



Seasonal changes in the age and structure of dissolved organic carbon in Siberian rivers and streams

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[1] We examined the age and structural composition of dissolved organic carbon (DOC) transported in the Kolyma River, two large tributaries and several small upland and lowland streams in 2003. The sampling took place under ice through the winter and included the spring flood period. Radiocarbon measurements of the DOC indicated that the bulk of the annual DOC flux was modern in origin ($\Delta^{14}\text{C} > 100\%$) and pyrolysis-gas chromatography/mass spectroscopy techniques showed high concentration of terrestrial lignin monomers consistent with vigorous leaching of surface horizons during the spring thaw. By September 2003 however, little terrestrial lignin was present and the radiocarbon age became significantly older ($\Delta^{14}\text{C} < 0\%$) indicating that the mechanism of DOC generation transitions from surface to deeper soils or other terrestrial sources of old, previously stabilized C. **Citation:** Neff, J. C., J. C. Finlay, S. A. Zimov, S. P. Davydov, J. J. Carrasco, E. A. G. Schuur, and A. I. Davydova (2006), Seasonal changes in the age and structure of dissolved organic carbon in Siberian rivers and streams, *Geophys. Res. Lett.*, 33, L23401, doi:10.1029/2006GL028222.

1. Introduction

[2] Northern latitude rivers contain the highest concentrations of dissolved organic carbon (DOC) found in major global rivers [Hope *et al.*, 1994; Schlesinger and Melack, 1981] and represent an important and temperature-sensitive term in northern latitude carbon budgets [Freeman *et al.*, 2001]. Approximately 7% of northern Net Ecosystem Productivity (NEP) fluxes ($\sim 25 \text{ Tg C}$) [Gurney *et al.*, 2002] enters the Arctic Ocean annually as DOC [Opsahl *et al.*, 1999]. As a result, these fluxes play an important role in regional carbon balance, coastal margin biogeochemistry [Benner *et al.*, 2004] and carbon export to the North Atlantic [Amon and Meon, 2004; Benner *et al.*, 2005]. There has been widespread boreal warming through the last half of the 20th century [Serreze *et al.*, 2000] and the warming now appears to be accelerating [Giorgi *et al.*, 2001], causing increased freshwater discharge from the Siberian Arctic [Peterson *et al.*, 2002] and declines in

permafrost extent [Jorgenson *et al.*, 2001; Serreze *et al.*, 2000]. Over the next century, warming is likely to be greater in arctic environments than any other ecosystem on earth with major consequences for terrestrial and aquatic biogeochemical cycles [ACIA, 2005].

[3] In northeastern Siberia and other boreal regions, layers of partially humified Holocene carbon are underlain by vast deposits of loess known in Russian as Yedoma. The amount of carbon in these deposits is sizable; these deposits cover 1,000,000 km² of area in SE Siberia with an average thickness of 25 m [Romanovsky, 1993]. During the Pleistocene, much of the east Siberian Arctic was unglaciated and covered by grasslands and steppe tundra that supported extensive mammalian grazers [Gubin, 1997]. The resulting soil deposits contain organic carbon concentrations that average between 2–5% C with numerous pockets of nearly undecomposed organic matter [Dutta *et al.*, 2006; Walter *et al.*, 2006; Zimov *et al.*, 2006]. There are signs that Yedoma deposits are now thawing [Romanovsky *et al.*, 2001], and thaw rates are likely to increase as warming accelerates in the 21st century [Giorgi *et al.*, 2001]. In addition to Yedoma, Siberian lowland systems, like other boreal environments, also contain large amount of carbon distributed through surface soils, frozen sediments and peatlands [Gorham, 1991]. These complex settings yield numerous potential sources of soluble carbon that may be leached into arctic river systems. Although DOC fluxes in boreal settings are typically larger than those in temperate and tropical ecosystems, there is still limited information on the mechanisms that control DOC release to boreal and arctic aquatic ecosystems.

[4] Despite the large size, and potential environmental sensitivity of DOC fluxes from arctic rivers [Frey and Smith, 2005], there is limited information on DOC age, composition or seasonal patterns in northern environments. The available data suggest that DOC at the mouths of arctic rivers is much younger than in temperate rivers despite the large stores of ancient carbon in boreal and arctic soils [Benner *et al.*, 2004; Raymond and Bauer, 2001]. For example, in Siberia, late summer DOC in both the Ob and the Yenisey rivers is heavily labeled with radiocarbon derived from atmospheric nuclear bomb testing, suggesting a large proportion of decadal age DOC in these large river systems [Benner *et al.*, 2004]. There is also good evidence that arctic river DOC contains small amounts of lignin [Benner *et al.*, 2004, 2005; Lobbes *et al.*, 2000], indicating a terrestrial source for at least some of the material. However, most detailed measurements of arctic river DOC are made during the late-summer season and miss the bulk of DOC discharge from these environments which occur during spring floods when both flow and concentrations are

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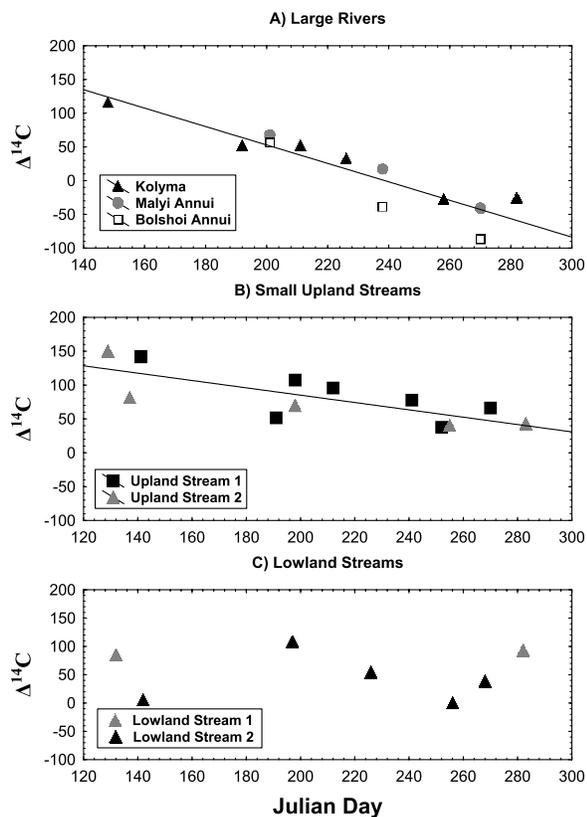


Figure 1. River and stream ^{14}C DOC through 2003. (a) The $\Delta^{14}\text{C}$ of DOC in the Kolyma River at Cherskii, Russia, and two large upstream tributaries. The regression has an $r^2 = 0.84$ ($p < 0.001$). (b) Seasonal changes in the ^{14}C content of DOC in two upland first-order streams that drain permafrost underlain basins. The regression has an $r^2 = 0.59$ ($p < 0.001$). (c) The $\Delta^{14}\text{C}$ of DOC in two lowland streams that drain seasonally flooded, non-permafrost areas around the Kolyma. There was no seasonal trend for Figure 1c.

high [Finlay *et al.*, 2006; Kohler *et al.*, 2003; Rember and Trefry, 2004]. In this study, we examined the seasonal cycle of DOC age and composition for the Kolyma River and several of its tributaries. Our research focused on measurement of both structure and age of DOC in order to obtain a better understanding of the controls over watershed-scale DOC production in boreal Siberia.

2. Methods

[5] We carried out a detailed seasonal study of DOC age, composition and flux in the Kolyma River and a range of tributaries including two large rivers that drain mountainous regions underlain by permafrost including the Malyi (middle) Annu and Bolshoi (big) Annu. We also sampled two streams draining seasonally flooded lowlands in the Kolyma River valley and two that drain upland areas. The seasonal sampling included overwinter samples taken under ice in the Kolyma and sampling during the spring flooding event. Our chemical analysis of the waters included measurement of DOC concentrations and the ^{14}C content of DOC. We also measured the specific UV Absorbance (SUVA) of DOC at 254 nm (normalized to DOC concentration) and carried

out pyrolysis-Gas Chromatography/Mass Spectroscopy (py-GC/MS) analysis on freeze-dried DOC to examine structural changes in this material. All of these methods are described in detail in the auxiliary materials¹ section of this paper.

3. Results and Discussion

[6] Both the age and chemical composition of DOC in the upland streams and large rivers indicate that the source of DOC to the Kolyma and upland streams shifts from decadal age organic matter during the snowmelt and early-summer period to progressively older carbon source(s) by August and September (Figure 1). In both the Kolyma mainstem and two large, upstream tributary rivers, there were substantial, linear declines in ^{14}C of DOC through the season to late August $\Delta^{14}\text{C}$ values of -27 to -86 ‰. These values correspond to a ^{14}C -DOC age up to 675 years before present (Figure 1) and represent an increasing proportion of older material in late-season DOC. Despite this contribution of old carbon to DOC late in the growing season, the total flux of DOC in these rivers and streams is overwhelmingly modern. In the Kolyma River, 86% of the 2003 flux of DOC occurred between May 16 (Julian Day 136) and July 19 (Julian Day 200) (Figure 2). DOC concentrations during the spring pulse reached a peak of approximately 14 mg/L (Table S1). Similar patterns occur in the small upland streams with peak runoff during the snowmelt period followed by large declines in flow (though not concentration) into the summer. Prior to this study, work on the radiocarbon age of Arctic river DOC had been based on a limited number of samples taken late in the growing season. In these studies [Amon and Meon, 2004; Benner *et al.*, 2004], late summer DOC ages on the Ob and the Yenisei were modern. While the bulk of the flux of the Kolyma DOC to the Arctic Ocean is also derived from recent primary production, the progressive increase in DOC age has not been observed before. The contrast between the Kolyma and other rivers in Siberia also illustrates that high latitude basins may have different controls over DOC flux with an apparent difference between permafrost-dominated, low peatland areas such as the Kolyma and the more peatland rich areas of western Siberia.

[7] Small upland streams that drain permafrost basins exhibit seasonal increases in DOC age. The upland stream $\Delta^{14}\text{C}$ DOC early growing season values are similar to those in larger rivers, but decline to ~ 50 ‰ by September; a value that is substantially higher than the late-season, large-river values. The lowland streams have variable but largely contemporary DOC ^{14}C signal through the growing season (Figure 1). Assuming that these upland and lowland streams are representative of the lower Kolyma watershed, changes in the age of DOC in these lower basin streams cannot explain the trends observed in the Kolyma. Additionally, the modern DOC ages from the lowland streams demonstrate that there is a relatively continuous source of modern DOC to the Kolyma through the entire growing season. Given this continued input of modern carbon through the growing season, the magnitude of the seasonal changes in Kolyma

¹Auxiliary material data sets are available at <ftp://ftp.agu.org/apend/gl/2006GL028222>. Other auxiliary material files are in the HTML.

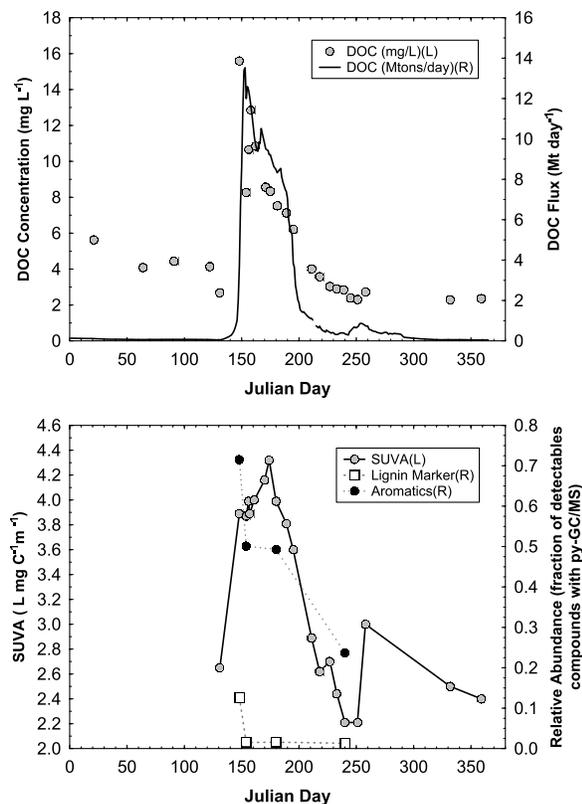


Figure 2. Kolyma concentration and flux and composition through 2003. (top) The concentration of ~ 30 DOC samples taken through the year with 7 under ice samples and a concentration of samples taken during the spring melt (Julian days 135–200). The solid line shows daily values for DOC based on a linear interpolation of data between measurement dates and daily flux values are based on the product of the daily river flow and DOC concentrations taken at mid channel. (bottom) SUVA values for DOC with higher numbers indicated greater aromaticity. The lines for lignin biomarkers and aromatics are based on py-GC/MS analyses and presented as a fraction of detectable compounds. More details on this measurement are provided in the auxiliary material section.

DOC age are even more surprising and must be the result of a significant upper-basin late-season source of old carbon.

[8] There are at least three possibilities for old carbon sources in the late season river samples. First, erosion of frozen soils as the Kolyma downcuts through Holocene and Pleistocene sediments could yield organic matter that then is solubilized in the river system. Second, permafrost thaw and deepening of active layer depths due to warming exposes older C to leaching and results in the solubilization and transport of this old carbon to river systems. A third possibility is that the DOC is being released from peatlands [Frey and Smith, 2005], but this appears unlikely because the Kolyma and Annuï rivers do not have extensive peatlands in the area upstream of the sampling locations. It is possible that in-stream processing of river DOC could selectively remove young DOC from the Kolyma late in the summer, but this possibility doesn't seem likely to account for the magnitude of observed changes given the

continued inputs of contemporary DOC from lowland streams. In addition, the seasonal increases in the age of upland streams where water has very short residence times (< 1 week), and the large seasonal increases in DOC age in the upstream Annuï tributaries make it clear that older sources of terrestrial carbon are entering these boreal watersheds during the latter portion of the growing season.

[9] Although it is impossible to directly quantify the old carbon fraction in the Kolyma due to the many possible sources and limited information on end-member age distributions, it is possible to bracket the potential size of this pool in the large rivers. To calculate a potential old carbon contribution we can assume a constant source of modern DOC with a fraction modern value of 1.04 based on the average early season value and an older soil source end-member from water extracts of upland Holocene soils (DOC fm = 0.60) or Pleistocene sediments (DOC fm = 0.16) (Table S2). This simple analysis indicates that the decline in large river DOC age could occur if 18% of the late-season Kolyma DOC was derived from the approximately 5000 year old mid-Holocene carbon or 9% derived from the 18,000 year old Pleistocene carbon (or some combination thereof). We emphasize that these rivers transport DOC with a range of ages and so this end-member calculation is useful only for bounding the potential contributions of differing sources of DOC to the rivers. Using the radiocarbon values alone, we cannot narrow our estimate of old carbon in the river system, nor can we clearly identify a source of the material. However, structural changes in DOC that accompany the increasing large river DOC ages provide some insight into the broader process level controls on DOC flux in the Kolyma River.

[10] The seasonal changes in DOC age are accompanied by a broad structural change in DOC that further supports an argument for a terrestrial source of the old carbon in the Kolyma and which has implications for understanding the role of DOC in both terrestrial and aquatic carbon budgets in the region (Figure 2). Across all streams and dates of sampling, there is a significant negative relation between $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ (Figure 3) with a tendency for the oldest carbon to be the most ^{13}C enriched. The most $\delta^{13}\text{C}$ depleted values of DOC tend to occur early in the growing season and are consistent with a source in freshly deposited organic matter. The 3 to 4 per mil enrichment in $\delta^{13}\text{C}$ in older vs. younger DOC is consistent with a more isotopically enriched source such as partially decomposed organic matter [Agren *et al.*, 1996]. Both surface soils and much older Yedoma soils have $\delta^{13}\text{C}$ signals similar to the late season DOC values in the Kolyma (Table S2). In addition to the enrichment of ^{13}C in old DOC, the specific UV absorbance (SUVA) at 254 nm of DOC is also much lower in older material than in younger material. Higher values of SUVA generally indicate the presence of dissolved aromatic carbon and the range of SUVA values in these samples from Siberian rivers imply a very sizeable difference in DOC structure across samples [Weishaar *et al.*, 2003]. In the Kolyma alone, SUVA values declined from an absorbance of $4 \text{ L}^{-1} \text{ mg DOC}^{-1} \text{ m}^{-1}$ in May to $2 \text{ L}^{-1} \text{ mg DOC}^{-1} \text{ m}^{-1}$ in September implying a very steep decline in the contribution of aromatic carbon to DOC through the growing season.

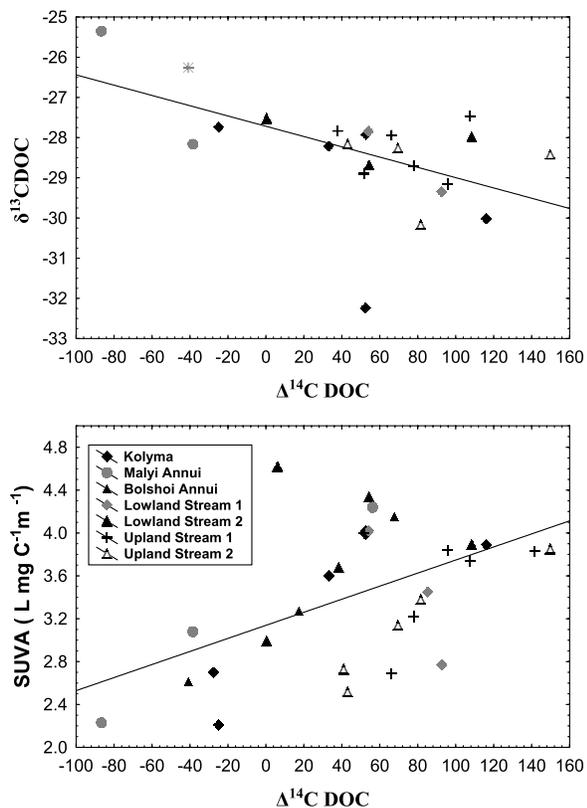


Figure 3. Markers of DOC chemistry and source. (top) The relation between ^{14}C and ^{13}C for all stream samples in the Kolyma basin. There is a strong, general correspondence between ^{14}C and ^{13}C in DOC ($r^2 = 0.47$, $p < 0.001$) if one early season Kolyma $\delta^{13}\text{C}$ value > -32 is excluded and an $r^2 = 0.29$ ($p < 0.001$) if all the points are used. (bottom) A general positive relationship between SUVA and ^{14}C that shows higher aromaticity in more modern DOC ($r^2 = 0.2598$, $p = 0.003$).

[11] The range of changes in DOC age and structure all point to shifting sources of DOC through the growing season in Siberia. During spring melt, there is significant flushing of water through very shallow surface litter and soil horizons due to frozen soils. The SUVA of DOC in the river during the spring flood suggests the presence of dissolved aromatic carbon consistent with leaching of recently deposited plant matter. Pyrolysis-gas chromatography/mass spectrometry (py-GC/MS) analysis of DOC in small streams and the Kolyma indicate that samples during the peak runoff period have relatively high concentrations of DOC attributable to relatively undecomposed terrestrial lignin as well as high overall aromatic content which corresponds to the SUVA measurements described above. These lignin monomers provide strong evidence for vigorous leaching of terrestrial organics out of surface horizons in the early growing season [Page *et al.*, 2001; Templier *et al.*, 2005]. By the end of the summer season, the relative abundance of the lignin biomarkers has declined to $\sim 2\%$ of the detectable compounds during pyrolysis. If we assume that pyrolysis detects all carbon in the sample, this relative abundance translates into $\sim 50 \mu\text{mol C/l}$ concentrations of lignin markers late in the season, consistent with observations

made with late-season CuO oxidation techniques in the Ob and Yenisey Rivers [Benner *et al.*, 2004]. Combined, the SUVA, aromatic and lignin observations all point to a seasonal shift in DOC generation from surface soils to older, structurally distinct sources late in the growing season. The seasonal decline in both lignin monomers and aromatic compounds in stream and river DOC is consistent with a seasonal shift from litter to older, humified, aromatic and lignin-poor, soil organic matter pools [Kogel-Knabner *et al.*, 1992; Lichtfouse *et al.*, 1998]. Taken together with observed changes in age and $\delta^{13}\text{C}$, the structural patterns are suggest that there is an old source of terrestrial C entering the Kolyma and its large tributaries during the growing season.

4. Conclusions

[12] DOC fluxes play an important role in both terrestrial and aquatic C budgets in northern latitudes. As a loss of carbon from terrestrial systems, DOC is a relatively small, but uncertain, term in C balance studies. For the Kolyma basin, the increase in DOC concentrations that occurs alongside increases in water flow suggests that the vast majority of DOC is produced and transported during this very brief period in the spring, probably from leaching of up-river settings where snowmelt occurs earlier in the season. The composition and age of this material is contemporary and all signs point to vigorous leaching or surface scouring of litter and surface horizons as the source of early season DOC. The overwhelming dominance of this short time period on DOC fluxes has implications for predictions of DOC flux change in the future and suggests that the annual loss of DOC will be most affected by what happens early in the growing season. Intensification of the hydrologic cycle, earlier (and perhaps elongated) spring melt periods, and changes in snowcover in the Kolyma watershed all could play more important roles in changing annual DOC fluxes than alteration of permafrost cover or decomposition cycles. Our study suggests that the major export of terrestrial DOC, and specifically lignin biomarkers, may happen very quickly during a period of a few weeks in the spring season.

[13] As the climate, vegetation cover and seasonal cycles of northern latitude systems change into the future, DOC fluxes will likely change. While this study suggests that DOC fluxes will be most affected by changes in the brief period of late spring peaks in DOC concentration and water flux, the late season fluxes of DOC may be the first to show signs of changes in terrestrial C cycling due to warming and increases in active layer thickness. The appearance of old and structurally distinct DOC in the Kolyma and our inability to assign a precise source to this carbon is intriguing. In and of itself, the appearance of old DOC in a boreal river is noteworthy because it illustrates that previously stabilized, old carbon pools are vulnerable to leaching in the Kolyma watershed; a result that differs from prior studies of northern latitude river systems. While we don't know whether or not this late-season signal is specific to the Kolyma watershed, related to ongoing regional warming or simply part of a normal seasonal cycle, it is clear that for the future, the structural and age-related signals embedded in these dissolved molecules could be

an important sign of changing hydrologic and biogeochemical cycles of boreal ecosystems.

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