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# Selective preservation of old organic carbon fluvially released from sub-Arctic soils

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[1] Amplified climate warming in the Arctic may cause thaw-remobilization of its large soil organic carbon (SOC) pool. Here we assess the remobilization and preservation of old SOC by the watershed-integrated radiocarbon signature of molecular SOC markers released from northernmost Scandinavia. The radiocarbon analyses revealed a remarkable fractionation for identical vascular plant markers (~420‰ or ~6000 $^{14}$C years) upon settling from surface water to the underlying sediments. From this, we infer fluvial export of two SOC pools; a young surface peat component, and an old deep mineral soil component. The young pool exists as an easily degradable humic suspension, while the old pool is matrix protected from degradation and ballasted for preferential settling. The two soil types with highest OC in Arctic permafrost evidently exhibit different susceptibilities to degradation. Hence, a significant part of the thaw-released OC may simply be fluvially relocated to sediments instead of being emitted to the atmosphere. **Citation:** Vonk, J. E., B. E. van Dongen, and Ö. Gustafsson (2010), Selective preservation of old organic carbon fluvially released from sub-Arctic soils, *Geophys. Res. Lett.*, *37*, L11605, doi:10.1029/2010GL042909.

## 1. Introduction

[2] Half of the global SOC pool resides in Arctic permafrost, mainly in the top three meters [*Tarnocai et al.*, 2009]. This is more than double the atmospheric C pool [*Schuur et al.*, 2008]. However, our mechanistic understanding of the fate of permafrost OC in a warming climate [*Zwiers*, 2002; *Richter-Menge et al.*, 2006] is poor, particularly regarding its propensity to be degraded to the greenhouse gases (GHG) $CO_2$ and $CH_4$ during land-to-ocean transport. The imprint of thaw-release of old SOC is mostly manifested in particulate organic carbon (POC) [*Guo and Macdonald*, 2006], while the dissolved organic matter is more reflecting changes in plant ecology [*Guo et al.*, 2007; *Raymond et al.*, 2007]. We therefore focus on POC and sediment OC to infer the fate of permafrost OC as recorded in the coastal environment. Since such bulk OC is a mixture of both marine and terrestrial sources, this study utilized specific *n*-alkanes and *n*-alkanoic acids derived from the waxes of land-based vascular plants. These compounds are established as markers for recalcitrant soil organic matter [*Smittenberg et al.*, 2006; *Goñi et al.*, 2005; *Drenzek et al.*, 2007; *van Dongen et al.*, 2008a]. Here, molecular radiocarbon analysis is used to source-apportion different terrestrial pools (i.e., young or pre-aged) and to reveal the fate of these pools during processing in a sub-Arctic river-ocean continuum. Our objective is to obtain a watershed-integrated signal of terrestrial SOC release that allows addressing remobilization and relative preservation of recalcitrant SOC components.

## 2. Study Area and Sampling

[3] The northern Scandinavian river Kalix (Figure 1) drains a catchment of tundra and boreal forests, overlying peat and till, and is one of the largest unregulated river systems in Europe. Geochemical studies show that its river chemistry represents a watershed-integrated signal [e.g. *Ingri et al.*, 2005]. Furthermore, the Kalix River exhibits similar hydrogeochemical properties to the western Eurasian Arctic rivers Ob and Yenisey [*Gustafsson et al.*, 2000; *Guo et al.*, 2004; *Ingri et al.*, 2005; *van Dongen et al.*, 2008b; *Vonk et al.*, 2008] (Figure 1). All three rivers drain a region rich in peatland underlain by discontinuous permafrost. Due to their locations along the 0°C annual isotherm these ecosystems are particularly sensitive to a warming climate.

[4] The Bothnian Bay recipient exhibits weak tidal currents and a high riverine inflow, resulting in a water column that is stably stratified in summer, ideal for process-oriented studies [*Gustafsson et al.*, 2000; *van Dongen et al.*, 2008b; *Vonk et al.*, 2008]. The high riverine inflow of total organic carbon (790 kt/year) [*Pettersson et al.*, 1997] in combination with a semi-enclosed character of the Bothnian Bay, gives this water body a high terrestrial impact. This is confirmed by high sedimentation rates (~6–7 mm/year; Vonk and Gustafsson, unpublished data, 2008). The strong isostatic uplift in the region likely increases erosion in the Kalix catchment, exposing preglacial relict regions that underlied 20–25% of the Fennoscandian ice sheet during the Last Glacial Maximum [*Fabel et al.*, 2002] (Figure S1 of the auxiliary material).[3]

[5] Suspended particulate matter and surface sediments were collected onboard R/V 'KBV005' (Umeå Marine Research Center, Sweden) from late May to early June 2005 during spring flood (Figure S2). Technical details of the sampling can be found elsewhere [*van Dongen et al.*, 2008b; *Vonk et al.*, 2008]. Samples were collected along a transect (river mouth – estuary – open bay (Figure 1)) to assess the age and degradation of released SOC during early diagenesis in the river - coastal system. The overwhelming terrestrial impact in the bay is confirmed by bulk and molecular geo-

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[3]Auxiliary materials are available in the HTML. doi:10.1029/2010GL042909.

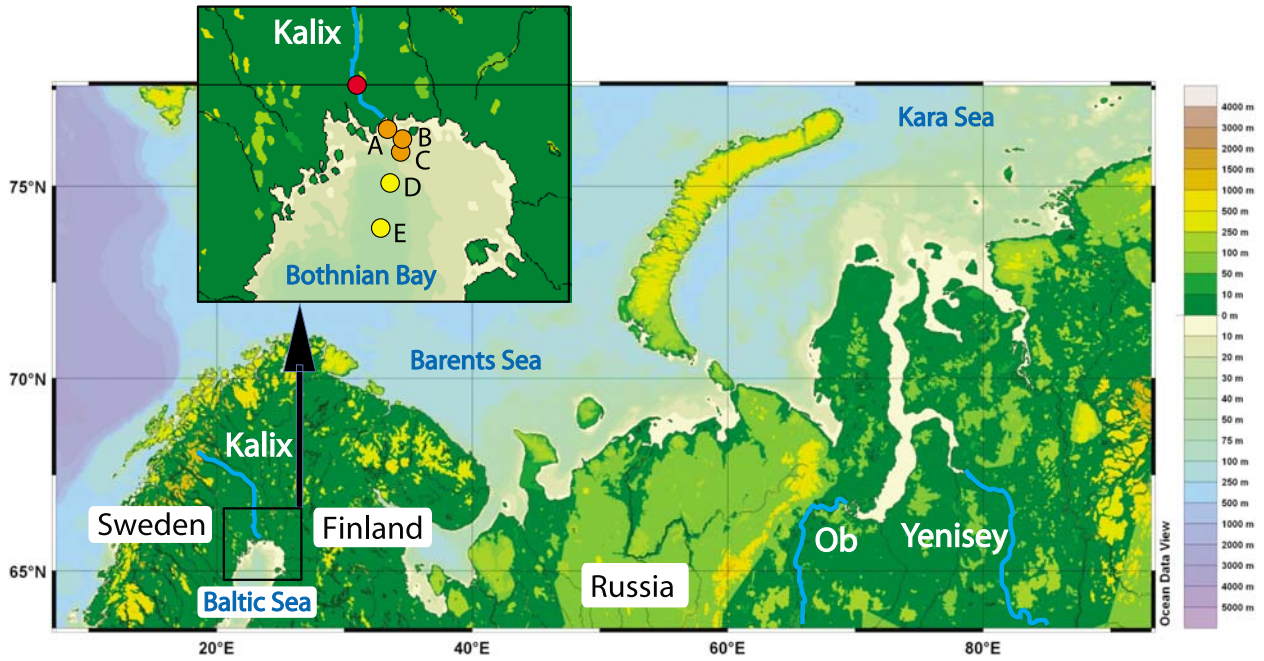


**Figure 1.** Topographical and bathymetrical map of northwestern Eurasia and its coastal seas illustrating the location of the Kalix, Ob and Yenisey Rivers, with an inset detailing our sampling locations (Kalix station in red, estuarine stations A, B, C in orange, open bay stations D and E in yellow).

chemical measurements such as the strongly depleted $\delta^{13}$C-OC (−27.4 to −25.8‰ for sediment OC; −29.4 to −28.7‰ for POC) and the high relative abundance of high-molecular-weight ($C_{27}$ + $C_{29}$ + $C_{31}$) compared to medium-molecular-weight *n*-alkanes ($C_{17}$ + $C_{19}$) [*van Dongen et al.*, 2008b; *Vonk et al.*, 2008]. Detailed fingerprinting of the terrestrial long-chain *n*-alkanes suggest that *Sphagnum*, a key plant in circum-Arctic ecosystems, is the overwhelming source of the river-exported organic matter (Figure 2a) [*Vonk and Gustafsson*, 2009]. The clear terrestrial imprint in both water column and sediment matrices provides integrated diagnostics of catchment carbon release, and the transect thus enables investigation of degradation dynamics of the released SOC during both horizontal (transport off-river) and vertical (water column to surface sediment) transport.

## 3. Methods

[6] Freeze-dried sediment and POC samples were Soxhlet extracted and fractionated as described elsewhere [*van Dongen et al.*, 2008b; *Vonk et al.*, 2008]. The HMW *n*-alkanoic acids ($C_{24}$ + $C_{26}$ + $C_{28}$) and HMW *n*-alkanes ($C_{27}$ + $C_{29}$) were isolated from the purified extracts with preparative capillary gas chromatography for offline $\Delta^{14}$C measurements. Quality control and further method information can be found in Text S1.

## 4. Results

[7] The ratio of long-chain *n*-alkanoic acids (sum of $C_{20}$ to $C_{30}$) to long-chain *n*-alkanes (sum of $C_{20}$ to $C_{32}$) systematically decreases along the transect in both suspended particulate matter and surface sediments, as well as from surface water to surface sediments (Figure 2b). The radiocarbon signal of bulk OC in the surface sediments shows contemporary values from around 0‰ in the estuary to −118‰ in the open bay (Table S1). In contrast, the compound-specific radiocarbon signal of HMW *n*-alkanoic acids shows older values, with $\Delta^{14}$C contents in POC of −153, −208 and −183‰ for the Kalix river, the estuary and the open bay, respectively (~1300–1800 $^{14}$C years (Figure 2c and Table S1)). In sharp contrast, $\Delta^{14}$C of HMW *n*-alkanoic acids in surface sediments (underlying a shallow 15–60 m water column) was −629‰ for the estuary and −568‰ for the open bay (~6700–7900 $^{14}$C years). This very old signal was further supported by HMW *n*-alkanes in surface sediments, which showed $\Delta^{14}$C values of −537, −852 and −817‰ in the estuary and −821 to −816‰ in the open bay (~6100–15,000 $^{14}$C years (Figure 2c and Table S1)).

## 5. Discussion

[8] Since POC settling times in this coastal system are on the order of days to weeks [*Gustafsson et al.*, 2000; *van Dongen et al.*, 2008b], the settling times cannot explain the $\Delta^{14}$C fractionation (corresponding to ~6000 $^{14}$C years (Table S1)) for identical compounds in suspended POC versus surface sediments. We hypothesize that the terrestrial organic carbon traced by HMW *n*-alkanoic acids is fluvially exported in two pools (Figure 3). One pool is primarily released as a humic suspension from the upper profiles of peatland, whereas the second pool originates from deeper (mineral) soil layers. The young humic SOC pool is amenable to rapid degradation processes and settles only slowly [*Gustafsson et al.*, 2000], while the old deeper SOC pool is tightly sorbed to mineral particles and hence matrix-protected against degradation [*Keil et al.*, 1994; *Huguet et al.*, 2008]. This mineral association also leads to rapid settling due to ballasting [*Gustafsson et al.*, 2000; *Klaas and Archer*, 2002]. The higher $\Delta^{14}$C values in the water column indicate an

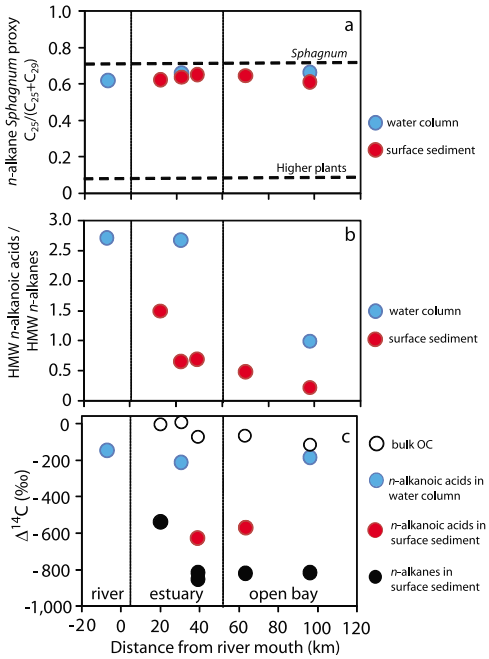


**Figure 2.** Molecular characterization of organic matter collected from surface water column and surface sediments in the Kalix River and Bothnian Bay; (a) the *n*-alkane *Sphagnum* proxy $C_{25}/(C_{25} + C_{29})$ indicative of terrestrial organic matter from peatlands [*Vonk and Gustafsson*, 2009], (b) long-chain *n*-alkanoic acids (sum of $C_{20}$ to $C_{30}$) over long-chain *n*-alkanes (sum of $C_{20}$ to $C_{32}$) indicative of degradation [*van Dongen et al.*, 2008a; *Vonk et al.*, 2008] and (c) $\Delta^{14}C$ (‰) of bulk OC (open circles), HMW *n*-alkanoic acids ($\Sigma C_{24} + C_{26} + C_{28}$) in water column (blue circles) and sediment (red circles) as well as HMW *n*-alkanes ($\Sigma C_{27} + C_{29}$) in sediment (black circles) for Kalix River, estuarine and open bay stations. Analytical uncertainties are smaller than the size of the symbols (Table S1).

influence of the fresh/young humic SOC pool, whereas the lower $\Delta^{14}C$ values in the surface sediments reflect stronger influence of the pre-aged mineral soil SOC pool, being selectively preserved. This mineral-bound SOC pool is thus remobilized but instead of being efficiently degraded to GHG, it is preferentially reburied in coastal sediments.

[9] The young pool of remobilized SOC is likely to be released from upper soil horizons and shallow peatlands in the Kalix River watershed [*Ingri et al.*, 2005] and is actively remineralized during the short passage/sedimentation period. In the circum-Arctic permafrost region, these shallow peatlands are the soils with the highest mean SOC content (69.6 kg $m^{-2}$, 0–1 m) [*Tarnocai et al.*, 2009]. This fresh terrestrial organic matter (OM) is bioavailable during fluvial and coastal transport as it mostly occurs in an amorphous buoyant suspension not protected by any mineral matrix association. Active breakdown of SOC released to these sub-Arctic coastal surface waters is supported by large atmospheric evasion of $CO_2$ [*Algesten et al.*, 2006], by a box-model study showing rapid POC degradation [*van Dongen et al.*, 2008b] and by the loss of functional groups (Figure 2b). DOC exported from Arctic rivers generally has a younger $^{14}C$ age [*Benner et al.*, 2004; *Guo and Macdonald*, 2006; *Raymond et al.*, 2007]. A fraction of this young, humic DOC aggregates into larger sizes [*Gustafsson et al.*, 2000] and contributes a young component to our POC samples. Along the transect, the $\Delta^{14}C$ of HMW *n*-alkanoic acids in POC stays fairly constant (Figure 3), which is likely a combination of ongoing degradation of fresh OM (depleting POC-$\Delta^{14}C$) and settling of older OM (enriching POC-$\Delta^{14}C$).

[10] We hypothesize that the old pool of remobilized SOC consists of terrestrial OM originating from (often deeper) mineral soils, the soil type that after the peat soil holds the next-highest mean SOC content in the circum-Arctic (32.2 kg $m^{-2}$, 0–1 m) [*Tarnocai et al.*, 2009]. The SOC markers used as tracers for the old pool represent a recalcitrant fraction both because of their structure (particularly *n*-alkanes, demonstrated by their older age) and their physical or chemical protection. The following old OC sources can be defined: (1) deeper soils in the catchment that started to develop at the beginning of the Holocene [*MacDonald et al.*, 2006], (2) deep relict organic remnants [*Lagerbäck*, 1988] from preglacial times that are now being eroded and (3) fossil OC. The latter source can be excluded since we find no evidence for this (Text S1). The remaining sources both originate from deeper down in the soil profiles. Groundwater base flow through these deeper layers is known to increase [*Frey and McClelland*, 2009] as a consequence of permafrost thaw in the catchment [*Christensen et al.*, 2004] and this would transport these old reservoirs toward the rivers. Radiocarbon dating of bulk POC from (sub-)Arctic rivers in Alaska has indeed shown release of old POC [*Guo and Macdonald*, 2006; *Guo et al.*, 2007].

## 6. Implications for Future Climate

[11] Taken together, this finding of different vulnerabilities toward degradation for the two key circum-Arctic soil types requires refining the assumption that there is a direct link between thawing of permafrost carbon and addition of GHG to the atmosphere [*Intergovernmental Panel on Climate Change*, 2007; *Khvorostyanov et al.*, 2008; *Schuur et al.*, 2008]. We evaluated the effect that this SOC recalcitrancy would have on attenuating the anticipated GHG releases to the atmosphere with a simple radiocarbon mass balance model (Text S1). The isotopic mass balance model for sub-Arctic Scandinavia suggested a preservation potential for mineral OC that was 22–162 times higher than for OC from non-mineral-soil origin (Text S1), which is in the same range as reported by others [*Huguet et al.*, 2008]. Assuming that the relative recalcitrancy of peat versus mineral soil SOC from sub-Arctic Scandinavia is representative for the relative recalcitrancies of these soil forms for the circum-Arctic, this implies that a considerable part of the remobilized C will simply be fluvially relocated to sediments instead of being emitted to the atmosphere as $CO_2$ or $CH_4$. Hence, the assumption of a causal relationship between thawing of permafrost and GHG emissions needs revision, since the two types of Arctic soil with highest OC content apparently exhibit different susceptibilities towards degradation.

## 7. Conclusions

[12] The compound-specific radiocarbon results for surface water and surface sediments suggest that SOC released

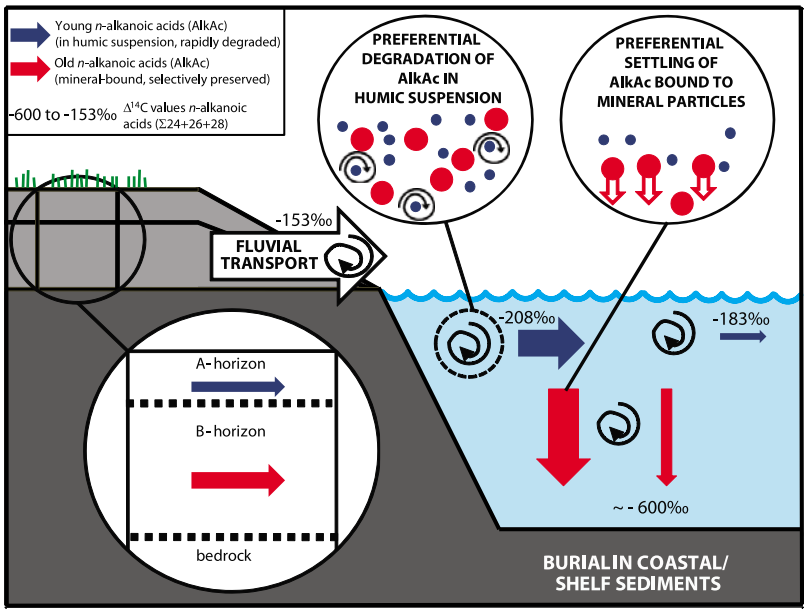


**Figure 3.** The release of sub-Arctic terrestrial organic matter (as traced by HMW *n*-alkanoic acids) in two different pools; a young, humic pool (blue) released from top soils that is easily degraded upon arrival in the coastal system and an old pool (red) released from deeper soil layers that is mineral-associated and thereby both recalcitrant to degradation and ballasted for rapid sedimentation. Numbers (−153 to −600‰) represent $\Delta^{14}C$ of HMW *n*-alkanoic acids ($\Sigma C_{24} + C_{26} + C_{28}$).

from sub-Arctic Scandinavia is transported in two separate pools to the Bothnian Bay. The youngest pool is derived from surficial peat and degrades rapidly already in the coastal water column, while a pre-aged SOC pool from deeper mineral soil layers shows selective preservation and settling. We thus infer that these two pools, originating from the two circum-Arctic soil types with the highest SOC content, demonstrate different potentials for positive feedback to climate warming. Thaw-induced release of the peat-OC with its high degradation potential would be expected to add GHG to the atmosphere. In contrast, thaw-induced release of the mineral soil-OC would be expected to be preserved selectively due to its mineral-matrix protection with restorage in coastal sediments.

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