



Mobilization pathways of organic carbon from permafrost to arctic rivers in a changing climate

Laodong Guo,¹ Chien-Lu Ping,² and Robie W. Macdonald³

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[1] Arctic warming may cause the release of vast amounts of soil organic carbon (SOC) from permafrost, which will manifest itself in the fluxes and composition of organic carbon in northern rivers and Arctic coastal regions. To elucidate the transport pathways of SOC, radiocarbon composition was measured for dissolved organic carbon (DOC), particulate organic carbon (POC), sediments and SOC from the Mackenzie, Sagavanirktok, and Yukon river basins, and soil leaching experiments were conducted. The radiocarbon ages of riverine suspended POC and sediments ranged from 4430 to ~7970 yr BP, while DOC was much younger (390–1440 yr BP) except samples from the Sag River. Soil leaching experiments released <1% of SOC as DOC. The decoupling in age and partitioning between POC and DOC indicates that POC in these rivers is dominated by old SOC derived from permafrost thawing and river-bank erosion in contrast to DOC, which is more readily influenced by modern terrestrial biomass, especially in large river basins which also drain subarctic regions. These observations imply that melting of permafrost will be manifest in the age and amounts of POC in arctic rivers whereas change in DOC will reflect altered plant ecology. **Citation:** Guo, L., C.-L. Ping, and R. W. Macdonald (2007), Mobilization pathways of organic carbon from permafrost to arctic rivers in a changing climate, *Geophys. Res. Lett.*, *34*, L13603, doi:10.1029/2007GL030689.

1. Introduction

[2] The fate and transport of old soil organic carbon (SOC), which is stored in vast quantities in Arctic regions, have been receiving increased attention due to concerns about the consequences of observed and projected amplified polar warming [Serreze *et al.*, 2000; *Arctic Climate Impact Assessment*, 2005]. However, the manner in which SOC interacts with the hydrological cycle has been practically overlooked [Davidson and Janssens, 2006], even though release of organic carbon from this large pool promises to be an important manifestation of warming in the cryosphere [Frey and Smith, 2005; Striegl *et al.*, 2005]. More than two decades ago, Schell [1983] suggested that old particulate organic carbon (POC) transported by rivers was an important component of the Alaskan arctic coastal organic carbon budget. Recently, it has been shown that ancient POC from

arctic river basins and coastal erosion is an important carbon source to the arctic shelf and its sediments [Guo *et al.*, 2004; Goni *et al.*, 2005; Drenzek *et al.*, 2007]. The difficulty in understanding the consequences of projected Arctic climate change for the organic carbon cycle is that we lack systematic studies to elucidate sources, transport mechanisms and fate of old SOC transported by rivers. With warming and melting, we can anticipate the release of frozen organic carbon, potentially shifting the incorporation of this carbon into riverine and coastal foodwebs.

[3] The release of dissolved organic carbon (DOC) into rivers from peatlands and permafrost is also proposed to occur under a warming climate [Freeman *et al.*, 2001; Frey and Smith, 2005]. Presently, however, DOC in arctic and subarctic river waters is dominated by contemporary sources [Benner *et al.*, 2004; Guo and Macdonald, 2006; Neff *et al.*, 2006], implying that DOC is not strongly coupled with old SOC. If increased DOC export due to warming were to be derived mostly from the release of old SOC stored in peatlands and permafrost, the DOC in arctic rivers should then become older, reflecting the age of that reservoir [Schell and Ziemann, 1983; Matheus *et al.*, 2003]. To better understand relationships between carbon dynamics and arctic climate change requires, therefore, the integrated study of radiocarbon composition, not only for river DOC and POC, but also sediments, soils, and other organic reservoirs such as soil-DOC and plants.

[4] Stable isotopes and radiocarbon have proven to be essential tracers for deciphering source and turnover time of OC in terrestrial and marine environments, but to date there have been no simultaneous measurements of radiocarbon in DOC, POC, sedimentary OC and SOC pools in arctic environments. Here, we present $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ determinations for DOC, POC, river sediments, and tundra SOC in three arctic river basins in North America, including the Mackenzie, Sagavanirktok (Sag), and Yukon Rivers. We note that the Mackenzie and Yukon rivers include large subarctic and temperate components in their watersheds whereas the Sag River basin lies wholly within the Arctic and drains tundra [Benke and Cushing, 2005]. We have also conducted soil leaching experiments to examine the potential of soil to release DOC under warming conditions.

2. Methods

2.1. Sample Collection and Analysis

[5] River water samples and sediments were collected during 2004 summer growing season at a station near the Arctic Red River/Canada for the Mackenzie River, at Sagwon/northern Alaska for the Sag River, and at Pilot Station/Alaska for the Yukon River (Table 1). River water was pumped through a 0.45 μm Nuclepore cartridge (pre-rinsed with acid

¹Department of Marine Science, University of Southern Mississippi, Stennis Space Center, Mississippi, USA.

²Palmer Research Center, School of Natural Resources and Agriculture Sciences, University of Alaska Fairbanks, Palmer, Alaska, USA.

³Institute of Ocean Science, Fisheries and Oceans Canada, Sidney, British Columbia, Canada.

Table 1. Sampling Locations, Instantaneous Discharge, and Other Hydrographic Data^a

River	Basin Area, km ²	Ice Cover Duration, days	Sampling Location	Sampling Date, 2004	Q, m ³ /s	Conductivity, μ S/cm	DOC, μ M	POC, μ M
Mackenzie	1,706,388	~200	67.46°N; 133.62°W	Jun–22	17,400	114	343	277
				Aug–31	8,640	144	259	236
Sag	14,239	~210	69.22°N; 148.43°W	Jun–15	141	212	63	158
				Aug–30	73	283	118	52
Yukon	855,000	~180	61.94°N; 162.87°W	Jul–15	10,765	103	269	456
				Aug–25	8,924	233	218	642

^aQ, instantaneous discharge; DOC, dissolved organic carbon; POC, particulate organic carbon; Sag, Sagavanirktok River.

and milli-Q water) to separate dissolved from particulate phases [Guo *et al.*, 2003]. The first 10 liters of filtrate was discarded and subsequent filtrate was collected into polypropylene carboys. Aliquots of filtrate were sampled for the analysis of DOC and nutrients. POC samples were collected on GF/F glass fiber filters, freeze-dried and acid fumed before analysis [Guo and Macdonald, 2006]. An aliquot of DOC samples was freeze-dried to obtain powder DOC samples for isotope analysis. Sediments were collected from riverbed at 0–20 cm and stored frozen until freeze drying. Tundra soils and selected natural soil DOC samples were collected from Sagwon Hills (Sag River basin), northern Alaska. A tundra soil sample (2–15 cm) was also collected from northern Canada (near our Arctic Red River sampling site in the Mackenzie River basin) and freeze-dried for isotopic characterization. Sediments and soils were treated with 1 M HCl, and the freeze-dried powdered DOC and soil DOC samples were treated with acid (HCl) fume before measurements of OC, stable isotopes and radiocarbon [Guo and Macdonald, 2006].

2.2. Soil Leaching Experiments

[6] Soil DOC yields were measured through the leaching of tundra soil samples from both surface and 90 cm deep, with a soil/water ratio of 1:10 at a temperature of 2°C and 22°C, respectively [Xu, 2005]. Nanopure water with a DOC background of ~2 μ M was used for the leaching experiments. After 24 hr of leaching, the soil suspension was centrifuged and filtered through precombusted GF/F glass fiber filters. Filtrate from two consecutive leachings was combined for the measurements of DOC concentration to calculate DOC yield based on the mass of SOC.

3. Results and Discussion

[7] DOC concentrations are similar within the Mackenzie and Yukon rivers (218–343 μ M) but considerably lower in the Sag River (63–118 μ M). POC concentrations are comparable to DOC implying that both are important components of export flux for these rivers (Table 1).

[8] Stable C isotope compositions of DOC and POC from all three rivers (–28.90 to –26.25‰; Table 2), suggest a pervasive C3 terrestrial vegetation source [Yunker *et al.*, 1991]. However, in marked contrast to the bulk compositional data, the radiocarbon ages of these DOC and POC pools (Table 2) are distinctly different especially for the Mackenzie and Yukon Rivers (Figure 1). In the Mackenzie River, for example, $\Delta^{14}\text{C}$ values for DOC correspond to ages between 390 to 1,440 yr BP compared to 6010–7840 yr BP for POC (Table 2). Likewise, in Yukon River waters the age of the DOC is 1210–1370 yr BP compared to 6030–6510 yr BP for POC. These ^{14}C ages of bulk DOC from the Yukon River at downstream Pilot Station are similar to those measured for low-molecular-weight (LMW, <1 kDa) DOC (850–1060 y BP) collected from the upper Yukon River [Guo and Macdonald, 2006], but older than those of high-molecular-weight (HMW, 1 kDa–0.45 μ m) DOC fractions (>modern).

[9] The isotopic signatures of bulk DOC and POC samples, which contain mixtures of organic carbon from multiple sources, nevertheless provide clear evidence that we are dealing with a common C3 vegetation source of organic carbon but, in the cases of the Mackenzie and Yukon rivers, the organic carbon was produced predominantly in time periods that differ by ~4500 to 7500 years. These bulk ages imply that modern terrestrial biomass production is the important source for DOC whereas old SOC sequestered in permafrost or deep soil horizons is the important source for POC.

[10] DOC from the Sag River appears considerably older (4950 and 2170 y BP) in view of the $\Delta^{14}\text{C}$ values (–463‰ to –242‰). These $\Delta^{14}\text{C}$ values are similar to those of soil DOC collected from the Sag River basin (–579 to –256‰, Table 3). Old soil DOC could be readily released into the Sag River through active layer and soil cryoturbation (frost heave) which mixes deep soils into surface soils in arctic tundra [Ping *et al.*, 1998]. The release of old soil DOC into river waters could be amplified during low discharge years, like the one observed during summer, 2004. Together, these results suggest that the Sag River DOC is supported mainly

Table 2. Radiocarbon and Stable Isotope Composition of Riverine DOC, POC, and Sediments

River	Sampling Date	DOC			POC			Sediments		
		$\delta^{13}\text{C}$, ‰	$\Delta^{14}\text{C}$, ‰	^{14}C age, yr BP	$\delta^{13}\text{C}$, ‰	$\Delta^{14}\text{C}$, ‰	^{14}C age, yr BP	$\delta^{13}\text{C}$, ‰	$\Delta^{14}\text{C}$, ‰	^{14}C age, yr BP
Mackenzie	Jun-22	–26.57	–54	390 ± 35	–28.90	–625	7,840 ± 50	–27.06	–632	7,970 ± 55
	Aug-31	–26.25	–169	1,440 ± 30	–26.79	–530	6,010 ± 40			
Sag	Jun-15	–27.78	–463	4,950 ± 45	–26.85	–465	4,970 ± 45	–25.88	–529	6,000 ± 45
	Aug-30	–27.16	–242	2,170 ± 35	–27.04	–466	4,980 ± 40			
Yukon	Jul-15	–28.08	–162	1,370 ± 35	–28.10	–558	6,510 ± 55	–26.05	–428	4,430 ± 40
	Aug-25	–27.17	–145	1,210 ± 35	–26.62	–531	6,030 ± 55			

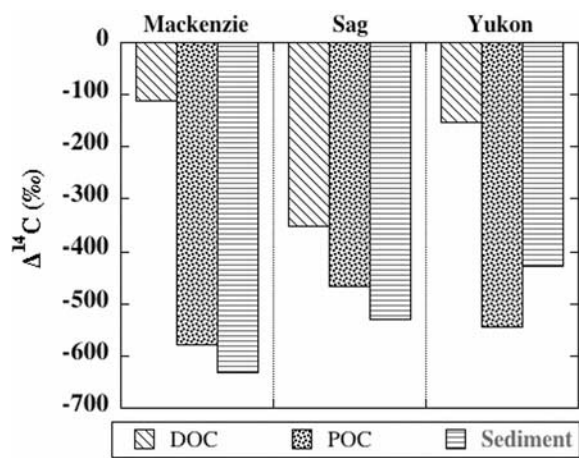


Figure 1. Variations of $\Delta^{14}\text{C}$ values among dissolved organic carbon (DOC), particulate organic carbon (POC), and sedimentary OC in the Mackenzie, Sagavanirktok (Sag), and Yukon Rivers.

by old SOC (~80%) with the remaining 20% supported by modern vegetation (Figure 2), whereas DOC in the Mackenzie and Yukon Rivers is largely supported by modern terrestrial biomass (~64–72%), although the relative sources may depend on sampling season and hydrological regime [Schell and Ziemann, 1983; Guo and Macdonald, 2006; Neff *et al.*, 2006].

[11] There are no literature data on the radiocarbon composition of bulk DOC for the Mackenzie and Sag rivers. Previous studies have shown that bulk DOC is heterogeneous with younger components being predominantly in the HMW phase and older components in the LMW phase in both river and sea waters [Guo *et al.*, 1996; Guo and Macdonald, 2006]. In addition, DOC sources can differ with sampling time and hydrological conditions [Hope *et al.*, 1994; Zou *et al.*, 2006].

[12] The tundra soils of the Sag River basin increase in ^{14}C age with depth, from the surface organic horizon (Oe; 105 yr BP), to the 40–50 cm mineral horizon (Bg; 3,260 yr BP), to the 90–100 cm cryoturbated horizon in the upper permafrost (Cf/Oe_{jj}; 7,220 yr BP) (Table 3). Surface tundra soils (upper 20 cm) from the Mackenzie Basin also contain relatively younger SOC (580 ± 35 yr BP, Table 3). In contrast, riverbed and suspended sediments from all three rivers exhibit old ^{14}C ages (4,400 – 8,000 yr BP, Table 2).

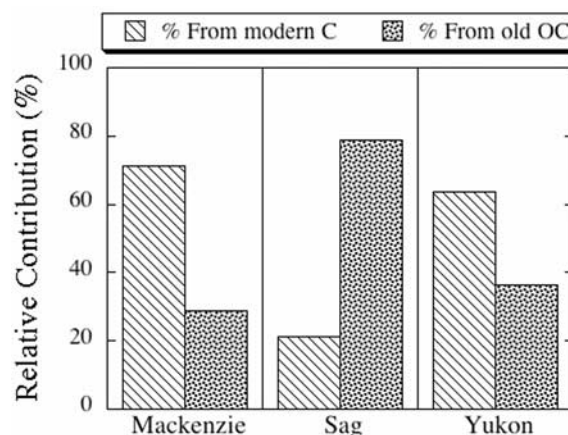


Figure 2. Relative contributions of DOC from modern primary production and old POC or soil organic carbon in the Mackenzie, Sagavanirktok (Sag), and Yukon River basins.

Clearly, the old POC transported by these rivers is derived mostly from old SOC stored in the upper permafrost, which likely enters the river through bank erosion or thawing of permafrost, rather than from surface soils which contain relatively young SOC (Table 3). Over a longer time period, the old POC delivered to the Arctic coast by these rivers has provided an important, partially-preserved component of the sedimentary OC accumulating in estuarine and coastal regions of the Siberian Arctic [Guo *et al.*, 2004] and Beaufort Seas [Goni *et al.*, 2005; Drenzek *et al.*, 2007].

[13] The increase in river DOC concentration has been ascribed to climate warming [Freeman *et al.*, 2001], but for arctic drainage basins, the connection between river DOC and old OC stored in permafrost is not at all clear. For example, there are projections of either amplified DOC release during the coming century [Frey and Smith, 2005] or decreased DOC export upon continued warming [Striegl *et al.*, 2005] and DOC production during the cold season [Finlay *et al.*, 2006]. The poor quantification of DOC sources in arctic rivers and of the relationship between DOC and climate and environmental change remains an impediment to evaluating the consequences of arctic warming and permafrost thawing. While there is growing evidence that permafrost, peatlands and lakes in Arctic regions are undergoing rapid change [Hinzman *et al.*, 2005; Jorgenson *et al.*, 2006], the younger DOC observed

Table 3. Stable Isotope and Radiocarbon Composition of End-Member Samples From Arctic Tundra

Sample	$\delta^{13}\text{C}$, ‰	Fraction of Modern	$\Delta^{14}\text{C}$, ‰	^{14}C age, yr BP
Tundra Soils (Alaska)				
0–20 cm	-26.44	0.9848 ± 0.0041	-20	105 ± 35
40–50 cm	-26.79	0.6660 ± 0.0032	-338	$3,260 \pm 40$
90–100 cm	-26.03	0.407 ± 0.0027	-596	$7,220 \pm 55$
Tundra Soils (Mackenzie)				
0–15 cm	-26.09	0.9302 ± 0.0038	-76	580 ± 35
Tundra plant-leaf	-29.06	1.0787 ± 0.0040	+72	>Modern
Tundra plant-branch	-28.56	1.0780 ± 0.0045	+71	>Modern
Soil DOC-1	-26.33	0.4229 ± 0.0024	-580	$6,910 \pm 45$
Soil DOC-2	-25.94	0.7489 ± 0.0033	-256	$2,320 \pm 35$
Soil DOC-3	-26.20	0.6206 ± 0.0030	-384	$3,830 \pm 40$
Soil DOC-4	-26.02	0.5313 ± 0.0030	-472	$5,080 \pm 45$
Soil DOC-5	-26.32	0.6447 ± 0.0034	-359	$3,520 \pm 40$

here and elsewhere for arctic rivers suggests that release of old DOC from permafrost into the hydrological cycle is not substantial. Our data show that river DOC is presently not strongly coupled with old POC and SOC for the Mackenzie and Yukon rivers, but may be quite strongly coupled for the Sag River. Reconciling the observations of age structure in SOC with the age distributions of POC and DOC in individual rivers requires not only field measurements as presented here, but also laboratory experiments to elucidate the potential for arctic soils to produce DOC.

[14] Results of soil leaching experiments show that, on average, arctic tundra soils release <1% of their total SOC under a soil/water ratio of 1:10 and a temperature of 2 or 22°C. These results are similar to those reported previously for high-latitude soils using long-term or short-term leaching experiments [Neff and Hooper, 2002; Xu, 2005]. Therefore, soil DOC yields are consistently low regardless of leaching time and water temperatures although leaching at 22°C produced slightly higher soil DOC than at 2°C. Based on radiocarbon compositions and soil leaching experiments, we infer that only limited amounts of DOC can be released from tundra soils and that mobilization of old SOC occurs predominantly through the particulate phase (Table 2).

[15] Soil DOC collected from a soil profile in the Sag River basin had a $\Delta^{14}\text{C}$ value ranging from -256 to -579‰ , which corresponds to ^{14}C ages from $\sim 2300 \pm 35$ to 6900 ± 45 yr BP (Table 3). These values compare well with those of tundra soils from the same profile, indicating that soil DOC and SOC resemble each other. The low DOC yield from tundra soils and the low DOC concentration observed for the Sag River indicate an overall limited supply of leached old SOC. In addition, older ^{14}C ages for Sag River DOC imply less modern terrestrial DOC for this basin, likely due to its relatively lower terrestrial primary production in arctic tundra. In contrast, for the Mackenzie and Yukon rivers the relatively young river DOC of higher concentration together with very low DOC yields from SOC imply a limited supply of leached old SOC but a much more vigorous supply of modern terrestrial DOC [Guo and Macdonald, 2006].

[16] Assuming river DOC is derived from two major sources, modern terrestrial biomass, with a $\Delta^{14}\text{C}$ value of $+70\text{‰}$ measured for tundra plants (Table 3), and old POC with $\Delta^{14}\text{C}$ values appropriate to each river basin (Table 2), the relative contribution of these two sources can be approximated. On average, only $\sim 28\%$ of the DOC in the Mackenzie River and 36% in the Yukon River derive from old POC, compared to 79% for the Sag River (Figure 2). However, these values are likely to change depending on climate, hydrology, and actual end-member $\Delta^{14}\text{C}$ values for old soils and plant litter. Interestingly, the contribution of modern DOC to the Sag River ($\sim 21\%$) is similar to that estimated for the terrestrial fraction ($\sim 33\%$) in fluvial transport in the north slope [Schell and Ziemann, 1983]. Younger DOC in the Mackenzie and Yukon rivers may be derived from subarctic and boreal regions of the basins which support higher terrestrial primary production with more willow and leafy plants [Chapin et al., 2000; Sturm et al., 2001]. Therefore, climate change manifested as an invasion of leafy plants into regions presently supporting only tundra (e.g., the Sag River basin) would tend to augment the DOC concentration in the Sag River and

decrease its ^{14}C age. The fractions of riverine DOC contributed from POC or SOC estimated from radiocarbon composition are considerably higher than the DOC yields from SOC measured experimentally (<1%), indicating that current old POC export by arctic rivers could be significantly higher than export of DOC or that other old DOC sources exist.

[17] From the data presented here, we infer that Arctic warming will manifest itself by increased DOC concentrations in arctic rivers supported mostly by increases in terrestrial primary production (e.g., a shift from tundra to leaf-bearing plants) and much less by increased release of DOC through melting of the permafrost (Table 2). In contrast, warming will likely lead to increased POC release due to melting of ice-bonded soils, a process that will be accompanied by a maintenance or increase in the age of riverborne POC. An important component of the entry of old POC into rivers likely will involve accelerated erosion of river banks releasing SOC from previously frozen soil horizons [e.g., Matheus et al., 2003]. To understand how the organic system is responding to changes already underway, and to provide a means to evaluate such changes at the basin scale, it is critical to establish baselines for a variety of rivers and to monitor the riverborne organic carbon components for size and age structure.

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L. Guo, Department of Marine Science, University of Southern Mississippi, Stennis Space Center, 1020 Balch Boulevard, MS 39529, USA. (laodong.guo@usm.edu)

R. W. Macdonald, Institute of Ocean Science, Fisheries and Oceans Canada, P.O. Box 6000, 9860 West Saanich Road, Sidney, BC, Canada V8L 4B2.

C.-L. Ping, Palmer Research Center, School of Natural Resources and Agriculture Sciences, University of Alaska Fairbanks, 533 E. Fireweed Avenue, Palmer, AK 99645, USA.