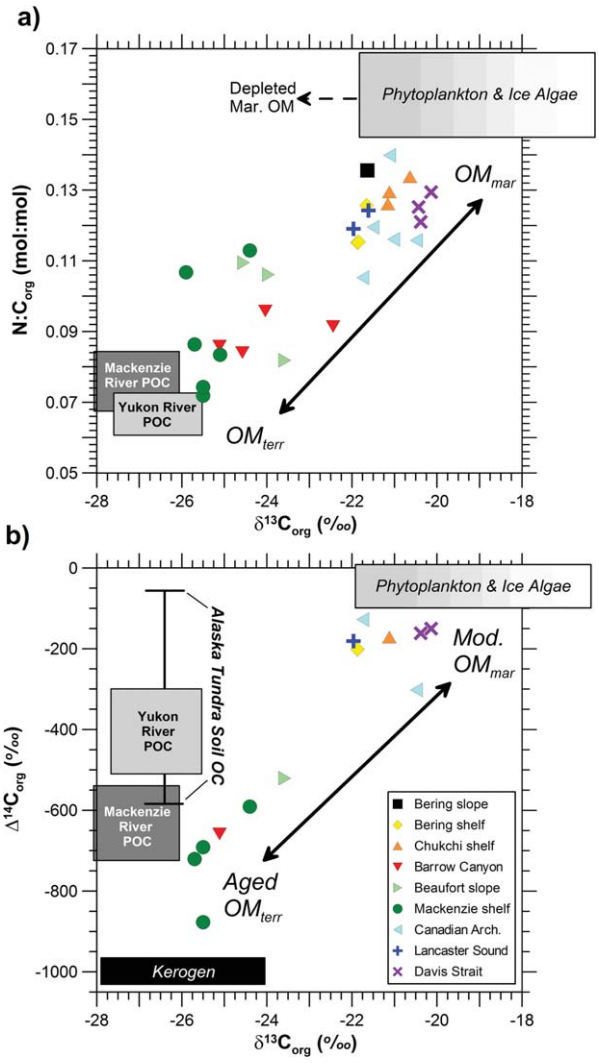

Figure 8. Plots of (a) molar carbon:nitrogen ratios ($N{:}C_{org}$) and (b) radiocarbon compositions ($\Delta^{14}C_{org}$) versus stable carbon compositions of bulk organic matter ($\delta^{13}C_{org}$) in surface sediments from the North American Arctic margin. The compositional ranges of suspended particulate organic matter from the Mackenzie River [*Goñi et al*., 2005] and the Yukon River [*Guo et al*., 2007], bedrock-derived kerogen [*Lewan*, 1986] and marine phytoplankton and ice algae [*Schubert and Calvert*, 2001] are included in the plots. Note that the $\Delta^{14}C_{org}$ compositional ranges of phytoplankton and ice algae assume that they reflect the radiocarbon compositions of DIC in regional surface waters [e.g., *Griffith et al*., 2012].

mixtures of marine organic matter (OM_{mar}) and terrigenous organic matter (OM_{terr}) sources. There is marked variability in the compositional characteristics of individual samples, but based on the bulk tracers (Figure 8a), it appears that sediments from the Mackenzie shelf, Barrow Canyon, and Beaufort slope are characterized by higher contributions from OM_{terr} relative to the other areas in the eastern (Davis Strait, Canadian Archipelago) and western (Bering and Chukchi Seas) regions of the North American margin of the Arctic Ocean. The contrast between the Beaufort and Chukchi/Bering samples is consistent with previous studies [e.g., *Naidu et al*., 2000; *Macdonald et al*., 2004], whereas the compositions of sediments from Lancaster Sound and the Canadian Archipelago are novel given the paucity of information on sedimentary OM from this region.

[36] The variability exhibited among the analyzed samples reflects broad variations in the compositions of OM_{mar} [e.g., *Schubert and Calvert*, 2001; *Magen et al*., 2010; *Connelly et al*., 2012] and OM_{terr} endmembers [e.g., *Guo et al*., 2004, 2007; *Goñi et al*., 2005] along the Arctic Ocean margin. Hence, for example, the $\delta^{13}C$ signatures of OM_{mar} can range from relatively enriched values (−15 to −18‰) in the case of ice algae to relatively depleted values (−20 to −26‰) in cases of slow-growing phytoplankton under high pCO_2 conditions [*Schubert and Calvert*, 2001; *Stein and Macdonald*, 2004]. Furthermore, contrasts in degradation history also can result in isotopic and elemental compositional differences [e.g., *Magen et al*., 2010; *Connelly et al*., 2012]. Similarly, although suspended sediments from the Mackenzie and Yukon Rivers plotted in Figure 8 display comparable N:C and $\delta^{13}C_{org}$ compositions, OM_{terr} inputs to the North American Arctic include multiple sources (e.g., vegetation, soils, bedrock) exported from fluvial/glacial watersheds and mobilized by coastal erosion [e.g., *McGuire et al*., 2009; *Ping et al*., 2011; *Vonk et al*., 2012]. Such end-member variability is reflected in the distinct projections along the PC2 axis (t2 in Figure 4c) for samples from different regions (e.g., Barrow Canyon versus Mackenzie shelf) with high OM_{terr} contributions and helps explain the variation along the general mixing line in Figure 8a.

[37] The stable and radiocarbon compositions of selected surface sediment samples also exhibit a general positive relationship (Figure 8b) that is consistent with variable mixtures of modern OM_{mar} and aged OM_{terr}. Sediments from the Beaufort Sea (Barrow Canyon, Mackenzie shelf, and Beaufort slope) are enriched in OM_{terr} that is significantly older than the OM_{mar}. Sediment samples with organic matter most enriched in ^{13}C display $\Delta^{14}C$ values that are somewhat lower than those expected for algal sources utilizing DIC from surface waters [e.g., *Griffith et al*., 2012], indicating that these samples contain predominantly modern OM_{mar}. It is important to note that the most negative $\Delta^{14}C$ values in samples from the Mackenzie shelf and Barrow Canyon are consistent with significantly older ages than those of OM in suspended samples from the Yukon River [*Guo and Macdonald*, 2006], and at least in one case, also older than OM in suspended samples from the Mackenzie River [*Guo et al*., 2007; *Goñi et al*., 2005]. These results suggest that there are sources of OM_{terr} entering these regions that are older than the bulk OM exported by these rivers and/or that the fraction most depleted in ^{14}C is preferentially preserved in surface sediments. For example, the Mackenzie River delivers petrogenic hydrocarbons to the Beaufort Sea and Canadian Basin principally as eroded Devonian shales or bitumens [*Yunker et al*., 2002, 2011] and this material would be expected to be preferentially preserved over plant-related terrigenous debris. In Figure 8b, we include the compositional ranges for kerogen (bedrock-derived, fossil carbon), which along with old OM in deep permafrost soils, could be the source of such ^{14}C-depeleted materials. The differences in $\Delta^{14}C$ between suspended sediments from the Yukon and Mackenzie Rivers

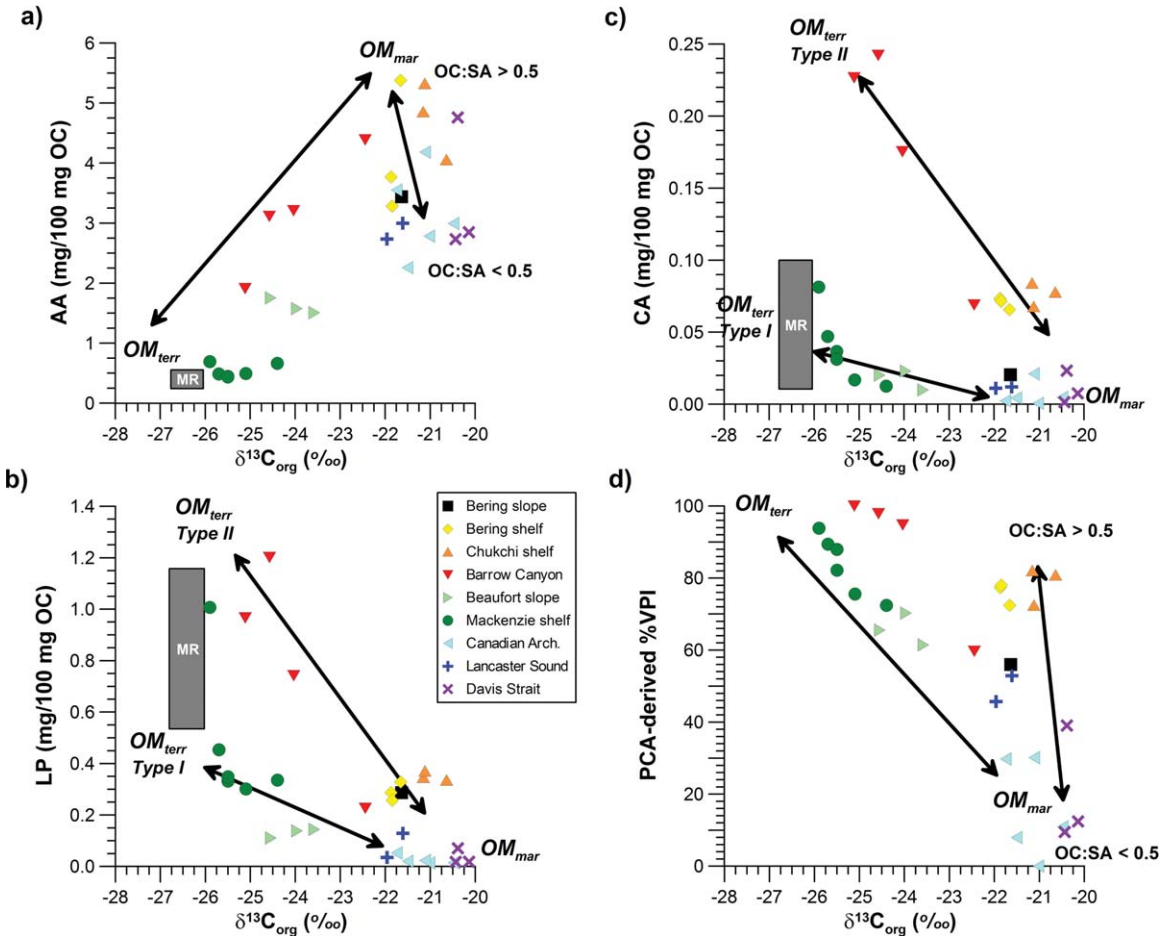

Figure 9. Plots of organic carbon-normalized yields of (a) amino acid-derived products (AA), (b) lignin phenols (LP), (c) cutin acids (CA), and (d) PCA-derived relative percent terrestrial vascular plant inputs (%VPI, from the relative distance along t1 in Figure 4c; values provided in Table 4) versus stable carbon compositions of bulk organic matter ($\delta^{13}C_{org}$) in surface sediments from the North American Arctic margin. See Figure 8 and text for details. The compositional ranges of suspended particulate organic matter from the Mackenzie River (MR) [*Goñi et al*., 2005] are included for reference.

illustrate the heterogeneity of OM_{terr} sources (both biogenic and petrogenic) and highlight the need for additional characterization in order to better quantify terrigenous contributions to OM in Arctic sediments.

[38] One way to further differentiate OM contributions from the various sources is to combine biomarker-based parameters with bulk compositions [e.g., *Goñi et al*., 2008; *Tesi et al*., 2010]. In the case of samples analyzed in this study, Figure 9a shows a general trend of increasing OC-normalized yields of amino acid products with enriched $\delta^{13}C_{org}$ compositions. Because these products are predominantly derived from nonvascular plant sources, we conclude, as we did based on the N:C versus $\delta^{13}C_{org}$ plot (Figure 8a), that there are enhanced contributions of protein-rich OM_{mar} as we move from Mackenzie shelf to Beaufort slope and Barrow Canyon sediments. Most other sediment samples deviate from this trend and instead display decreasing yields of amino acid products while retaining relative enriched $\delta^{13}C_{org}$ values. Among the Canadian Archipelago samples, contributions from reworked coastal (largely glacigenic) sediments, in which OM is highly degraded, may in part explain this trend. Furthermore, examination of the OC:SA values shows that samples with lower yields of amino acid products have lower surface area-normalized OC loadings ($0.3 > OC:SA > 0.5$) relative to their counterparts with higher amino acid products ($0.6 > OC:SA > 1.2$). Therefore, we conclude that the variations in amino acid products at nearly constant $\delta^{13}C_{org}$ values displayed by samples from the eastern and western regions of the study area reflect contrasts in OC preservation, which are related to the provenance of the material and the redox conditions and oxidation history at each site (see Figure 6).

[39] Plots of terrigenous biomarkers and the relative percentage of terrestrial vascular plant input (%VPI, based on the PCA model) versus $\delta^{13}C_{org}$ highlight contrasts in the compositions and provenance of OM_{terr} in surface sediments along the North American Arctic margin (Figures 9b–9d). Our results show that the OC-normalized yields of both lignin phenols and cutin acids and the %VPI reflected in t1 of the PCA are negatively correlated with $\delta^{13}C_{org}$, consistent with higher contributions of OM_{terr} in those samples with more depleted $\delta^{13}C_{org}$ values. However, in all cases, the relationships show distinct trends for samples

from different regions. For example, the Mackenzie shelf, Beaufort slope, Lancaster Sound, Bering slope, and Canadian Archipelago samples all plot along a trend line that could be interpreted as mixing between an OM_{terr} endmember with relatively low lignin and cutin contents (OM_{terr} Type I) and a ^{13}C-enriched OM_{mar} endmember devoid of terrigenous biomarkers. In contrast, samples from Barrow Canyon, Bering shelf, Chuckchi shelf, and the inshore sample (G1) from Mackenzie shelf display a different trend that suggests a mixture of OM_{mar} with a terrigenous endmember (OM_{terr} Type II) that is much more enriched in lignin and cutin biomarkers. Possible sources of OM_{terr} include old mineral soils and bedrock (Type I) and organic-rich soils and peats (Type II). These distinct sources may help explain the trends along the PC2 axis in the PCA results, which showed marked separation in t2 for samples from Barrow Canyon and the Bering and Chukchi shelves and those from the Mackenzie shelf (Figure 4c).

[40] The relative terrestrial vascular plant inputs in each sediment sample calculated from the PCA model (%VPI in Table 4) also show variable relationships with $\delta^{13}C_{org}$ compositions (Figure 9d). As we observed in the case of amino acid product yields (Figure 9a), samples with enriched $^{13}C_{org}$ values ($>\sim-22‰$) display variations in relative terrestrial contributions that are positively related to carbon loadings. Thus, for a given (enriched) $^{13}C_{org}$ value, samples with higher %VPI (i.e., those from Bering and Chukchi shelves) had higher OC:SA ratios (>0.5) than those with lower terrestrial contributions (samples from Davis Strait and Canadian Archipelago). Such a trend suggests that redox conditions in sediments affect the preservation of not only OM_{mar} but also OM_{terr}. As was the case in the plots of lignin phenols and cutin acids versus $^{13}C_{org}$ compositions (Figures 9b and 9c), the samples from Barrow Canyon and Bering and Chukchi shelves display a distinct relationship with $\delta^{13}C_{org}$ relative to samples from the Mackenzie shelf and Beaufort slope. Again, these differences are consistent with the PCA model results (Figure 4c) and suggest regionally distinct sources of OM_{terr} along the North American Arctic margin.

[41] One way to further explore the possibility that there are regional differences in the provenance of OM_{terr} is to investigate the compositional signatures of specific terrigenous biomarkers by investigating trends in their ratios (Table 4). For example, marked regional contrasts in the abundances of cutin acids relative to lignin phenols (CA:LP ratios) are evident between surface sediments from the western region of the study area (Bering shelf, Chukchi shelf, Barrow Canyon all with CA:LP > 0.15) and those from the Mackenzie shelf and Canadian Archipelago (CA:LP < 0.1). Compositionally, all the lignin appears to be angiosperm derived, with syringyl:vanillyl phenol ratios > 0.5 for all samples analyzed (see Table 4). However, the composition of cutin acids reveals that terminal hydroxylated fatty acids (w-OH) make up a much greater fraction of the total cutin acids in samples from the Mackenzie shelf, Mackenzie River, Canadian Archipelago, and Lancaster Sound (w-OH:CA > 0.4) compared to those from the Bering shelf, Chukchi shelf, Barrow Canyon, and Beaufort slope. Elevated w-OH:CA values have been measured in soils and sediments where contributions from subsurface vascular plant biomass (i.e., roots) dominate over contribution from aerial tissues (i.e., leaf litter) [*Crow et al*., 2009a, 2009b]. Thus, it is possible that the contrasts in CA:LP and w-OH:CA ratios reflect contributions from different sources, such as for example, watershed soils mobilized by fluvial and glacial erosion versus inputs from the erosion of coastal permafrost soils [e.g., *Ping et al*., 2011; *Vonk et al*., 2012].

[42] In addition to distinct sources, we have to consider that differences in the transport and cycling of OM_{terr} among the different regions of the North American Arctic margin may contribute to the contrasts illustrated in Figure 9 and Table 4. For example, work on other margins has shown that fine, lithogenic-rich particles derived from

Table 4. Compositional Parameters Based on Terrestrial Biomarker Ratios[a]

Sample	SP:VP	CP:VP	CA:LP	w-OH/CA	w,x-OH/CA	% VPI
Bering Sea Slope						
BS3A	0.85	0.00	0.06	0.00	1.00	56
Bering Sea Shelf						
SLIP1	0.59	0.21	0.26	0.19	0.81	77
SLIP3	0.61	0.27	0.28	0.18	0.82	78
SLIP4	0.57	0.20	0.20	0.18	0.82	72
Chukchi Sea Shelf						
UTN3	0.65	0.25	0.23	0.18	0.82	81
UTN5	0.65	0.25	0.24	0.18	0.82	82
UTN7	0.60	0.22	0.18	0.20	0.80	72
Barrow Canyon						
BC3	0.47	0.30	0.30	0.62	0.38	60
BC4	0.64	0.19	0.20	0.19	0.81	98
BC5	0.66	0.22	0.24	0.17	0.83	95
BC6	0.66	0.22	0.23	0.16	0.84	100
Beaufort Sea Slope						
CG1	0.81	0.47	0.07	0.05	0.95	61
CG2	0.67	0.36	0.17	0.19	0.81	70
CG3	0.58	0.34	0.18	0.21	0.79	66
Mackenzie Shelf						
G1	0.58	0.16	0.08	0.47	0.53	94
G7	0.81	0.21	0.11	0.44	0.56	82
GRM1	0.76	0.18	0.10	0.49	0.51	89
G12	2.12	0.30	0.04	0.54	0.46	72
G9	1.21	0.32	0.06	0.55	0.45	76
G10	0.93	0.26	0.09	0.65	0.35	88
Canadian Archipelago						
QM1	1.49	n.a.[b]	0.03	0.45	0.55	30
VS1	n.a.	0.06	0.01	0.51	0.49	0
FS1	n.a.	0.08	0.03	0.52	0.48	0
BE2	n.a.	0.02	0.33	0.70	0.30	30
PS1	n.a.	0.03	0.12	0.84	0.16	11
PS2	n.a.	0.08	0.10	0.21	0.79	8
Lancaster Sound						
CAA2	n.a.	0.05	0.04	0.43	0.57	9
CAA1	0.89	0.20	0.08	0.78	0.22	53
BB11	0.43	0.44	0.17	0.75	0.25	46
Davis Strait						
DS5	n.a.	0.01	0.19	0.34	0.66	39
DS2	n.a.	0.11	0.03	0.45	0.55	10
DS1	n.a.	0.07	0.18	0.07	0.93	12

[a]Compound codes: VP, vanillyl phenols; SP, syringyl phenols; CP, cinnamyl phenols; LP, lignin phenols (VP + SP + CP); CA, cutin acids; w-OH, terminally hydroxylated cutin acids w,x-OH, internally hydroxylated cutin acids; %VPI, percent vascular plant input estimated from PCA model (see text for details).

[b]n.a., not applicable due to compound yields at/below detection limits.

mineral soils are characterized by lower biomarker yields that reflect diagenetic processes (e.g., microbial decay, sorption/desorption) during pedogenesis, whereas coarser, less-degraded, vascular plant detritus is typically characterized by higher biomarker yields [e.g., *Hatten et al*., 2012; *Goñi et al*., 2013]. Differential transport of these two broadly defined terrigenous particle classes can result in distinct spatial biomarker distributions [e.g., *Bianchi et al*., 2007; *Goñi et al*., 2008; *Kuzyk et al*., 2008; *Tesi et al*., 2010]. Hence, it is possible that the patterns revealed by our analyses could in part reflect differential transport processes of terrestrial materials between the central/eastern and western regions of the study area.

[43] In Arctic margins, transport processes include atmospheric (dust) transport [*Yunker et al*., 2005, 2011], entrainment and transport by ice [e.g., *Eicken*, 2004; *Eicken et al*., 2005; *Dethleff*, 2005], water column transport involving resuspension on the shelf and margins followed by advection to the interior ocean [e.g., *O'Brien et al*., 2006, 2011], transport by eddies [*O'Brien et al*., 2011, 2013], and density flows off the shelves, likely enhanced at canyons [e.g., *Pickart et al*., 2005]. Each of these processes has the potential to alter the compositional signatures of the OM_{terr} delivered to different regions of the margin and basin. Hence, the remarkably high contributions of biomarker-rich OM_{terr} in distal locations along Barrow Canyon may be explained by the focusing of terrigenous materials at the head of the Canyon and their efficient down-canyon transport to the adjacent basin during downwelling-favorable conditions and dense water cascades [e.g., *Pickart et al*., 2005]. These processes, which have been observed in other canyon systems [e.g., *Tesi et al*., 2008; *Pasqual et al*., 2011], can lead to the rapid transfer of OM to deep depositional centers where enhanced accumulation minimizes microbial degradation and can result in efficient burial. Our results, together with those of *Macdonald and Gobeil* [2012], suggest that canyon-facilitated transport of OM may be more significant in offshore OM transport along the Arctic Ocean margin than previously recognized.

5. Closing Remarks

[44] The results of this study show marked regional contrasts in the distribution of organic and inorganic carbon in surface sediments from the North American Arctic margin. These differences reflect variable inputs, transport pathways, and distinct sedimentary OM preservation regimes associated with oxidative conditions in the seabed. There are large differences in the distribution of land-derived materials, including bedrock carbonates and terrigenous organic matter, which are related to the surficial geology and geomorphology of the adjacent land masses, rates of coastal erosion and transport processes responsible for the redistribution of allochthonous materials. In view of these strong regional contrasts, it is not possible to give a uniform prediction for how climate-induced changes such as decreased sea ice cover, permafrost degradation, and increased fluvial runoff may impact benthic environments across the whole Arctic margin. Any prediction should be regionally grounded and consider the distinct compositions like those identified in this study before extrapolating from one area to another.

[45] With these caveats in mind, we suggest that the enhanced ice-free conditions now observed in the Arctic will have the largest impact in areas such as the Canadian Archipelago, Lancaster Sound, and Davis Strait. These regions will experience considerable increases in the influx of both marine and terrestrial organic matter as a result of higher productivity, enhanced coastal erosion, and greater sediment resuspension. Areas such as the Beaufort slope and shelf will likely also experience elevated inputs of terrestrial materials as result of these processes favored by extended periods of ice-free conditions, which will likely result in more efficient cross-margin transport to deeper parts of the basin through bathymetric features such as canyons. Changes in the influx of inorganic sediment and in the magnitude and composition of organic matter reaching the seafloor will directly affect benthic communities in these regions of the Arctic margin.

[46] The high abundance of bedrock carbonate in sediments from the eastern region of the study area presents an intriguing scenario in terms of the impacts of ocean acidification on the benthic fauna. Recent studies have shown that surface waters in parts of the Bering Sea and western Arctic Ocean are already undersaturated with respect to aragonite [e.g., *Fabry et al*., 2009; *Yamamoto-Kawai et al*., 2011], and models project that aragonite undersaturation will be prevalent in even larger areas of the Arctic Ocean and parts of the subarctic Pacific by as early as 2023 [e.g., *Steinacher et al*., 2009; *Feely et al*., 2009]. Dolomitic carbonate in surface sediments from the eastern region of the North American Arctic margin could provide a buffer against elevated CO_2 concentrations in pore waters and thus limit the effects of acidification on benthic fauna. In contrast, regions like the Bering and Chukchi shelves contain little carbonate in surface sediments and appear to be characterized by high benthic remineralization rates. Based on these characteristics, benthic fauna at these locations may be far more vulnerable to impacts of ocean acidification.

[47] **Acknowledgments.** We gratefully acknowledge the Canadian International Polar Year Program and the Natural Sciences and Engineering Research Council of Canada for their support in the collection of the 2007–2008 core and analyses of the samples including the results reported here. We greatly appreciate the efforts of the officers and crews of the CCGS Wilfrid Laurier (2007) and Louis St. Laurent (2008) who got us to our stations and helped retrieve the sediment cores. The paper benefited from insightful comments by Jochen Knies and an anonymous reviewer. Finally, we appreciate technical assistance in sampling and sample handling by D. Dubien, M.-E. Randlett, Y. Alleau, and B. Klein. Funding for Alison O'Connor contribution was through the U.S. National Science Foundation REU program at Oregon State University. The NSERC CERC program provided support for Zou Zou Kuzyk. Miguel Goñi's contribution to the work also was supported through grant 0902850 from the U.S. National Science Foundation's P2C2 program.

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