Characterization of Siberian Arctic coastal sediments: Implications for terrestrial organic carbon export

Laodong Guo,[1] Igor Semiletov,[1,2] Örjan Gustafsson,[3] Johan Ingri,[4] Per Andersson,[5] Oleg Dudarev,[2] and Daniel White[6]

Received 29 April 2003; revised 14 December 2003; accepted 7 January 2004; published 10 March 2004.

[1] Surface sediments were collected during the 2000 TransArctic Expedition along the Siberian Arctic coastline, including the Ob, Yenisey, Khatanga, Lena, and Indigirka estuaries. Sediments were characterized for elemental composition (total organic carbon, TOC, black carbon, BC, and total N, as well as major and trace elements), isotopic signature ($\delta^{13}C$, $\delta^{15}N$, $\Delta^{14}C$, ε_{Nd}, $^{87}Sr/^{86}Sr$), and organic molecular composition to better understand river export variations over the large spatial scale of the Siberian Arctic. On average, 79 ± 9% of the total C in sediments was organic while 21 ± 9% was inorganic. BC made up 9 ± 4% of the TOC pool, with a general increasing trend from west to east along the Siberian coast. The combined Nd- and Sr-isotopes (ε_{Nd} and $^{87}Sr/^{86}Sr$) were used to define two distinct sediment sources between east and west Siberian regions with the Khatanga River as a boundary. Data from pyrolysis-GC/MS of the sedimentary organic carbon (SOC) indicated an increase in the freshness of the organic matter from west to east on the Siberian Arctic coast, with increasing relative abundance of furfurals (polysaccharides) with respect to nitriles. Values for the $\delta^{13}C$ of SOC ranged from −27.1‰ (mostly terrigenous) to −23.8‰, while $\delta^{15}N$ increased from east to west (3.1 to 5.2‰) with a significant correlation with C/N ratio. Values for the $\Delta^{14}C$ of SOC ranged from −805 to −279‰, with a consistent trend increasing from the east (Indigirka River) to the west (Ob River). These $\Delta^{14}C$ values corresponded to a ^{14}C age of 2570 ± 30 yBP in the Ob estuary and 13,050 ± 50 yBP in the Indigirka estuary. Most importantly, $\Delta^{14}C$ values were significantly correlated with the ratio of BC/TOC ($R^2 = 0.91$, n = 6), consistent with the distribution pattern of increasing permafrost zone from the west to the east along the Siberian coast. Together, our results suggest that older OC was derived from the release of recalcitrant BC during permafrost thawing and riverbank and coastal erosion, likely enhanced by ongoing environmental changes in the northern ecosystem. *INDEX TERMS:* 1040 Geochemistry: Isotopic composition/chemistry; 1055 Geochemistry: Organic geochemistry; 1615 Global Change: Biogeochemical processes (4805); 4806 Oceanography: Biological and Chemical: Carbon cycling; *KEYWORDS:* Arctic Ocean, organic carbon, sediment

Citation: Guo, L., I. Semiletov, Ö. Gustafsson, J. Ingri, P. Andersson, O. Dudarev, and D. White (2004), Characterization of Siberian Arctic coastal sediments: Implications for terrestrial organic carbon export, *Global Biogeochem. Cycles*, *18*, GB1036, doi:10.1029/2003GB002087.

[1]International Arctic Research Center, University of Alaska Fairbanks, Fairbanks, Alaska, USA.
[2]Pacific Oceanological Institute, Russian Academy of Science, Vladivostok, Russia.
[3]Institute of Applied Environmental Research (ITM), Stockholm University, Stockholm, Sweden.
[4]Division of Applied Geology, Lulea University of Technology, Lulea, Sweden.
[5]Laboratory for Isotope Geology, Swedish Museum of Natural History, Stockholm, Sweden.
[6]Water and Environmental Research Center, University of Alaska Fairbanks, Fairbanks, Alaska, USA.

0886-6236/04/2003GB002087$12.00

1. Introduction

[2] Arctic rivers are a globally important source of terrestrial organic carbon to the ocean. They make up about 10% of the global river water runoff and contain high concentrations of dissolved and particulate organic carbon [e.g., *Lobbes et al.*, 2000; *Holmes et al.*, 2002; *Guéguen et al.*, 2003]. Therefore, terrestrial inputs of organic carbon to the continental margin waters of the Arctic Ocean are high [*Anderson et al.*, 1998; *Kattner et al.*, 1999; *Stein and MacDonald*, 2003].

[3] The terrestrial Arctic has historically been an overall significant sink for carbon. As such, large quantities of soil organic carbon are stored in the region. It is estimated that northern ecosystems have accumulated 25–33% of the

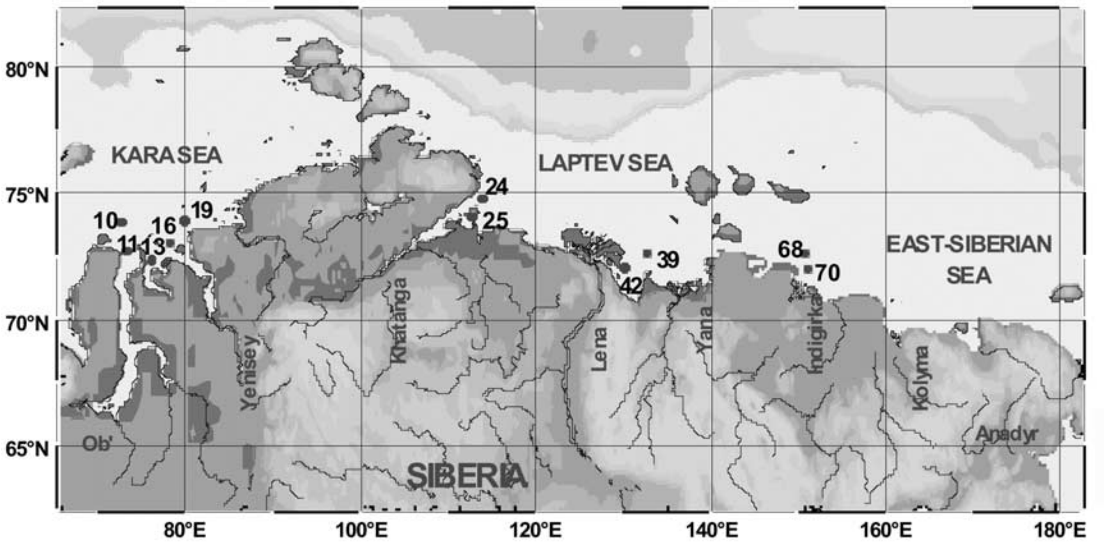

Figure 1. Map of the Siberian Arctic region and sampling locations. See color version of this figure at back of this issue.

world's soil carbon [*Waelbroeck et al.*, 1997; *Oechel et al.*, 2000]. Recent environmental changes, however, may have a profound impact on this ecosystem and result in a major C export to the Arctic Ocean. For example, there is evidence for increasing river discharge and changes to the hydrological regime in the Siberian region especially after 1970 [e.g., *Savelieva et al.*, 2000; *Semiletov et al.*, 2000; *Peterson et al.*, 2002; *Yang et al.*, 2002]. In addition, permafrost and ice complex shrinking and increased coastal erosion in the northern regions have been widely reported [e.g., *Semiletov*, 1999, 2000; *Jorgenson et al.*, 2001; *Brown et al.*, 2003]. Permafrost contains a vast amount of old organic matter that could actively participate in biogeochemical cycles if liberated by thermokarst, coastal erosion, and an increase in the seasonal thaw depth of permafrost. It is suggested that carbon currently sequestered in permafrost is likely to be exposed to physicochemical and biological degradation in a warming climate, potentially creating a positive feedback to climate change [e.g., *Davidson et al.*, 2000]. A hypothesized climate-change-driven increase in terrestrial organic carbon inputs to the Arctic Ocean through permafrost thawing, increasing river runoff, and accelerated coastal erosion could dramatically alter carbon budgets and biogeochemical cycles [e.g., *Gibson et al.*, 2000; *Savelieva et al.*, 2000; *Freeman et al.*, 2001; *Stein and MacDonald*, 2003]. While the fate and transport processes of terrestrial organic carbon across the Arctic land/ocean margin are largely unknown, they are critical to our understanding of environmental change on a timescale of human concern. Whether there is a sign of old terrestrial organic carbon export due to recent environmental and climate changes in the Arctic region is still not clear. The quantitative importance of pre-aged terrestrial organic carbon export to the Arctic Ocean owing to warmer conditions needs to be better understood especially on a pan Arctic perspective.

[4] During the 2000 TransArctic expedition, sediment samples were collected from the Siberian Arctic coastline, including the Ob, Yenisey, Khatanga, Lena, and Indigirka estuaries. The sediments were characterized for elemental composition (including organic carbon species and major and trace elements) and isotopic signatures (δ^{13}C, δ^{15}N, Δ^{14}C, ε_{Nd}, ^{87}Sr/^{86}Sr). Preliminary results were used to examine river export of terrestrial organic matter in a changing climate over the large spatial scale of the Siberian Arctic.

2. Experimental

2.1. Study Area and Sampling

[5] Surface sediments were collected using a van Veen grab sampler during the Russian 2000 TransArctic expedition along the Siberian Arctic coastline (Figure 1). The five major Siberian estuarine regions (i.e., Ob, Yenisey, Khatanga, Lena, and Indigirka) that we sampled have different basin areas, hydrology, annual discharges, water and soil chemistry, and permafrost distribution (Table 1) [*Huh et al.*, 1998; *Holmes et al.*, 2002]. Similar surface sediment samples from the Kalix River draining the northern boreal zone of the Scandinavia were also collected to extend the spatial and climatological coverage westward. Sediment samples were kept frozen until processing in the laboratory. Bulk sediments from the top 2 cm (0–2) were dried at 80°C and then homogenized. The specific sedimentation rates for these samples were not measured, but sediments from 0–2 cm should represent the most recent deposited sediments in the study areas. Detailed sampling locations and other parameters are listed in Table 1.

[6] The watersheds of east Siberia are situated mainly in the permafrost area that occupies about one half of the territory of Russia. On the other hand, the northern European and west Siberian rivers are mainly located in the region of non-permafrost and/or permafrost islands (Figure 2). As shown in Figure 2, the Khatanga, Lena, Indigirka, and Kolyma are rivers in the permafrost region,

Table 1. Sampling Locations, Hydrological Data, and Ancillary Parameters in Siberian Arctic and Kalix Rivers

Sample ID	River/Estuary	Location	Surface Salinity, ‰	Sediment Type	Basin Area,[a,b,c] 10^6 km^2	Water Runoff,[a,b,c] km^3/yr	Suspended Sediment,[a,b,c] 10^6 t/yr	Dissolved Matter,[a,b,c] 10^6 t/yr	Dissolved/Suspended Ratio	TOC,[a,b,c] 10^6 t/yr	Fe,[a] 10^6 t/yr	Mn,[a] 10^6 t/yr
KR-R	Kalix	66°02′07N 22°49′08E	0	silt	2.36×10^{-2}	10	20.4×10^{-3}	32×10^{-2}	16	4.32×10^{-2}	75×10^{-4}	1.94×10^{-5}
KR-E	Kalix	65°45′75N 23°18′90E	2	silt								
N-10	Ob	73°50′33N; 72°42′4E	12.1	silty mud	2.54–2.99	429–433	13.4–16.5	34–47.2	2.9–3.5	3.05–3.69	397	3.95
N-11	Ob	72°40′20N; 73°22′40E	7.5	silty mud								
N-13	Yenisey	72°20′N; 76°12′8E	8.3	silty mud	2.44–2.59	555–620	13.2–14.5	43–59.9	4.1–4.5	4.13–4.59	67	3.33
N-16	Yenisey	73°00′17N; 78°22′23E	8.6	Sandy mud								
N-19	Yenisey	73°51′21N; 80°00′36E	20.9	silty mud								
N-24	Khatanga	74°45′N; 113°59′93E	17.8	silty mud	0.275–0.364	70–85.3	1.7	7.93	4.7	0.54[c]	-	-
N-25	Khatanga	74°01′92N; 112°42′84E	13.2	mictite								
N-39	Lena	72°33′84N; 132°48′70E	12.9	mud	2.46–2.48	505–525	11.7–17.6	41–59.6	3.4–4.8	5.06–5.30	22	3.53
N-42	Lena	72°00′71N; 130°14′39E	3.8	silty mud								
N-68	Indigirka	72°34′1N; 150°56′8E	21.3	silt	0.358–0.362	47.5–61	12.9	3.1–3.57	0.3	0.47[c]	4	0.67
N-70	Indigirka	71°59′13N; 151°13′67E	21.9	muddy silt								

[a]Data from *Telang et al.* [1991].
[b]Data from *Holmes et al.* [2002].
[c]Data from *Gordeev et al.* [1996].

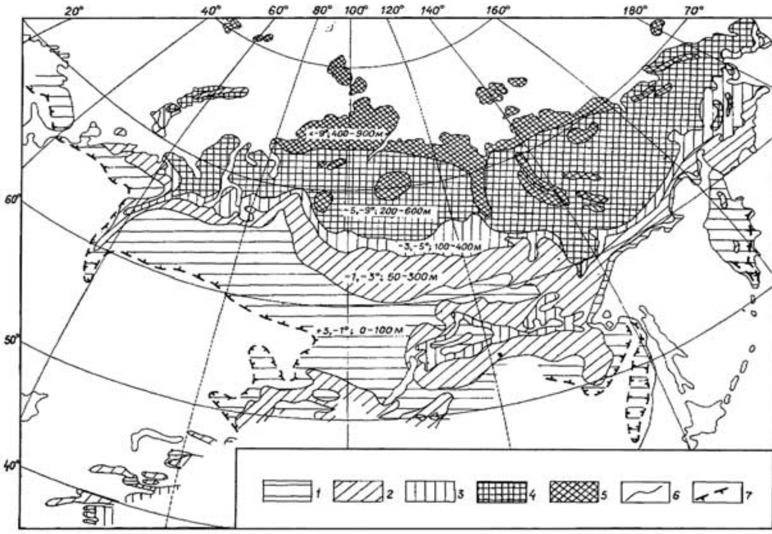

Figure 2. Distribution of permafrost zone increasing from west to east coast. Numbers given in key: 1: the zone of island permafrost with the mean temperature (T) from +3°C to −1°C, and thickness (H) from 0 to 100 m; 2–5: the zone of continuous permafrost with the different mean temperature and thickness: 2: T = from −1°C to −3°C, H = 50–300 m; 3: T = from −3°C to −5°C; 4: T = from −5°C to −9°C; 5: T < −9°C; 6: boundary between the different permafrost zones; and 7: the southern boundary of island permafrost distribution (boundary of the permafrost and permafrost islands regions are adopted from *Danilov* [1990]).

whereas the upper and mid-stream of the Yenisey is flowing in the region of permafrost islands, and the upper and mid-stream of the Ob is flowing in non-permafrost region with only a small part of the low-stream in the continuous permafrost region. Thus discharge (surface runoff)/precipitation (D/P) ratios are higher in the watersheds underlain by a shallow permafrost table in east Siberia compared with the watersheds with a deeper permafrost table in west Siberia. Indeed, D/P ratio increases from 0.30 in the west to 0.74 in the east [*Semiletov et al.*, 2000], whereas the evaporation/precipitation ratio (E/P) decreases from 0.70 to 0.26.

[7] As summarized in Table 1, the Eurasian Arctic rivers have very low sediment yields (tons/km^2/yr), compared with other major rivers. In addition, the ratios of dissolved/suspended matter in the Eurasian Arctic rivers are high. This indicates the dominance of chemical weathering and low transport rates for suspended particles in the low relief river basins of the Eurasian Arctic. There is also a trend with the dissolved/suspended matter ratio increasing from the Ob to the Lena River (Table 1).

2.2. Measurements of Carbon and Nitrogen

[8] Sediment samples were measured for total organic carbon (TOC), inorganic carbon (IC), and pyrogenic black carbon (BC) contents as well as for the total nitrogen (TN) concentrations. Total C and N were measured on a Carlo Erba elemental analyzer [*Guo et al.*, 2003a]. Organic C was measured after samples were treated with HCl [*Santschi et al.*, 2001]. Inorganic C was then calculated from the difference between TC and OC concentrations (i.e., IC = TC − OC).

[9] Black carbon (BC) content was determined with the chemothermal oxidation method [*Gustafsson et al.*, 1997, 2001]. Briefly, the sediment was first finely ground for 20 min in an automatic ball grinder. Then, IC was removed by a mild acidification in situ in Ag capsules. Amorphous non-pyrogenic OC was subsequently removed in a thoroughly tested thermal oxidation procedure at 375°C in a tube furnace continuously supplied with an atmosphere of air. This CTO375 method of BC determination in surface sediments has proven to return results that are isotopically and geochemically consistent and able to provide improved accountability of the environmental distribution of co-produced molecular combustion markers [e.g., *Gustafsson et al.*, 2001; *Middelburg et al.*, 1999; *Persson et al.*, 2002; *Currie et al.*, 2002].

2.3. Major and Trace Element Analysis Using ICP-AES, ICP-MS, and TIMS

[10] For major element determinations, the sediment was digested following the procedure described by *Burman et al.* [1978]. Sediment samples were fused with $LiBO_2$ at 1000°C, and the beads thus formed were dissolved in 5% suprapur HNO_3. The major elements were then measured by inductively coupled plasma-atomic emission spectrometry (ICP-AES).

[11] Trace elements were dissolved by a digestion procedure using 50% suprapur HNO_3 in Teflon bombs in a microwave oven. The solutions were then centrifuged, diluted, and finally analyzed by ICP-AES and inductively coupled plasma-mass spectrometry (ICP-MS). The accuracy and precision of the analyses were checked by analyzing reference sediments. The instrumental precision, determined as ±1 SD, for three to four runs on the same sample was generally better than 1% for the major elements and 10% for the trace elements. Isotopic ratios of Nd and Sr were

Table 2. Elemental and Isotopic Characteristics of Carbon and Nitrogen in Surface Sediments of Eurasian Arctic River/Estuaries[a]

River/Estuary	Sample ID	OC, mg-C/g	IC, mg-C/g	BC, mg-C/g	N, mg-N/g	C/N	BC/OC	$\delta^{13}C$, ‰	^{14}C age,[b] yBP	f_{modern}[c]	$\delta^{14}C$, ‰	$\delta^{15}N$, ‰
Kalix	KR-R	41.2 ± 0.3	n.d.	0.39 ± 0.03	3.5 ± 0.1	14.9	0.0094	−27.63 ± 0.05	630 ± 25	0.9246 ± 0.0031	−81 ± 3	1.49 ± 0.17
Kalix	KR-E	44.0 ± 0.6	n.d.	0.75 ± 0.23	3.4 ± 0.1	16.3	0.017	−26.72 ± 0.07	600 ± 35	0.9279 ± 0.0039	−78 ± 5	0.75 ± 0.51
Ob	N-10	5.0 ± 0.6	1.4	0.19 ± 0.12	0.6 ± 0.1	9.7	0.039	−25.70 ± 0.09	2570 ± 30	0.7258 ± 0.0026	−279 ± 3	5.12 ± 0.40
Ob	N-11	5.6 ± 0.1	1.4	0.57 ± 0.03	0.6 ± 0.02	11.1	0.102	−27.14 ± 0.10				5.21 ± 0.42
Yenisey	N-13	12.8 ± 0.7	8.5	1.29 ± 0.48	1.6 ± 0.1	9.5	0.101	−26.38 ± 0.11				5.00 ± 0.13
Yenisey	N-16	3.6 ± 0.1	0.6	0.15 ± 0.08	0.4 ± 0.0	11.6	0.041	−26.15 ± 0.13	3600 ± 30	0.6390 ± 0.0036	−365 ± 5	4.04 ± 0.56
Yenisey	N-19	9.3 ± 0.5	4.5	0.52 ± 0.12	1.1 ± 0.1	9.9	0.056	−26.87 ± 0.59				5.16 ± 0.55
Khatanga	N-24	10.7 ± 0.7	2.8	0.68 ± 0.10	1.2 ± 0.0	10.9	0.064	−23.83 ± 0.01	5950 ± 30	0.4766 ± 0.0017	−526 ± 3	4.62 ± 0.40
Khatanga	N-25	9.6 ± 0.9	4.7	1.12 ± 1.17	1.2 ± 0.0	9.1	0.117	−24.53 ± 0.05				4.97 ± 0.36
Lena	N-39	13.6 ± 1.9	3.3	1.41 ± 0.01	1.4 ± 0.2	11.6	0.104	−25.30 ± 0.25	6400 ± 30	0.4510 ± 0.0016	−552 ± 3	3.87 ± 0.08
Lena	N-42	15.0 ± 1.6	2.2	2.53 ± 0.26	1.4 ± 0.2	12.6	0.168	−25.63 ± 0.27				3.10 ± 0.25
Indigirka	N-68	5.8 ± 0.2	1.1	0.95 ± 0.26	0.6 ± 0.04	10.6	0.163	−25.92 ± 0.09	13050 ± 50	0.1968 ± 0.0013	−805 ± 3	3.80 ± 0.27
Indigirka	N-70	7.8 ± 0.4	0.5	1.14 ± 0.20	0.8 ± 0.04	11.1	0.146	−26.35 ± 0.18				3.99 ± 0.04

[a]Errors are those of $\pm 1\sigma$.
[b]Here yBP is years before the present.
[c]Here f_{modern} is fraction of modern.

measured by thermal ionization mass spectrometer (TIMS) [*Andersson et al.*, 2000].

2.4. Organic Molecular Characterization by Pyrolysis-GC/MS

[12] Organic matter in sediment samples was fingerprinted using pyrolysis-gas chromatography/mass spectrometry (py-GC/MS) according to methods given by *White and Beyer* [1999]. Approximately 100 of the most abundant compounds in each pyrogram were identified using a mass spectral library. An index set of 21 compounds, divided into four categories, was selected to show key differences between samples. The ion chromatogram of the most abundant ion in each of the 21 index compounds was extracted from the total ion chromatogram. The area assigned to each index compound was equal to the area under the trace of the most abundant ion only. This facilitated separation of overlapping peaks in the total ion chromatogram. A "fraction of index" for each category was calculated by summing the areas of all compounds in the category and dividing by the sum of areas of all index compounds for the sample.

[13] The index categories were furfurals, alkylbenzenes, nitriles, and other. The category of "other" represented all groups of compounds that did not compose more than 10% of any one sample. The analysis was not intended to represent all compounds in the organic carbon pool, but rather use the available information to show differences between the sediment samples. Pyrolysis-GC/MS conditions were maintained according to *White et al.* [2002].

2.5. Measurements of $\delta^{13}C$, $\delta^{15}N$, and $\Delta^{14}C$

[14] Sediment samples were also measured for organic C and N isotopic composition, including $\delta^{13}C$, $\delta^{15}N$, and $\Delta^{14}C$. Before measurements, inorganic carbon was removed from sediment samples by HCl treatment. Stable carbon and nitrogen isotope ratios were calculated in terms of $\delta^{13}C$ and $\delta^{15}N$, $(R_{sample}/R_{standard} - 1) \times 1000$, where R is the ratio of $^{13}C/^{12}C$, or $^{15}N/^{14}N$, in sediment samples or standards (PDB for carbon and atmospheric N_2 for nitrogen). Both $\delta^{13}C$ and $\delta^{15}N$ were measured by continuous flow isotope ratio mass spectrometry [*Guo et al.*, 2003b]. The precision and accuracy of $\delta^{13}C$ and $\delta^{15}N$ analysis were ±0.1‰, and ±0.2‰, respectively, as determined by replicate analysis of standards and samples. Values of $\Delta^{14}C$ were measured using accelerator mass spectrometry (AMS) at the National Ocean Science AMS Facility at Woods Hole Oceanographic Institution [*Guo et al.*, 1996]. One-sigma errors are given in fraction of modern and $\Delta^{14}C$ values and ^{14}C ages.

3. Results and Discussion

3.1. Variations of Organic and Inorganic Carbon Species

[15] Measurements of organic and inorganic C concentrations (Table 2) showed that TOC varied from 3.6 to 15 mg/g dried sediment (mg/gds), with higher OC concentrations in the Kalix River but no obvious trend along the Arctic coastline or among Siberian rivers. Concentrations of IC were considerably lower than those of OC, ranging from 0.5 to 8.5 mg/gds (Table 2), with an average of 2.8 ±

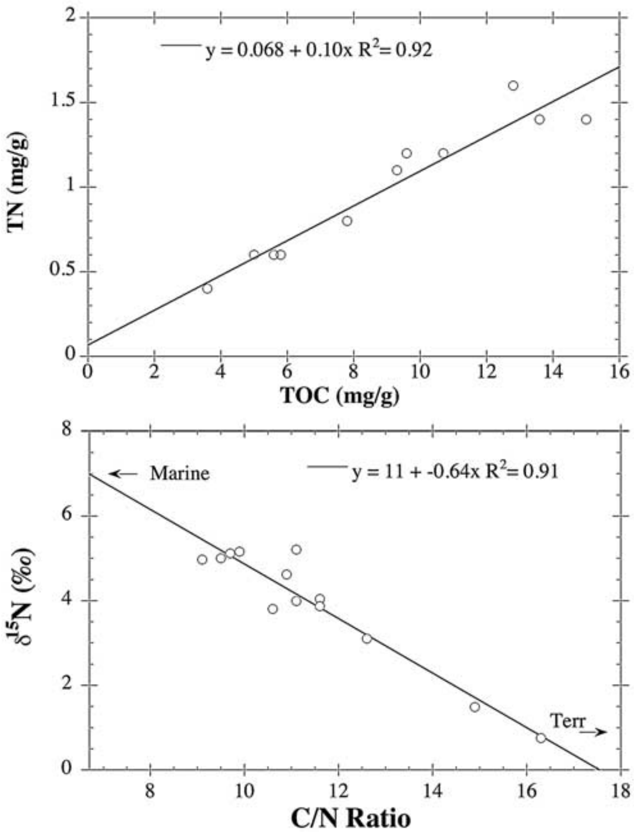

Figure 3. Correlations (top) between total organic carbon (TOC) and total nitrogen (TN) and (bottom) between $\delta^{15}N$ and C/N ratio, in surface sediments from the Siberian Arctic coast. Data of Kalix River are not plotted in the TOC/TN relationship. The intercept of 0.07 (mg/g) represents the inorganic N content at zero TOC (top panel) while $\delta^{15}N$ values for interpolated marine and terrestrial end-members were 7 and ~1, respectively.

2.3 mg/gds. Most of the C in sediments was OC, comprising 60–94% of the total C (with an average of 79 ± 9%). While both OC and IC concentrations differed from station to station, their percentage in the total C pool seemed relatively constant, i.e., 79 ± 9% for OC/TC and 21 ± 10% for IC/TC among the five Siberian estuaries.

[16] Concentrations of BC varied from 0.15 to 2.53 mg/gds with an average of 0.9 ± 0.6 mg/gds and a median of 0.75 mg/gds (Table 2). Within the TOC pool, BC comprised 1–17% of the OC, with an average of 9 ± 4% (median 10%). This fractional contribution to the bulk OC was in the same broad range as has recently been reported for other contemporary ocean margin sediments such as the Gulf of Maine [*Gustafsson and Gschwend*, 1998], the North Sea [*Middelburg et al.*, 1999], the Mississippi River plume [*Mitra et al.*, 2002], the Santa Monica Basin [*Masiello et al.*, 2002], and a Norwegian fjord [*Persson et al.*, 2002]. There did not appear to be any consistent trend in absolute BC concentrations among rivers/estuaries in the Eurasian Arctic. However, there appeared to be an increasing trend in the percentage of BC in the TOC pool going from the western Arctic rivers, such as the Kalix, Ob, and Yenisey via Khatanga to the east Siberian Lena and Indigirka rivers (Table 2).

[17] While BC in marine sediments from the temperate and industrialized regions can be derived from anthropogenic sources such as fossil fuel combustion, it is likely that BC now found in these remote Arctic rim sediments originated from vegetation- or wild-fires in their respective drainage basins. Such biomass-derived BC is believed to represent 90% of the BC released to the environment [*Kuhlbusch and Crutzen*, 1995]. The BC now found in surficial river/estuarine sediments should provide an integrated picture of the burning history and soil/permafrost dynamics of the corresponding river drainage basin. Therefore the consistent increase in the BC/TOC ratio from west to east along the Siberian coastline points to a dynamically varying source function of BC from combined fire, drainage, and thawing permafrost processes. Peat and forest fires are common in the permafrost region of Russia and Siberia and have been reported to affect an average area as large as 1.2 million ha/yr for the recent period 1990–1999 [*Shivdenko and Goldammer*, 2001]. In particular, the Siberian Far East region differs from other parts by frequent forest fires due to the specific climatic and forest vegetation characteristics [*Davidenko*, 2000; *Shivdenko and Goldammer*, 2001]. Moreover, *Zimov et al.* [1993] reported that fires covering more than 0.3 million ha of the Kolyma lowland between early 1960s and late 1980s caused a significant increase in the active layer depth from 0.3–0.6 to 2.0 m. These independent reports of geographical forest fire patterns are consistent with our findings of a west-to-east increase in BC/TOC of river mouth surface sediments (Table 2).

[18] Concentrations of total nitrogen (TN) in the sediment samples ranged from 0.4 to 1.6 mg/g, with an average of 0.98 ± 0.40 mg/g. As shown in Figure 3, concentration of TN was significantly correlated with TOC, with an intercept value of 0.068, similar to those reported in Arctic sediments [*Stein and MacDonald*, 2003]. On the basis of these TOC and TN concentrations, a C/N ratio was calculated. As shown in Table 2, the C/N molar ratio averaged 10.7 ± 1.0, ranging from 9 to 12.6. Higher C/N ratios in the northern region were consistent with their terrestrial nature, with lighter $\delta^{13}C$ values and older apparent ^{14}C ages (Table 2).

3.2. Fingerprinting of Organic Molecules

[19] The py-GC/MS technique was used to provide information about the molecular composition of the SOC. Results for the bulk SOC samples were summarized in Figure 4. In terms of total index, furfurals, alkybenzenes, and nitriles were the major categories in the SOC pool. Furfurals, a representative of polysaccharides, ranged between ~32% of the index in Ob sediments to 42% in the Indigirka sediments. Alkybenzenes varied from 16% of the index in the Ob to 26% in the Khatanga. Nitriles ranged from 23% of the index in the Indigirka sediments to 36% in the Ob, with an inverse distribution trend compared to that of furfurals. Other minor organic components made up an average of 14% of the total index (Figure 4).

[20] In general, nitriles are derived from polypeptides and indicate a well-humified environment. Alkylbenezenes are most abundant in soils formed under anaerobic conditions

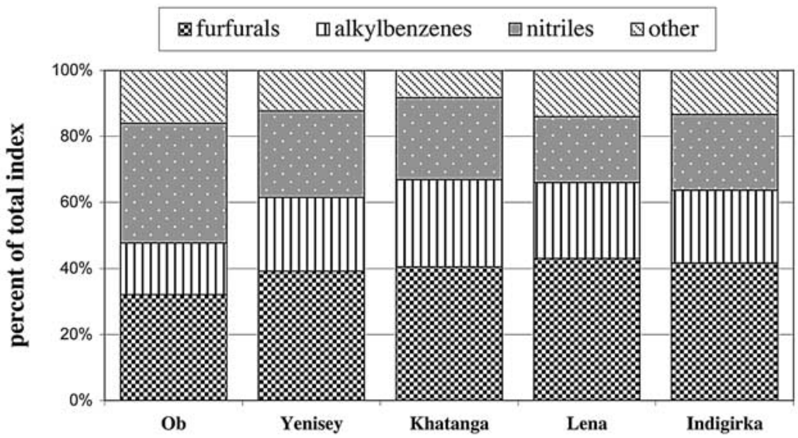

Figure 4. Relative abundances (percent of index) of organic molecules in sediments from the west to the east coast, as identified by pyrolysis-GC/MS using four major compound classes.

where O- and N-depletion has occurred. Furfurals, derived from polysaccharides, represent fresh organic matter that has undergone little decomposition [*Bracewell et al.*, 1989].

[21] The comparison plots in Figure 4 show that the relative abundance of different types of materials varied from sample to sample. The most noticeable difference was the general decrease in the relative abundance of nitriles and the increase in relative abundance of furfurals from west to east. This condition suggested that the SOC in the eastern river basins was less decomposed. This finding was in agreement with the observation that the watersheds in the east have a greater abundance of permafrost that tends to preserve organic matter in a fresh state. These results were therefore consistent with data on BC/TOC ratios and radiocarbon analyses, and variation in the permafrost distribution along the Siberian Arctic coastline.

3.3. Major and Trace Elements, Including Sr and Nd Isotopes

[22] Among the major element oxides, SiO_2 and Al_2O_3 were the most abundant, ranging from 55 to 77% and 8.8 to 16.1%, respectively, followed by Fe_2O_3, Na_2O, K_2O, CaO, MgO, TiO_2, P_2O_5, and MnO_2 (Table 3). The sum of oxides is shown in Table 3, along with concentrations of trace elements, including As, Co, Cr, Cu, Ni, Pb, Zn, and Zr (in mg/kg for trace elements).

[23] Using element/Al concentration ratio as an index, the Ti/Al ratio in the Lena river mouth sediment was almost identical to the average continental crust ratio and the ratio found in suspended particles in "average river waters" (Table 3). This indicates that both Al and Ti were mainly present in rock forming primary minerals in the sediment. Similarly, the Si/Al ratio was between the continental crust ratio and the ratio in "average river suspended matter." In the Yenisey samples, P appeared to follow the Fe pattern, similar to As (Table 3).

[24] Several of the analyzed trace elements (Co, Cu, Ni, Pb, and Zn) showed a relative enrichment in the formed authigenic Mn-rich phase (Table 3). A decreasing trend of the Fe/Al ratio was seen in the sediments, going from west to east, with a relative enrichment of Fe in the Yenisey samples. Most of the enriched Fe/Al ratios in the sediments more likely represented aggregated and sedimented colloidal Fe from the rivers.

Table 3. Major Elements, Normalized to Al (wt%), and Selected Trace Elements (ppm) in Surface Sediments From the Siberian Arctic Estuaries[a]

Sample ID	Sum of Oxides	Al	Si/Al	Fe/Al	Na/Al	K/Al	Ca/Al	Mg/Al	Ti/Al	P/Al	Mn/Al	As	Co	Cr	Cu	Ni	Pb	Zn	Zr
KR-R	83.3	5.45	4.24	1.68	0.31	0.30	0.30	0.21	0.075	0.033	0.141	7	22	70	31	20	12	93	182
KR-E	85.6	6.03	3.98	1.37	0.33	0.34	0.26	0.23	0.064	0.034	0.059	66	22	98	36	27	18	134	178
OB-10	95.1	5.15	6.63	0.55	0.35	0.39	0.18	0.15	0.073	0.013	0.014	20	13	90	14	22	14	44	259
OB-11	95.9	4.97	6.98	0.61	0.34	0.38	0.17	0.14	0.081	0.018	0.024	21	12	91	12	19	15	42	357
Yenisey-13	90.8	7.57	3.43	0.89	0.23	0.25	0.13	0.23	0.071	0.018	0.097	31	28	122	37	52	20	93	172
Yenisey-16	98.6	4.66	7.71	0.57	0.34	0.33	0.35	0.23	0.091	0.010	0.015	6	9	151	12	20	10	35	335
Yenisey-19	92.8	6.46	4.54	0.79	0.32	0.30	0.19	0.25	0.068	0.015	0.017	24	20	110	30	41	16	79	170
Khatanga-24	92.8	7.68	3.64	0.65	0.29	0.28	0.16	0.23	0.066	0.013	0.008	24	19	106	28	38	18	86	177
Khatanga-25	94.8	6.14	5.22	0.51	0.32	0.35	0.25	0.17	0.057	0.015	0.020	16	15	75	20	25	15	54	265
Lena-39	91.9	8.52	3.19	0.57	0.24	0.31	0.08	0.18	0.052	0.015	0.015	21	17	98	23	34	28	92	184
Lena-42	91.9	8.05	3.49	0.54	0.26	0.30	0.10	0.17	0.054	0.018	0.015	22	13	86	18	29	22	83	201
Indigirka-68	96.4	6.78	4.87	0.39	0.35	0.31	0.12	0.11	0.068	0.013	0.011	12	10	78	12	19	15	55	377
Indigirka-70	94.8	6.99	4.53	0.46	0.32	0.30	0.12	0.12	0.072	0.015	0.010	20	11	94	13	22	18	63	480
Continental Crust			3.98	0.52	0.21	0.35	0.65	0.24	0.05	0.01	0.01								
Average suspended river[b]			3.03	0.51	0.08	0.21	0.23	0.13	0.06	0.01	0.01								

[a] Sum of oxides: $SiO_2 + Al_2O_3 + Fe_2O_3 + Na_2O + K_2O + CaO + MgO + TiO_2 + P_2O_5 + MnO_2$ in percent. Aluminum normalized ratios based on weight percent. Trace metals, As to Zr, in ppm.

[b] Data from *Martin and Whitfield* [1983].

[25] Within each river/estuarine system the sediments yielded a small range in ε_{Nd} ($[(^{143}Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{CHUR}-1] \times 10^4$), generally less than 0.6 ε-units (Figure 5). Given the large distance between the samples taken within each river estuary the sediment ε_{Nd} was relatively homogeneous, which indicates that the ε_{Nd} represent an average signal from each drainage basin. There were significant deviations in ε_{Nd} between the river systems, and two isotopically distinct groups can be identified for the Ob and Yenisey, −7.2, and for the Khatanga, Lena, and Indigirka, −12.0. The difference between the groups most likely reflects the geological variation of northern Siberia, with generally older bedrock exposed to weathering and transport in the eastern part. Thus the ε_{Nd} was a good parameter to separate regional erosional sources of detrital sedimentary particles along the Siberian coast [*Andersson et al.*, 2003].

[26] The $^{87}Sr/^{86}Sr$ data show a large range (0.7115 to 0.7166), but within the ranges reported for suspended particulate and dissolved samples from the Arctic rivers [*Huh et al.*, 1998; *Millot et al.*, 2003]. Within each river/estuarine sample, there was a significant variation in the $^{87}Sr/^{86}Sr$ (Figure 5). However, the general relationship between ε_{Nd} and $^{87}Sr/^{86}Sr$ indicate that both Nd and Sr isotopic composition are varying due to changing provenance [*Andersson et al.*, 2003]. Data from combined isotopes clearly show the differentiation between at least two major sediment sources in the Siberian Arctic, consistent with the distribution pattern of organic composition and carbon isotopic signatures discussed in other sections.

3.4. Stable Isotopes ($\delta^{13}C$, $\delta^{15}N$) and Radiocarbon ($\Delta^{14}C$)

[27] Values of $\delta^{13}C$ varied from −27.6 to −23.8‰ (Table 2). There existed a significant difference in stable C isotopic composition of SOC between estuaries, with lighter $\delta^{13}C$ values in the west and east Siberian Arctic coastal sediments and heavier $\delta^{13}C$ values in the Khatanga coast. For example, samples from the west Siberian coast, such as Ob and Yenisey, had $\delta^{13}C$ values ranging from −27.14 to −25.70‰, while sample from the east Siberian coast, such as Lena and Indigirka, had $\delta^{13}C$ values from −26.35 to −25.30‰. A $\delta^{13}C$ value of −27.70‰ was also reported for sediment samples from the Dmitry Laptev Strait at the east Siberian Sea where coastal erosion and fluvial transport of terrestrial OC dominates [*Semiletov*, 1999; *Naidu et al.*, 2000]. Lighter $\delta^{13}C$ values in the Siberian coastal region are indicative of terrestrial OC either from coastal erosion or riverine inputs [*Semiletov*, 1999; *Rachold et al.*, 2000; *Stein and MacDonald*, 2003], as also suggested by their higher C/N ratios and old ^{14}C ages (Table 2).

[28] Values for $\delta^{15}N$ ranged from 3.1‰ in the coast of the Lena River to 5.2‰ in the Ob River coast. Along the Siberian coastline, values of $\delta^{15}N$ seemed to decrease from the western coast (4.0–5.2‰) to the eastern coast (3.1–3.9‰), whereas $\delta^{13}C$ values show a decrease toward both west and east with the Khatanga River as a transition zone. This distribution pattern agrees well with those of Nd and Sr isotopic composition. Higher $\delta^{15}N$ values in the west Arctic region are likely the results of mixing with more marine or

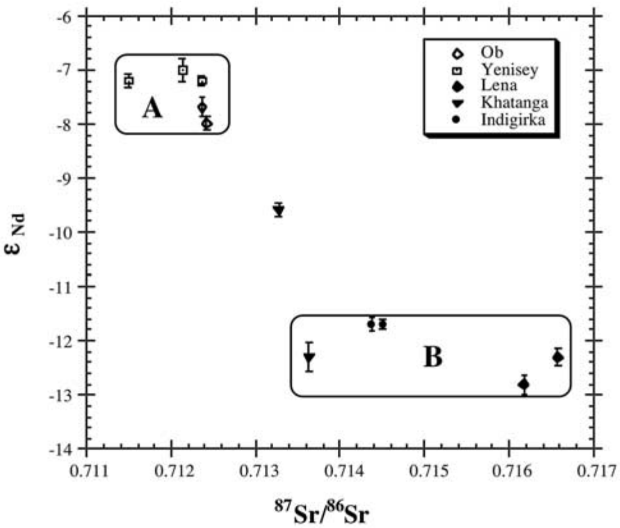

Figure 5. Variations of $\varepsilon_{Nd versus}$ $^{87}Sr/^{86}Sr$ in the Arctic coastal sediments. Group A includes the Ob and Yenisey and group B includes the Indigirka, Lena, and Khatanga. Note that 2σ errors for $^{87}Sr/^{86}Sr$ are smaller than the symbol.

diagenetically altered materials. Interestingly, $\delta^{15}N$ was significantly correlated with C/N ratio, with an interpolated $\delta^{15}N$ value of ∼7 for marine and ∼1 for terrestrial end-member, respectively (Figure 3).

[29] Results of radiocarbon measurements show a more consistent distribution feature. Values of $\Delta^{14}C$ consistently decreased from −279 ± 3‰ in the western Siberian Ob coast to −805 ± 3‰ in the eastern coast of the Indigirka River (Table 2). These $\Delta^{14}C$ values correspond to apparent ^{14}C ages ranging from 2570 ± 30 yBP in the west Siberian coast of the Ob River to 13,050 ± 50 yBP in the east coast of the Indigirka River. This trend extends even farther westward as an even younger radiocarbon signature was found for two samples from Kalix River drainage basin with an average $\Delta^{14}C$ value of −79 ± 5‰ (Table 2). Interestingly, the wideness and thickness of the permafrost zone in the Siberian Arctic region also decrease from the east to the west (Figure 2). Therefore the older SOC in the Indigirka coast and the decrease in ^{14}C ages from the eastern to the western Siberian coast (Figure 6) are likely related to a consistent variation in composition of the terrestrial soil organic matter input from each river basin. More importantly, $\Delta^{14}C$ values were significantly correlated with the ratio of BC/TOC in the sediments (R^2 = 0.91, n = 6) (Figure 6), indicating that the old SOC was mostly derived from recalcitrant black carbon being released from long-term storage in continental soils, especially through permafrost thawing and riverbank and coastal erosions. It follows that the accompanying non-pyrogenic OC has a relatively modern origin (∼124 yBP at zero BC from the relationship between ^{14}C age and BC/TOC ratio in Siberian Arctic samples, figure not shown). Furthermore, the $\Delta^{14}C$ value was also correlated with ε_{Nd} values of the sediments (R^2 = 0.67, n = 6, figure not shown), suggesting that permafrost

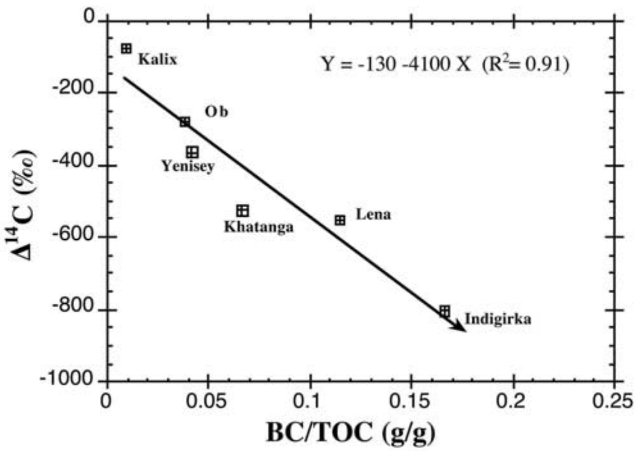

Figure 6. Relationship between $\Delta^{14}C$ values of sedimentary organic matter and the ratio of black carbon (BC) to total organic carbon (TOC) in the Siberian Arctic coast and the Kalix River. Note that the decreasing trend of $\Delta^{14}C$ with increasing BC/TOC ratio is from the west to the east coast.

dynamics and soil/rock chemistry of each river basin may co-regulate the export and biogeochemical cycling of trace elements and terrestrial OC.

[30] The Pleistocene ice-complex is a common feature of the east Siberian and is being damaged by thermokarst processes [*Romanovskii et al.*, 2000]. Rapid coastal erosion (up to 10 m per summer) has been observed mostly in the east Siberian coast and in the Kolyma-Indigirka lowland [*Semiletov*, 1999, 2000]. Therefore the degradation of the coastal and riverbank ice-complex in the Siberian region may play an important role in the offshore transport of old terrestrial OC and thus control the west-east distribution pattern of organic matter freshness and ^{14}C ages as well as hydrochemical anomalies observed in the Siberian coast [*Semiletov*, 1999; *Dudarev et al.*, 2001]. More studies are needed to quantitatively examine the export of terrestrial OC (especially pyrogenic versus non-pyrogenic OC) to the Arctic and the biogeochemical consequences of ongoing climate and environmental change in the northern region.

4. Summary and Conclusions

[31] Along the Siberian Arctic coast, two significantly distinct groups of sediments were defined from the ε_{Nd} and $^{87}Sr/^{86}Sr$ as the westerly group, with the Ob and Yenisey Rivers, and the easterly group with Khatanga, Lena, and Indigirka Rivers. The ratio of black carbon to TOC (average BC/TOC ratio = 0.09 ± 0.04) showed a general trend increasing from the west to the east Siberian coast. Organic matter composition fingerprinted by py-GC/MS revealed a general increase in the relative abundance of furfurals and a decrease in nitriles from west to east, suggesting an increase in the organic matter freshness from west to east. Results of radiocarbon measurements demonstrated a consistent distribution feature, with values of $\Delta^{14}C$ consistently decreasing from −279 ± 3‰ in the Ob to −850 ± 3‰ in the east coast of the Indigirka. These distribution patterns and the extent of preservation of terrestrial organic matter from the west to the east Arctic coast were consistent internally and in agreement with the distribution pattern of permafrost zone in terms of wideness and thickness in the Siberian region.

[32] The significant correlation between $\Delta^{14}C$ values and the ratio of BC/TOC ($R^2 = 0.91$, n = 6) suggest that the old SOC was mostly derived from the release of old carbon from long-term storage in continental soils and ice-complex, likely through permafrost thawing and coastal and riverbank erosion.

[33] **Acknowledgments.** We wish to thank Zofia Kukulska for BC measurements, Sarah Garland for performing pyrolysis-GC/MS, Tim Howe for ^{15}N and ^{13}C analysis, and Lou Codispoti, Robie Macdonald, and two anonymous reviewers for constructive comments on an earlier version of the manuscript. This study was supported, in part, by the Frontier Research System for Global Change/International Arctic Research Center, the Russian Foundation for Basic Research (grants 99-05-65066, 01-05-64018, 02-05-65258), NSF (OPP-0230455), and the Swedish Natural Science Research Council (VR grant G-5103-1165-1999).

References

Anderson, L. G., K. Olsson, and M. Chiersi (1998), A carbon budget for the Arctic Ocean, *Global Biogeochem. Cycles*, *12*, 455–465.

Andersson, P. S., R. Dahlqvist, J. Ingri, and O. Gustafsson (2000), The isotopic composition of Nd in a boreal river: A reflection of selective weathering and colloidal transport, *Geochim. Cosmochim. Acta*, *65*, 521–527.

Andersson, P., L. D. Guo, I. Semiletov, O. Gustafsson, J. Ingri, O. Dudarev, and D. White (2003), Nd- and Sr- isotopes in Siberian Arctic estuarine sediments: Implications for sediment provenance, paper presented at 2003 EGS/AGU/EUG Meeting, Eur. Geophys. Soc., Nice, France.

Bracewell, J., K. Haider, S. Larter, and H.-R. Schulten (1989), Thermal degradation relevant to structural studies of humic substances, in *Humic Substances II*, edited by M. Hayes et al., pp. 181–222, John Wiley, New York.

Brown, J., M. T. Jorgenson, O. P. Smith, and W. Lee (2003), Long-term rates of coastal erosion and carbon yields, Elson Lagon, Barrow, Alaska, paper presented at 8th International Permafrost Conference, Int. Permafrost Assoc., Zurich, Switzerland.

Burman, J.-O., C. Pontér, and K. Boström (1978), Metaborate digestion procedure for ICP-AES, *Anal. Chem.*, *50*, 679–680.

Currie, L. A., et al. (2002), A critical evaluation of interlaboratory data on total, elemental, and isotopic carbon in the carbonaceous particle reference material, NIST SRM 1649a, *J. Res. Natl. Inst. Stand. Technol.*, *107*, 279–298.

Danilov, I. D. (1990), *Ground Ice* (in Russian), Nedra, Moscow.

Davidenko, E. P. (2000), Ecological and economic evaluation of the consequences of catastrophic fires in the Russian Far East: The Khabarovsk Territory example of 1998, *Int. Forest Fire News*, *22*, 53–62.

Davidson, E., S. E. Trumbore, and E. Amundson (2000), Soil warming and organic carbon content, *Nature*, *408*, 789–790.

Dudarev, O. V., I. P. Semiletov, A. I. Botsul, I. V. Utkin, A. N. Charkin, V. V. Anikiev, G. M. Kolesov, and D. Y. Sapozhnikov (2001), The coastal erosion as a significant source of the particulate matter into the Arctic shelf, paper presented at Second Wadati Conference on Global Change and the Polar Climate, IARC/Univ. of Alaska Fairbanks, Tsukuba, Japan.

Freeman, C., C. D. Evans, D. T. Monteith, B. Reynolds, and N. Fenner (2001), Export of organic carbon from peat soils: Warmer conditions may be a blame for the exodus of peatland carbon to the oceans, *Nature*, *412*, 785.

Gibson, J. A. E., W. F. Vincent, B. Nieke, and R. Pienitz (2000), Control of biological exposure to UV radiation in the Arctic Ocean: Comparison of the roles of ozone and riverine dissolved organic matter, *Arctic*, *53*, 372–382.

Gordeev, V. V., J. M. Martin, I. S. Sidorov, and M. V. Sidorova (1996), A reassessment of the Eurasian river input of water, sediment, major elements, and nutrients to the Arctic Ocean, *Am. J. Sci.*, *296*, 664–691.

Guéguen, C., D. Wang, and L. Guo (2003), Optical properties pf natural organic matter in the Yukon River Basin, *Tohoku Geophys. Res.*, *36*, 434–438.

Guo, L., P. H. Santschi, L. A. Cifuentes, S. Trumbore, and J. Southon (1996), Cycling of high molecular weight dissolved organic matter in the Middle Atlantic Bight as revealed by carbon isotopic (^{13}C and ^{14}C) signatures, *Limnol. Oceanogr.*, *41*, 1242–1252.

Guo, L., J. L. Lehner, D. M. White, and D. S. Garland (2003a), Heterogeneity of natural organic matter the Chena River, Alaska, *Water Res.*, *37*, 1015–1122.

Guo, L., N. Tanaka, D. M. Schell, and P. H. Santschi (2003b), Nitrogen and carbon isotopic composition of HMW dissolved organic matter in marine environments, *Mar. Ecol. Prog. Ser.*, *252*, 51–60.

Gustafsson, Ö., and P. M. Gschwend (1998), The flux of black carbon in surface sediments on the New England continental shelf, *Geochim. Cosmochim. Acta*, *62*, 465–472.

Gustafsson, Ö., F. Haghseta, C. Chan, J. MacFarlane, and P. M. Gschwend (1997), Quantification of the dilute sedimentary "soot-phase": Implications for PAH speciation and bioavailability, *Environ. Sci. Technol.*, *31*, 203–209.

Gustafsson, O., T. D. Bucheli, Z. Kukulska, M. Andersson, C. Largeau, J. N. Rouzaud, C. M. Reddy, and T. I. Eglinton (2001), Evaluation of a protocol for the quantification of black carbon in sediments, *Global Biogeochem. Cycles*, *15*, 881–890.

Holmes, R. M., J. W. McClelland, B. J. Peterson, I. A. Shiklomanov, A. I. Shiklomanov, A. V. Zhulidov, V. V. Gordeev, and N. N. Bobrovitskaya (2002), A circumpolar perspective on fluvial sediment flux to the Arctic Ocean, *Global Biogeochem. Cycles*, *16*, 1098, doi:10.1029/2001GB001849.

Huh, Y., M. Y. Tson, A. Zaitsev, and J. M. Edmond (1998), The fluvial geochemistry of the rivers of Eastern Siberia: I. Tributaries of the Lena River draining the sedimentary platform of the Siberian Craton, *Geochim. Cosmochim. Acta*, *62*, 1657–1676.

Jorgenson, M. T., C. H. Racine, J. C. Walters, and T. E. Osterkamp (2001), Permafrost degradation and ecological changes associated with a warming climate in central Alaska, *Clim. Change*, *48*, 551–579.

Kattner, G., J. M. Lobbs, H. P. Fitznar, R. Engbrodt, E. M. Nothig, and R. J. Lara (1999), Tracing dissolved organic substances and nutrients from the Lena River through Laptev Sea (Arctic), *Mar. Chem.*, *65*, 25–39.

Kuhlbusch, T. A. J., and P. J. Crutzen (1995), Toward a global estimate of black carbon in residues of vegetation fires representing a sink of atmospheric CO_2 and a source of O_2, *Global Biogeochem. Cycles*, *9*, 491–501.

Lobbes, J. M., H. P. Fitznar, and G. Kattner (2000), Biogeochemical characteristics of dissolved and particulate organic matter in the Russian rivers entering the Arctic Ocean, *Geochim. Cosmochim. Acta*, *64*, 2973–2983.

Martin, J.-M., and M. Whitfield (1983), The significance of the river input of chemical elements to the ocean, pp. 265–269, in *Trace Metals in Sea Water*, edited by C. S. Wong et al., Plenum, New York.

Masiello, C. A., E. R. M. Druffel, and L. A. Currie (2002), Radiocarbon measurements of black carbon in aerosols and ocean sediments, *Geochim. Cosmochim. Acta*, *66*, 1025–1036.

Middelburg, J. J., J. Nieuwenhuize, and P. Van Breugal (1999), Black carbon in marine environments, *Mar. Chem.*, *65*, 245–252.

Millot, R., J. Gallardet, B. Dupre, and C. J. Allegre (2003), Northern latitude chemical weathering rates: Clues from the Mackenzie River Basin, *Canada, Geochim. Cosmochim. Acta*, *67*, 1305–1329.

Mitra, S., T. S. Bianchi, B. A. McKee, and M. Sutula (2002), Black carbon from the Mississippi River: Quantities, sources, and potential implications for the global carbon cycle, *Environ. Sci. Technol.*, *36*, 2296–2302.

Naidu, A. S., L. W. Copper, B. P. Finney, R. W. Macdonald, C. Alexander, and I. P. Semiletov (2000), Organic carbon isotope ratios ($\delta^{13}C$) of Arctic Amerasian continental shelf, *Int. J. Earth Sci.*, *89*, 522–532.

Oechel, W. C., G. L. Vourlitis, S. J. Hastings, R. C. Zulueta, L. Hinzman, and D. Kane (2000), Acclimation of ecosystem CO_2 exchange in the Alaska Arctic in response to decadal climate warming, *Nature*, *406*, 978–981.

Persson, N. J., Ö. Gustafsson, T. D. Bucheli, R. Ishaq, K. Naes, and D. Broman (2002), Soot-carbon influenced distribution of PCDD/Fs in the marine environment of the Grenlandsfjords, Norway, *Environ. Sci. Technol.*, *36*, 4968–4974.

Peterson, B. J., R. M. Holmes, J. W. McClelland, C. J. Vorosmarty, I. A. Shiklomanov, A. I. Shiklomanov, R. B. Lammers, and S. Rahmstorf (2002), Increasing river discharge to the Arctic Ocean, *Science*, *298*, 2171–2173.

Rachold, V., M. N. Grigoriev, F. E. Are, S. Solomon, E. Reimnitz, H. Kassens, and M. Antonow (2000), Coastal erosion vs. riverine sediment discharge in the Arctic shelf seas, *Int. J. Earth Sci.*, *89*, 450–460.

Romanovskii, N. N., H. W. Hubberten, A. V. Gavrilov, V. E. Tumskoy, G. S. Tipenko, M. N. Grigoriev, and C. Siegert (2000), Thermokarst and land-ocean interaction, Laptev Sea Region, Russia, *Permafrost Periglacial Proc.*, *11*, 137–152.

Santschi, P. H., L. Wen, and L. Guo (2001), Transport and diagenesis of trace metals and organic carbon in Palos Verdes shelf sediments affected by a sewage outfall, *Mar. Chem.*, *73*, 153–171.

Savelieva, N. I., I. P. Semiletov, L. N. Vasilevskaya, and S. P. Pugach (2000), A climate shift in seasonal values of meteorological and hydrological parameters for northeastern Asia, *Prog. Oceanogr.*, *47*, 279–297.

Semiletov, I. P. (1999), The failure of the coastal frozen rock as an important factor in biogeochemistry of the Arctic shelf waters, *Doklady Earth Sci.*, *369*, 1140–1143.

Semiletov, I. P. (2000), On biogeochemical consequences of coastal processes in the Eurasian Arctic seas, paper presented at Shelf-Basin Interactions Pan Arctic Meeting, U.S. Natl. Sci. Found., SBI Project Off., Callaway Gardens, Ga.

Semiletov, I. P., N. I. Savelieva, G. E. Weller, I. I. Pipko, S. P. Pugach, A. Gukov, A. Yu, and L. N. Vasilevskaya (2000), The dispersion of Siberian river flows into coastal waters: Meteorological, hydrological and hydrochemical aspects, in *The Freshwater Budget of the Arctic Ocean*, *NATO Meeting/NATO ASI Ser.*, edited by E. L. Lewis, pp. 323–367, Kluwer Acad., Norwell, Mass.

Shivdenko, A., and J. G. Goldammer (2001), Fire situation in Russia, *Int. For. Fire News*, *24*, 41–59.

Stein, R., and R. W. Macdonald (Eds.) (2003), *The Organic Carbon Cycle in the Arctic Ocean*, Springer-Verlag, New York.

Telang, S. A., R. Pocklington, A. S. Naidu, E. A. Romankevich, I. I. Gitelson, and M. I. Gladyshev (1991), Carbon and mineral transport in major North American, Russian Arctic, and Siberian Rivers: The St Lawrence, the Mackenzie, the Yukon, the Arctic Alaskan Rivers, the Arctic Basin Rivers in the Soviet Union, and the Yenisey, in *Biogeochemistry of Major World rivers*, *Scope Ser.*, vol. 42, edited by E. T. Degens, S. Kempe, and J. E. Richey, pp. 75–104, John Wiley, Hoboken, N. J.

Waelbroeck, C., P. W. Monfray, C. Oechel, S. Hastings, and G. Vourlitis (1997), The impact of permafrost thawing on the carbon dynamics of tundra, *Geophys. Res. Lett.*, *24*, 229–232.

White, D., and L. Beyer (1999), Pyrolysis-gas chromatography/mass spectrometry and pyrolysis-gas chromatography/flame ionization detection analysis of three Antarctic soils, *J. Anal. Appl. Pyrolysis*, *50*, 63–76.

White, D., D. Garland, X. Dai, and C.-L. Ping (2002), Fingerprinting soil organic matter in the arctic to help predict CO_2 flux, *Cold Regions Sci. Technol.*, *35*, 185–194.

Yang, D., D. L. Kane, L. D. Hinzman, X. Zhang, T. Zhang, and H. Ye (2002), Siberian Lena River hydrologic regime and recent change, *J. Geophys. Res.*, *107*(D23), 4694, doi:10.1029/2002JD002542.

Zimov, S. A., S. P. Daviodov, Y. V. Voropaev, S. P. Prosyannikoiv, and I. P. Semiletov (1993), Planetary maximum CO_2 and ecosystems of the North, in *Proceedings of the International Workshop on Carbon Cycling in Boreal Forest and Sub-Arctic Ecosystems*, edited by T. S. Vinson and T. P. Kolchugina, pp. 21–34, Oregon State Univ., Corvallis, Oreg.

P. Andersson, Laboratory for Isotope Geology, Swedish Museum of Natural History, S-104 05 Stockholm, Sweden. (per.andersson@nrm.se)

O. Dudarev, Pacific Oceanological Institute, Russian Academy of Science, Vladivostok, Russia. (dudarev@poi.dvo.ru)

L. Guo and I. Semiletov, International Arctic Research Center, University of Alaska Fairbanks, 930 Koyukuk Drive, Fairbanks, AK 99775, USA. (guol@iarc.uaf.edu; igorsm@iarc.uaf.edu)

Ö Gustafsson, Institute of Applied Environmental Research (ITM), Stockholm University, 10691 Stockholm, Sweden. (orjan.gustafsson@itm.su.se)

J. Ingri, Division of Applied Geology, Lulea University of Technology, S-971 87 Lulea, Sweden. (johan.Ingri@ltu.se)

D. White, Water and Environmental Research Center, University of Alaska Fairbanks, AK 99775, USA. (ffdmw@uaf.edu)

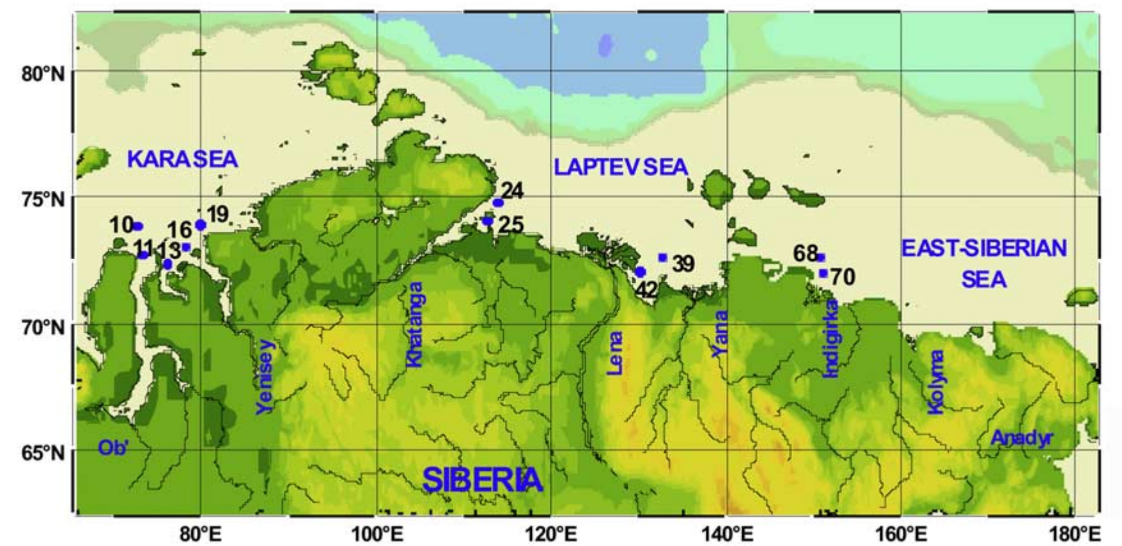

Figure 1. Map of the Siberian Arctic region and sampling locations.