Export of young terrigenous dissolved organic carbon from rivers to the Arctic Ocean

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[1] Soils in the drainage basins of Arctic rivers are a major global reservoir of aged organic carbon. The fate of this old carbon is of growing concern as the effects of climate change become more evident in the Arctic. We report natural abundance ^{14}C data indicating that dissolved organic carbon (DOC) from several Eurasian and North American rivers is predominantly young and largely derived from recently-fixed C in plant litter and upper soil horizons. Concentrations of dissolved lignin phenols, unique organic tracers of terrestrial plant material, and ^{14}C content in DOC were strongly correlated throughout the Arctic Ocean, indicating terrigenous DOC is mostly young and widely distributed in polar surface waters. These young ages of terrigenous DOC in rivers and the ocean indicate little of the old carbon stored in Arctic soils is currently being mobilized in the dissolved component of continental runoff. *INDEX TERMS:* 1040 Geochemistry: Isotopic composition/chemistry; 1615 Global Change: Biogeochemical processes (4805); 4207 Oceanography: General: Arctic and Antarctic oceanography; 4806 Oceanography: Biological and Chemical: Carbon cycling; 4850 Oceanography: Biological and Chemical: Organic marine chemistry. **Citation:** Benner, R., B. Benitez-Nelson, K. Kaiser, and R. M. W. Amon (2004), Export of young terrigenous dissolved organic carbon from rivers to the Arctic Ocean, *Geophys. Res. Lett.*, *31*, L05305, doi:10.1029/2003GL019251.

1. Introduction

[2] The Arctic Ocean receives about 10% of global riverine discharge and 25 Tg of terrigenous dissolved organic carbon (DOC) each year [*Aagard and Carmack*, 1989; *Opsahl et al.*, 1999]. Most of this organic carbon is derived from soils, one of the largest global reservoirs of organic carbon [*Dixon et al.*, 1994]. High latitude regions include the largest peat-bog systems on Earth and contain as much as half of global soil carbon content [*Dixon et al.*, 1994; *McGuire et al.*, 2002]. The fate of soil carbon in high latitude soils is uncertain as the effects of global warming and climate change are predicted to be magnified in the Arctic [*Serreze et al.*, 2000]. Thawing of the permafrost which underlies a substantial fraction of the Arctic could accelerate carbon losses from soils [*Goulden et al.*, 1998]. Freshwater discharge to the Arctic Ocean is expected to increase with increasing temperatures [*Peterson et al.*, 2002], potentially resulting in greater riverine export of terrigenous organic carbon to the ocean.

[3] Organic carbon in Arctic soils is typically old, with average radiocarbon ages ranging from centuries to millennia [*Schell*, 1983; *Schirrmeister et al.*, 2002], but little is known about the age of organic carbon in Arctic rivers. Particulate organic carbon in the Colville River, an Alaskan river that discharges into the Arctic Ocean, has a radiocarbon age of ~2000 yr BP and a substantial peat component [*Schell*, 1983]. DOC is the most abundant form of carbon in Arctic rivers [*Gordeev et al.*, 1996; *Lobbes et al.*, 2000]. Over 90% of the total organic carbon exported from Russian rivers to the Arctic Ocean is in dissolved form [*Lobbes et al.*, 2000], but radiocarbon ages of this DOC have not been reported.

[4] The large discharge of freshwater and associated terrigenous materials have a major impact on the physical features and biogeochemical cycles of the Arctic Ocean. Surface waters of the Arctic Ocean are stratified and have the highest concentrations of terrigenous DOC of any ocean basin. It is estimated that 5–33% of the total DOC in polar surface waters is of terrigenous origin [*Opsahl et al.*, 1999]. The age and fate of this DOC is largely unknown, but recent estimates indicate a substantial fraction is exported to the North Atlantic Ocean [*Opsahl et al.*, 1999; *Amon et al.*, 2003]. In the present study we report radiocarbon ages of DOC in Arctic rivers and the Arctic Ocean. These are among the first radiocarbon data reported for DOC in the Arctic, and they reveal novel insights about carbon cycling in both the terrestrial and marine environments.

2. Materials and Methods

[5] The Yenisey and Ob' rivers were sampled where they discharge into the Kara Sea during cruises on the R/V Boris Petrov. Surface water samples were filtered (<0.2 μm) and the DOC was isolated by tangential-flow ultrafiltration or vacuum evaporation [*Opsahl et al.*, 1999]. In 1997 ultrafiltration recovered 57–69% of the DOC, whereas in 1999 and 2000 the recovery of DOC by vacuum evaporation was ~100%. Surface waters were collected in 12 L carboys from the deltas of the Ikpikpuk and Kokolik rivers in 2002. After filtration (<0.2 μm), 24–43% of the DOC was recovered by C_{18} solid phase extraction [*Louchouarn et al.*, 2000].

[6] All seawater samples were collected using Niskin bottles on a CTD/rosette, and they were filtered (<0.2 μm) prior to processing. Two seawater samples were collected in 1997 and 1998 from the R/V Polarstern, and 34–36% of the DOC was recovered by ultrafiltration. In 2002 seawater samples were collected from the USCGC Healy and 24–34% of the DOC was recovered by C_{18} solid phase extraction.

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Table 1. Sampling Locations, Radiocarbon and Lignin Phenol Concentrations of DOC

Latitude (N)	Longitude (W)	Sampling Date	Depth (m)	Salinity	$\Delta^{14}C$ (‰)	Radiocarbon age (ybp)	Lignin phenols (ng/L)
Yenisey River		9/18/97	surface	1.20	150	Modern	31043
Yenisey River		9/12/00	surface	0.0	108	Modern	34437
Ob' River		9/14/97	surface	2.44	84	Modern	24512
Ob' River		9/3/99	surface	0.60	307	Modern	15982
Kokolik River		8/22/02	surface	0.15	−6	Modern	20808
Ikpikpuk River		6/4/02	surface	0.07	6	Modern	32133
72°37′24″	158°40′46″	8/6/02	11	29.874	−208	1820	210
70°40′52″	166°29′55″	8/21/02	21	31.112	−200	1740	186
72°17′30″	151°53′20″	7/29/02	33	30.919	−199	1730	187
71°23′18″	157°38′53″	7/21/02	51	32.773	−262	2380	133
81°41′20″	4°33′20″	8/21/97	80	33.968	−90	710	306
71°39′00″	155°45′48″	6/11/02	100	32.597	−190	1640	nd
72°11′28″	154°18′15″	6/7/02	101	32.860	−159	1335	296
72°33′34″	154°34′27″	6/9/02	120	32.989	−144	1200	137
72°05′31″	154°28′27″	6/5/02	127	33.434	−87	680	214
72°17′30″	151°53′20″	7/29/02	131	33.029	−323	3090	129
71°55′30″	154°50′46″	7/23/02	149	33.819	−353	3450	nd
73°40′41″	159°09′57″	8/13/02	155	33.218	−290	2700	113
72°28′41″	153°27′14″	7/27/02	170	33.449	−310	2930	128
72°09′12″	154°13′32″	7/26/02	173	33.533	−246	2220	105
72°29′24″	153°29′45″	7/27/02	1971	34.942	−379	3770	38
72°17′30″	151°53′20″	7/29/02	2949	34.957	−378	3760	22
72°29′24″	153°29′45″	7/27/02	3082	34.957	nd	nd	33
88°59′20″	102°28′10″	7/10/98	3200	34.950	−348	3385	33

[7] Radiocarbon analyses were performed at the National Ocean Sciences Accelerator Mass Spectrometry facility at the Woods Hole Oceanographic Institution and at the Leibnitz Labor Für Altersbestimmung und Isotopenforschung in Kiel, Germany. Dried samples of DOC were combusted to CO_2 and converted to graphite for ^{14}C analysis. Radiocarbon values are reported as $\Delta^{14}C$ and radiocarbon age according to *Stuiver and Polach* [1977] and *Stuiver* [1980]. Modern is defined as 95% of the radiocarbon concentration in 1950 of NBS Oxalic Acid I as described by *Olsson* [1970]. Errors averaged ±36 yr for radiocarbon ages.

[8] Lignin phenols were analyzed using the CuO oxidation method as described by *Hernes and Benner* [2003]. Lignin phenols were analyzed using a Hewlett Packard 5890A gas chromatograph with a DB5-MS capillary column and 5972 mass selective detector in selected ion monitoring mode. Errors associated with lignin phenol concentrations were $\leq$10% for river and polar surface water samples and $\leq$20% for deep-water samples.

3. Results and Discussion

[9] Water samples were collected from two of the largest Eurasian rivers, the Yenisey and Ob'. The Yenisey has a drainage area of 2.44×10^6 km^2 and annual discharge of 4.86 Tg DOC [*Lobbes et al.*, 2000]. The Ob' has a drainage area of 2.99×10^6 km^2 and an annual discharge of 3.69 Tg DOC [*Lobbes et al.*, 2000]. Both rivers drain vast areas of boreal forests (taiga) and extensive peat bogs. Together these rivers account for ~34% (8.55 Tg) of annual riverine discharge of DOC to the Arctic Ocean. In 1997 the concentrations of DOC were 537 μM in the Yenisey and 528 μM in the Ob', and during 1999 and 2000 the DOC concentrations were 698 μM in the Yenisey and 519 μM in the Ob'. Two relatively small rivers on the north slope of Alaska, the Ikpikpuk and Kokolik, were sampled in 2002. Arctic tundra dominates the watersheds of these rivers. The concentrations of DOC were 601 μM in the Ikpikpuk and 689 μM in the Kokolik.

[10] The $\Delta^{14}C$ of DOC in all Arctic river samples ranged from −6 to 307‰, indicating the presence of bomb ^{14}C from nuclear weapons testing and modern radiocarbon ages for all samples (Table 1). Young radiocarbon ages indicate the DOC is derived primarily from recently-fixed plant litter and near-surface soil horizons. DOC in the large Eurasian rivers was enriched in ^{14}C (mean $\Delta^{14}C$ = 162‰) relative to the small Alaskan rivers (mean $\Delta^{14}C$ = 0‰), possibly indicating differences in carbon cycling between taiga and tundra soils. A similar ^{14}C content ($\Delta^{14}C$ = −13‰; calculated from the average fraction modern) was reported for DOC in a small river in central Alaska [*Guo et al.*, 2003]. For comparison, the $\Delta^{14}C$ values of DOC from tropical and temperate rivers range from −158 to 365‰ with corresponding average radiocarbon ages of 1384 yr BP to modern [*Spiker*, 1981; *Hedges et al.*, 1986; *Raymond and Bauer*, 2001], indicating variable retention and aging in soils. Modern radiocarbon ages were observed previously in DOC from streams draining peat-dominated wetlands, demonstrating that the ages of soil organic carbon and DOC can differ by thousands of years [*Schiff et al.*, 1997; *Schiff et al.*, 1998; *Palmer et al.*, 2001].

[11] The $\Delta^{14}C$ data indicate that most DOC in these Arctic rivers is young, but they do not preclude the occurrence of older material. Bulk radiocarbon ages reflect an average age of the various components of DOC, so it is possible to have old DOC in a sample with a modern ^{14}C content. Seasonal variability in the ages of DOC in rivers has been linked to hydrology, with older DOC becoming apparent under base flow conditions [*Schiff et al.*, 1997; *Schiff et al.*, 1998]. In the present study all four Arctic rivers were sampled within ~3 months of peak discharge when most DOC is exported, but they are not necessarily representative of base flow conditions.

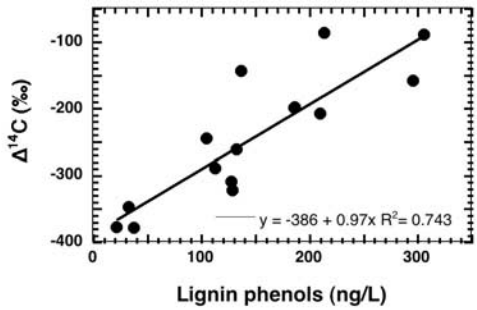

Figure 1. Relationship between the concentrations of dissolved lignin phenols and $\Delta^{14}C$ (‰) in DOC from various depths and locations in the Arctic Ocean. A linear regression of the data is shown.

[12] Freshwater discharge from Arctic rivers has residence times of 5–15 years in surface waters of the Arctic Ocean [*Schlosser et al.*, 1994]. The large reservoir of freshwater and terrigenous DOC in polar surface waters is a temporally-integrated mixture from all Arctic rivers, and it has survived biogeochemical processing in estuarine and coastal waters. Samples of DOC were collected from various depths and locations in the Arctic Ocean, including the Chukchi Sea, Canada Basin, Makarov Basin and East Greenland Current. The $\Delta^{14}C$ values of these DOC samples ranged from −379 to −87‰, corresponding to average radiocarbon ages of 3770 to 680 yr BP (Table 1). These DOC samples include both terrigenous and marine components, so the radiocarbon ages represent an average age of the mixture. The concentrations of dissolved lignin phenols, unique organic tracers of terrestrial plant material [*Opsahl and Benner*, 1997], were measured in these DOC samples to estimate the terrigenous component. Lignin phenol concentrations ranged from 22–306 ng L^{-1}, indicting the terrigenous fraction of these DOC samples spans a wide range of values (Table 1).

[13] A strong positive relationship exists between the $\Delta^{14}C$ of DOC in the Arctic Ocean and dissolved lignin phenol concentrations (Figure 1). Nearly 75% of the variability in $\Delta^{14}C$ is accounted for by variability in lignin phenol concentrations. These data indicate the terrigenous component of polar surface water DOC is enriched in ^{14}C and young relative to the marine component. These results are consistent with the modern radiocarbon ages of DOC in Arctic rivers, and they demonstrate general agreement between radiocarbon and lignin phenols as tracers of terrigenous DOC. The average radiocarbon age of DOC in Arctic rivers could change in the near future as Arctic soils warm. Radiocarbon ages of DOC in rivers will likely increase if soil moisture decreases and deeper soil horizons become a more important source of carbon [*Schiff et al.*, 1998]. Increasing average radiocarbon ages of DOC in Arctic rivers would provide strong evidence of the mobilization of the vast and relatively old carbon stored in soils.

[14] It is particularly interesting that samples with the greatest ^{14}C and lignin concentrations were found in halocline waters at depths of 80–130 m rather than the upper 50 m of the water column where phytoplankton biomass and rates of primary production are highest and salinity is lowest (Figure 2). The halocline plays a critical role in the heat budget of the Arctic Ocean, and understanding its formation and origins has been a topic of great interest for several decades [*Aagard et al.*, 1981; *Steele and Boyd*, 1998]. High concentrations of terrigenous DOC are clearly indicative of an Arctic river component in halocline waters and suggestive of halocline formation in shelf waters. Organic tracers like lignin phenols and $DO^{14}C$ can provide novel clues about the formation and origin of halocline waters and thereby enhance understanding of the consequences of climate change in the Arctic.

[15] The DOC in polar surface waters is relatively young and enriched in terrigenous DOC compared to DOC in surface waters of the north Pacific and Atlantic [*Druffel et al.*, 1992; *Opsahl and Benner*, 1997]. In contrast, the $\Delta^{14}C$ values (−379 to −348‰) and lignin phenol concentrations (22–38 ng L^{-1}) in DOC from the deep Arctic Ocean are similar to those (−414‰ to −375‰ and ∼30 ng L^{-1}) of deep-water DOC in the North Atlantic. Arctic deep waters are of Atlantic origin [*Jones et al.*, 1995], and these results indicate minimal changes in the isotopic and chemical composition of deep-water DOC occur during its residence in the Arctic.

[16] The East Greenland Current (EGC) is the major surface current exporting water and DOC from the Arctic Ocean [*Foldvik et al.*, 1988; *Amon et al.*, 2003]. High concentrations of dissolved lignin phenols and strong fluorescence signals from colored DOC indicate these waters are rich in terrigenous DOC [*Opsahl et al.*, 1999; *Amon et al.*, 2003]. Based on C-normalized yields of lignin and stable carbon isotopic compositions in the EGC sample and samples from the Yenisey and Ob' rivers, about 12 to 25% of the DOC is of terrigenous origin [*Opsahl et al.*, 1999]. Assuming the terrigenous component had the ^{14}C content of DOC in Yenisey and Ob' river water, the $\Delta^{14}C$ of the marine component of DOC in the East Greenland Current (EGC) was calculated using the following equation:

$$\Delta^{14}\text{C of DOC}_{\text{EGC}} = (\text{fraction}_{\text{ter}})(\Delta^{14}\text{C of DOC}_{\text{ter}}) + (\text{fraction}_{\text{mar}})(\Delta^{14}\text{C of DOC}_{\text{mar}})$$

where $\Delta^{14}C$ of DOC_{EGC} is −90‰, $\text{fraction}_{\text{ter}}$ is 0.12 or 0.25, $\Delta^{14}C$ of DOC_{ter} is 162‰, and $\text{fraction}_{\text{mar}}$ is 0.88 or 0.75. We estimate marine DOC in the EGC sample has an average $\Delta^{14}C$ value within the range of −124‰ to −174‰.

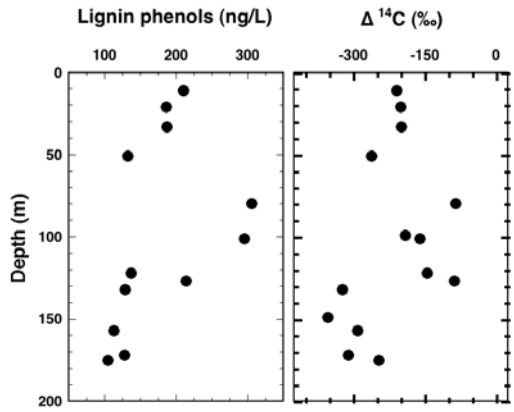

Figure 2. Concentrations of dissolved lignin phenols and $\Delta^{14}C$ (‰) in DOC from Arctic polar surface waters.

This range of $\Delta^{14}C$ values for marine DOC in surface waters exiting the Arctic is similar to the range of values (−137‰ to −238‰) for surface water DOC in the north Pacific and Atlantic [*Druffel et al.*, 1992]. It appears the terrigenous component of EGC DOC is considerably younger than the marine component.

[17] The EGC exports between 3 and 12 Tg of terrigenous DOC annually from the Arctic to the North Atlantic [*Opsahl et al.*, 1999; *Amon et al.*, 2003]. Thus, up to half of the young terrigenous DOC entering the Arctic from rivers could be exported to the North Atlantic. The development of a strong halocline and rapid transport of polar surface waters account for the export of terrigenous DOC from the Arctic Ocean, but it is surprising that relatively young terrigenous DOC escapes remineralization during its residence time in the Arctic. Photochemical processes appear to be critical for rapid and extensive decomposition of terrigenous DOC [*Kieber et al.*, 1990; *Miller and Zepp*, 1995; *Opsahl and Benner*, 1998], and these processes are currently limited in the ice-covered waters of the Arctic Ocean. At the present time it appears the young, terrigenous DOC exported from the Arctic is largely remineralized in the North Atlantic, as there is no isotopic or chemical evidence of this DOC in lower-latitude waters [*Druffel et al.*, 1992; *Opsahl and Benner*, 1997].

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