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Widespread release of old carbon across the Siberian Arctic echoed by its large rivers

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Abstract

Over decadal-centennial timescales, only a few mechanisms in the carbon-climate system could cause a massive net redistribution of carbon from land and ocean systems to the atmosphere in response to climate warming. The largest such climate-vulnerable carbon pool is the old organic carbon (OC) stored in Arctic permafrost (perennially frozen) soils. Climate warming, both predicted and now observed to be the strongest globally in the Eurasian Arctic and Alaska, caused thaw-release of old permafrost carbon from local tundra sites. However, a central challenge for the assessment of the general vulnerability of this old OC pool is to deduce any signal integrating its release over larger scales. Here we examine radiocarbon measurements of molecular soil markers exported by the five Great Russian-Arctic Rivers (Ob, Yenisey, Lena, Indigirka and Kolyma), employed as natural integrators of carbon release processes in their watersheds. Average radiocarbon ages of *n*-alkanes increased east-to-west from 6400 yr BP in Kolyma to 11 400 yr BP in Ob, consistent with a warmer climate and more degraded organic matter westward. The dynamics of Siberian permafrost can thus be probed via radiocarbon river signals. Old permafrost carbon is at present vulnerable to mobilization over continental scales. Climate-induced changes in the radiocarbon fingerprint of released permafrost carbon will likely depend on changes in both permafrost coverage and Arctic soil hydraulics.

1 Introduction

Recent upward revision of the shallow soil OC (SOC) stock of northern permafrost elevates it to half of the global SOC pool (Tarnocai et al., 2009). In contrast to dynamic changes in the extent of Arctic sea ice (Lindsay et al., 2009), which can be monitored over large scales by satellites, thawing of this Arctic belowground cryosphere – permafrost – is more evasive toward large-scale monitoring (Gruber et al., 2004; McGuire et al., 2009). A possible approach toward probing the large-scale characteristics of

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permafrost carbon release is provided by the realization that permafrost degradation mobilizes thawed-out organic matter (OM) to streams and rivers, ultimately emptying into the coastal Arctic Ocean (Stein and Macdonald, 2004; Guo et al., 2007; van Dongen et al., 2008; Frey and McClelland, 2009). Hence, to overcome the heterogeneity and upscaling challenges posed by the Arctic landscape mosaic, we here examine the five Great Russian-Arctic Rivers (GRARs; Ob, Yenisey, Lena, Indigirka and Kolyma; Fig. 1a), extended by the westward neighboring Kalix River draining sub-Arctic Scandinavia (Fig. 1a), as natural integrators and a means to study the carbon release processes in their watersheds (Bianchi and Allison, 2009).

Different riverine carbon forms trace different components of terrestrial OM. Previous studies have reported on the ^{14}C signal of dissolved OC (DOC) in the GRARs, with generally young ^{14}C ages in the range of a few hundred years (Benner et al., 2004; Guo and Macdonald, 2006; Neff et al., 2006; Guo et al., 2007; Raymond et al., 2007), reflecting that this component stems from fresh plant litter and thus traces vegetation dynamics in the drainage basin. In contrast, detailed radiocarbon studies on Arctic soil-leaching and river-release of OM imply that permafrost thawing is predominantly manifested in the age of the particulate OC (POC) form in Arctic rivers with reported ^{14}C ages of several thousand years (Goñi et al., 2005; Guo and Macdonald, 2006; Guo et al., 2007; Vonk et al., 2010a). Unfortunately, bulk POC may stem from multiple sources including peat, mineral soils and plankton (Schuur et al., 2008; Vonk et al., 2010a, b). To circumvent issues of unknown mixtures from multiple OC sources, this study utilized vascular plant-wax derived high-molecular-weight (HMW) *n*-alkanes and *n*-alkanoic acids, as established molecular markers for soil OM (Goñi et al., 2005; Smittenberg et al., 2006; van Dongen et al., 2008; Vonk et al., 2010b). With the overarching objective of assessing current patterns in thaw-induced mobilization of old permafrost carbon over Eurasian-Arctic scales, we here constrained the radiocarbon signature of molecular markers of SOC in surface sediments collected near the mouths of the GRARs.

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2 Materials and methods

2.1 Study area and sampling

The GRARs and Kalix River span over >5000 km (140 degrees longitude) and represent different climatology, permafrost coverage, vegetation zones and other drainage-basin and river export characteristics (Fig. 1a; Table 1). The Lena, Indigirka and Kolyma are predominantly located in the continuous permafrost region with a drier and colder climate and vast amounts of deciduous forest. This contrasts with the Ob, Yenisey and Kalix watersheds, located in the discontinuous or sporadic permafrost zone, which is wetter and thus holds more extensive peat and wetlands (Kremenetski et al., 2003; Tarnocai et al., 2009).

The Arctic surface sediments were collected in 2004 and 2005 using the H/V *Ivan Kireev* (Archangelsk) from the estuaries of the five GRARs during the second and third Russia-United States cruises. The Kalix estuary sediments were obtained in 2005 using the research vessel "KBV005" from the Umeå Marine Research Center (UMF, Norrbyn, Sweden). The complete sampling details for all six locations are described elsewhere (van Dongen et al., 2008; Vonk et al., 2008). The obtained sediments were kept frozen at −20 °C until processed in the laboratory.

2.2 Methods

2.2.1 Extraction and fractionation

The bulk sediments were thoroughly mixed prior to sub sampling and small amounts of material were used for bulk radiocarbon analyses. Sub samples, typically between 70–160 g, were freeze dried, grinded, solvent extracted, purified and separated into lipid fractions using column chromatography. Aliquots of the hydrocarbon and acid fractions were then analyzed using gas chromatography/mass spectrometry (GC/MS). Details of the analytical protocols are provided in the Supplement Methods.

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2.2.2 Preparative capillary gas chromatography and compound-specific radiocarbon analysis

Individual HMW *n*-alkanoic methyl esters and HMW *n*-alkanes were isolated from the purified extracts with preparative capillary gas chromatography (pcGC) (Supplement and Fig. S1). The pcGC system was constructed around an Agilent 6890N GC-FID with a 7683 Series injector (Agilent Technologies; Palo Alto, USA) with a cold injection system (CIS3) and preparative fraction collector (PFC) (both from Gerstel GmbH; Mülheim an der Ruhr, Germany). The isolated compounds (purities between 96 and 99%) were transferred from the PFC trap capillaries with dichloromethane. Blanks swab tests in the laboratory facilities all showed natural levels of ^{14}C (Supplement Methods). To obtain sufficient amounts of carbon for ^{14}C analyses, the isolated C_{27}+C_{29}+C_{31} *n*-alkanes and C_{24}+C_{26}+C_{28} *n*-alkanoic methyl esters were recombined, respectively. The ^{14}C results for *n*-alkanoic methyl esters were corrected for measured ^{14}C values of the methyl group from the derivatization agent (BF_3 in methanol) to obtain the inherent ^{14}C value for the *n*-alkanoic acids. Finally, the isolated fractions and bulk sediment samples were quantified at the US National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility of the Woods Hole Oceanographic Institution (WHOI, Woods Hole, USA) for $\Delta^{14}C$ content. See Supplement Methods for further details.

3 Results and discussion

3.1 Sources of organic matter

The estuarine surface sediments provide integrated diagnostics of land-based carbon release as demonstrated by terrestrial signatures of stable carbon isotopes of total OC (Table 1) and the dominance of terrestrial HMW *n*-alkanes over marine low-molecular-weight *n*-alkanes by an average factor 40 (Fig. 2a). The detailed fingerprint within the terrestrial HMW *n*-alkanes suggests that *Sphagnum*, a key plant in peatland ecosystems, is a major contributor to the river-exported OM (Fig. 2b), particularly in

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the western Eurasian Arctic, that holds the world's largest peatland (Kremenetski et al., 2003).

3.2 Degradation status of remobilized organic matter

The degradation status of mobilized SOC is an important property that relates to its propensity to be converted microbially to greenhouse gases. The degradation of OM is associated with a loss of functional groups. The ratio of HMW *n*-alkanoic acids to HMW *n*-alkanes is therefore a proxy for degradation status (e.g., Goñi et al., 2005; van Dongen et al., 2008; Vonk et al., 2010a). Here, the increasing contribution of HMW *n*-alkanoic acids relative to HMW *n*-alkanes from west to east among the GRARs (Fig. 2c; Table 1) follows the continent-scale eastward trend of colder climate and more extensive permafrost coverage (Fig. 1a). Simultaneously, an increasing ^{14}C age of released bulk OC eastward (Fig. 2d) presumably reflects that more extensive permafrost coverage yields higher reservoir ages. Hence, these observations indicate a continent-scale trend of older yet less degraded terrestrial OM being released toward the eastern reaches of Siberia. This combines with previous molecular-based findings from Russian-Arctic Rivers (Guo et al., 2004; van Dongen et al., 2008; Vonk et al., 2010a) to suggest that fresh biomass produced during the short vegetative season is preserved with little alteration in the "deep freezer" of the East Siberian Arctic. The more degraded nature of the OM fluvially released in western Siberia may indicate what may occur with the deep-frozen OM in the east if large-scale thawing were to take place in its watersheds, now experiencing the largest temperature increase on Earth (Richter-Menge and Overland, 2010).

3.3 Age and origin of organic matter

The compound-specific radiocarbon signal of HMW *n*-alkanes provides a more distinct source-specific picture of mobilized old SOC. Radiocarbon ages of HMW *n*-alkanes in the three eastern GRARs, whose drainage areas are largely located in the continuous

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permafrost zone (Table 1), were 6000–6800 yr BP (Fig. 2d). Moving west into watersheds with more discontinuous permafrost, the values increased systematically from 8600 yr BP for Yenisey via 11 400 yr BP for Ob to 13 600 yr BP for Kalix (Fig. 2d, Table 2). This ubiquitously depleted ^{14}C signal demonstrates for the first time that old SOC is now leaking out from across the entire Eurasian Arctic region. Probing this riverborne molecular radiocarbon signal over time thus offers the possibility to monitor climate-warming induced changes in large-scale releases of permafrost carbon.

3.4 Continental scale trends in carbon release

While there is close agreement in ^{14}C signal between bulk OC and HMW *n*-alkanes in East Siberia, there is an increasing ^{14}C age fractionation moving westward (Fig. 2d). What system processes give rise to these dichotomous geospatial trends? Fractionating contributions from either planktonic or petrogenic sources are ruled out based on molecular and isotopic compositions (Supplement Text). We hypothesize that the age offset between bulk carbon and molecular SOC markers in the western watersheds, but absence of such an age offset in the east, is reflecting their differences in permafrost characteristics, associated hydrology and resulting carbon releases. A proposed consequence of permafrost thawing is transition from surface-water dominated transport toward groundwater dominated transport with uncertain but potentially substantial implications for fluvial release of permafrost carbon (Frey et al., 2007; Bense et al., 2009; Frey and McClelland, 2009; Lyon and Destouni, 2010). We suggest that the west-east offset between bulk and molecular ^{14}C signals combined with the continent-scale trend of younger ^{14}C age of mobilized molecular SOC markers eastward is a manifestation of the biogeochemical implication of differing hydraulic pathways imposed by differences in permafrost distribution (Fig. 1b and c Schematics).

The discontinuous permafrost landscape in the wetter West Siberia and sub-Arctic Scandinavia is dominated by extensive peatlands (Supplement Fig. S2), which developed explosively 14 000–8000 yr BP (MacDonald et al., 2006). The high riverine bulk DOC load in western GRARs carries a young ^{14}C signal (Benner et al., 2004; Raymond

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et al., 2007); estuarine flocculation may contribute to the here younger ^{14}C of bulk OC. In contrast, the radiocarbon ages of the HMW *n*-alkanes, tracing the permafrost SOC (Guo and Macdonald, 2006; Guo et al., 2007), place constraints on wherefrom in the system soil carbon is mobilized. The comparable ages of the river-integrated molecular SOC markers and the peat basal ages in the western catchments suggest that deep hydraulic conduits contribute to OM mobilization. These old ^{14}C signals (8600 to 13 600 yr BP) from the Western GRARs and Kalix River suggest that the regime shift to increased groundwater flow for regions being (in the future) characterized by discontinuous permafrost coverage (Bense et al., 2009; Frey and McClelland, 2009; Lyon and Destouni, 2010) will bring carbon release from old dormant permafrost reservoirs. Further support for a role of a deep conduit in the mobilization of old tundra SOC over West Siberia is provided by a six-fold elevation of mineral-weathered inorganic solutes in permafrost-free compared to permafrost-influenced watersheds (Frey et al., 2007). Mobilization of old permafrost OC can also occur more abruptly through thaw slumping caused by thermokarst development, mostly confined to areas of discontinuous permafrost (Schuur et al., 2008; Frey and McClelland, 2009). The observed increase in drainage of thermokarst thaw lakes (Smith et al., 2005) will increase groundwater storage and discharge. Irrespective of exact system location, the current fluvial mobilization of old permafrost SOC across extensive Western Eurasian Arctic scales demonstrates the vulnerability of this huge belowground carbon pool toward re-entering the modern carbon cycle.

For the eastern GRARs, fluvial releases of recalcitrant SOC are impeded by the extensive coverage of continuous permafrost in their drainage basins (Fig. 1a; Table 1). Here, the water movement occurs largely in the thin active layer (seasonal thaw) above the permafrost table (Fig. 1c). The spring flood is likely to transport material mostly from the surface of the underlying frozen vegetation (Frey and McClelland, 2009), but as the active layer deepens throughout the summer, the zone of water movement is in the lower reaches of the active layer. The only mechanism whereby any deeper SOC can be fluvially released is via thermokarst, river bank erosion and permafrost cracks.

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Hence, we hypothesize that the younger, yet also old, ^{14}C ages of the recalcitrant SOC of the eastern GRARs (6000–6800 ^{14}C yr), along with a smaller offset (≤1000 yr) between bulk OC and molecular soil markers, largely reflect thaw release of permafrost carbon at the bottom of the active layer (Fig. 1c).

4 Conclusions and broad scale implications

The radiocarbon fingerprinting of molecular markers of old permafrost carbon released to the great rivers draining pan-arctica provides one means of moving beyond point-based studies (e.g. Dorrepaal et al., 2009; Schuur et al., 2009) toward a spatially-integrated assessment of the vulnerability of this massive belowground carbon pool. This initial assessment suggests that old permafrost carbon indeed is vulnerable to mobilization across the scale of the Eurasian Arctic with systematic variations in age and degradation status that can be related to permafrost coverage with an important role for groundwater-mediated carbon releases. Amplified warming of the Arctic permafrost region (Zwiers, 2002; ACIA, 2004; Richter-Menge and Overland, 2010) could bring an increase in active layer depth, enhanced river bank erosion, thermokarst formation and opening of dormant/deep hydrological flow paths for release of old permafrost carbon (Schuur et al., 2008; Bense et al., 2009; Frey and McClelland, 2009). From the data presented here, we infer that effects of Arctic warming on the Eurasian-Arctic belowground carbon pool will manifest itself by increased thaw-release of old SOC and hydraulic discharge at depth to the rivers. The thawing of the Arctic megapool of permafrost carbon can be monitored complementary through both changing river hydraulics (Frey et al., 2007; Bense et al., 2009; Lyon and Destouni, 2010) and the ^{14}C age of molecular SOC markers.

Supplementary material related to this article is available online at: http://www.biogeosciences-discuss.net/8/1445/2011/ bgd-8-1445-2011-supplement.pdf.

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Table 1. Sample location, hydrological and bulk geochemical data of the surface sediments of Eurasian Arctic River Estuaries.

	Kalix	Ob	Yenisey	Lena	Indigirka	Kolyma
Geological and physiographic regions[a]	Scandinavian Mountains	West Siberian Lowlands	West Siberian Lowlands	Central Siberian Plateau	East Siberian Highlands	East Siberian Highlands
Latitude; Longitude	65°37′ N; 23°25′ E–65°26′ N; 23°19′ E[b]	72°65′ N; 73°44′ E	72°61′ N; 79°86′ E	71°96′ N; 129°54′ E	72°06′ N; 150°46′ E–71°02′ N; 152°60′ E[b]	70°00′ N; 163°70′ E
Basin area (10^6 km^2)[c]	0.24	2.54–2.99	2.44–2.59	2.43–2.49	0.36	0.65–0.66
TOC/POC flux ($t\,km^{-2}\,y^{-1}$)[d]	1.4/0.099	1.1/0.14	1.8/0.066	1.9/0.49	1.2/0.47	1.5/0.48
Permafrost coverage[e]	5/15/80	5/26/69	12/76/12	35/65/0	100/0/0	100/0/0
TOC ($mg\,g^{-1}$)[f,g]	16.0 ± 0.1/15.4 ± 0.1	9.2 ± 0.2	19.4 ± 0.3	4.8 ± 0.2	14.6 ± 0.2	17.3 ± 1.1
$\delta^{13}C_{TOC}$ (‰)[g]	−27.3 ± 0.1/−26.1 ± 0.1	−27.4 ± 0.1	−26.5 ± 0.1	−25.0 ± 0.1	−26.6 ± 0.1	−26.7 ± 0.1
$TAR^{g,h}_{n-alkane}$	20	80	49	17	50	46
HMW *n*-alkanoic acids/ HMW *n*-alkanes[g,i]	0.59	0.15	0.24	0.57	0.33	0.83
$C_{25}/(C_{25}+C_{29})$ *n*-alkanes[g,j]	0.65	0.52	0.44	0.39	0.38	0.41

[a] according to AMAP (1998)
[b] combined surface sediments along a transect
[c] data from Gordeev et al. (1996); Holmes et al. (2002); Ingri et al. (2005); Rachold et al. (2004)
[d] kalix data from Ingri et al. (2005), GRAR data from Stein and Macdonald (2004)
[e] given as % continuous; % discontinuous; % non-permafrost calculated from Walker (1998) and estimated from Johansson et al. (2006)
[f] TOC, total organic carbon
[g] data from van Dongen et al. (2008) and Vonk et al. (2008), Kalix data are from stations with sample ID C and D
[h] $TAR_{n-alkane}$, ratio of terrigenous (sum of C_{27}, C_{29} and C_{31}) to aquatic (sum of C_{17} and C_{19}) *n*-alkanes
[i] ratio of high-molecular weight (HMW; sum of C_{20}–C_{30}) *n*-alkanoic acids to high-molecular weight (sum of C_{20}–C_{32}) *n*-alkanes
[j] according to Vonk et al. (2009)

Table 2. Carbon isotopic composition and age of bulk surface sedimentary organic carbon, high-molecular-weight (HMW) *n*-alkanes (C_{27}+C_{29}+C_{31}) and HMW *n*-alkanoic acids (C_{24}+C_{26}+C_{28}) in surface sediment of Eurasian Arctic River Estuaries. $\delta^{13}C$ and $\Delta^{14}C$ results are given in per mil measured relatively to VPDB and NBS Oxalic Acid, respectively. For ^{14}C isotope analysis the results are also presented as $Fm_{\delta^{13}C\,corr}$ (relative to NBS Oxalic Acid I).

Sample ID	$\delta^{13}C$ (‰)[a]	$\Delta^{14}C$ (‰)	^{14}C $Fm^{b}_{\delta^{13}C\,corr}$	^{14}C Age (yr BP)[c]
Bulk OC				
Kalix[d]	−26.7	−74.2 ± 37	0.932 ± 0.03	570 ± 250
Ob	−27.4	−314 ± 3	0.691 ± 0.003	3000 ± 35
Yenisey	−26.5	−175 ± 3	0.831 ± 0.003	1500 ± 30
Lena	−25.0	−609 ± 3	0.394 ± 0.003	7500 ± 60
Indigirka	−26.6	−527 ± 3	0.476 ± 0.003	6000 ± 50
Kolyma	−26.7	−502 ± 2	0.501 ± 0.003	5600 ± 50
n-alkanes (C_{27}+C_{29}+C_{31})				
Kalix[d]	−31.1	−818 ± 3	0.183 ± 0.004	13 600 ± 170
Ob	−25.0	−760 ± 5	0.242 ± 0.005	11 400 ± 160
Yenisey	−32.8	−660 ± 7	0.342 ± 0.007	8600 ± 160
Lena	−31.8	−573 ± 6	0.430 ± 0.006	6800 ± 110
Indigirka	−31.6	−530 ± 10	0.473 ± 0.01	6000 ± 170
Kolyma	−32.0	−551 ± 7	0.452 ± 0.007	6400 ± 120
n-alkanoic acids (C_{24}+C_{26}+C_{28})				
Kalix[d]	−32.1	−599 ± 43	0.404 ± 0.04	7300 ± 760
Ob	−32.1	−719 ± 6	0.283 ± 0.006	10 100 ± 170
Yenisey	−32.1	−525 ± 5	0.478 ± 0.005	5900 ± 80
Lena	−24.0	−500 ± 6	0.503 ± 0.006	5500 ± 100
Indigirka	−30.3	−693 ± 6	0.309 ± 0.006	9400 ± 150
Kolyma	−32.9	−532 ± 9	0.471 ± 0.009	6000 ± 150

[a] error of the $\delta^{13}C$ measurement is ±0.1‰
[b] Fm is fraction of modern
[c] yr BP is years before present
[d] data obtained from Vonk et al. (2010); average values of sediments at stations with sample ID C and D

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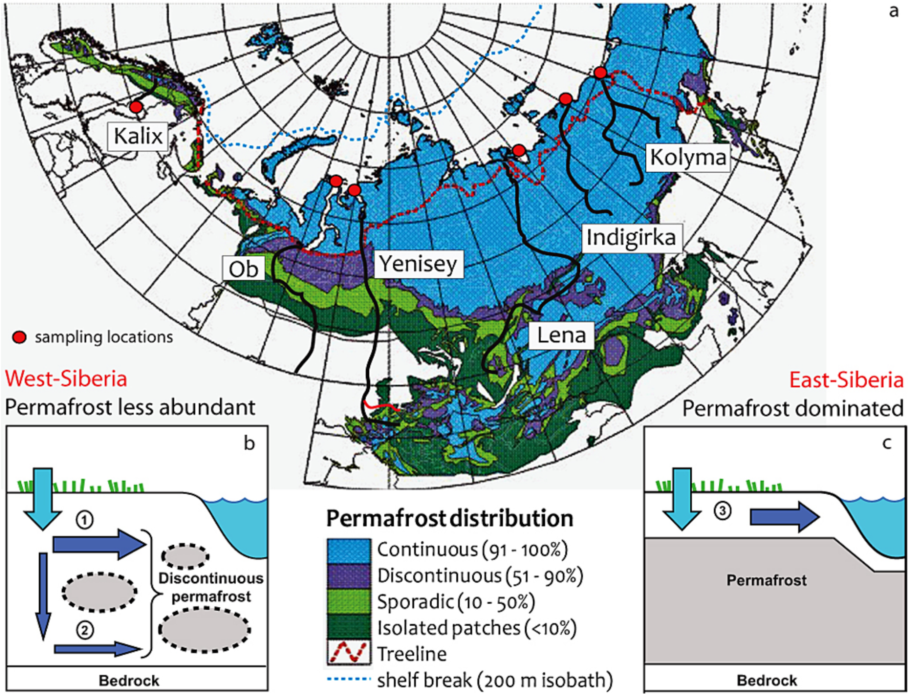

Fig. 1. The Eurasian Arctic with permafrost distribution, studied rivers and possible mechanism of fluvial carbon releases. **(a)** Permafrost distribution in the Eurasian Arctic (modified from Tarnocai et al., 2009) with Kalix and Russian Arctic rivers (black lines) and sampling locations (red circles). **(b)** In West-Siberian discontinuous permafrost, top soils (transport route 1) deliver bulk organic carbon (OC) with relatively young ^{14}C while groundwater and thermokarst-related transport (transport route 2) delivers more refractory, older OC. **(c)** Continuous permafrost in East-Siberia restricts carbon transport predominantly to the active layer (transport route 3) causing release of bulk OC and refractory lipids of similar ^{14}C age.

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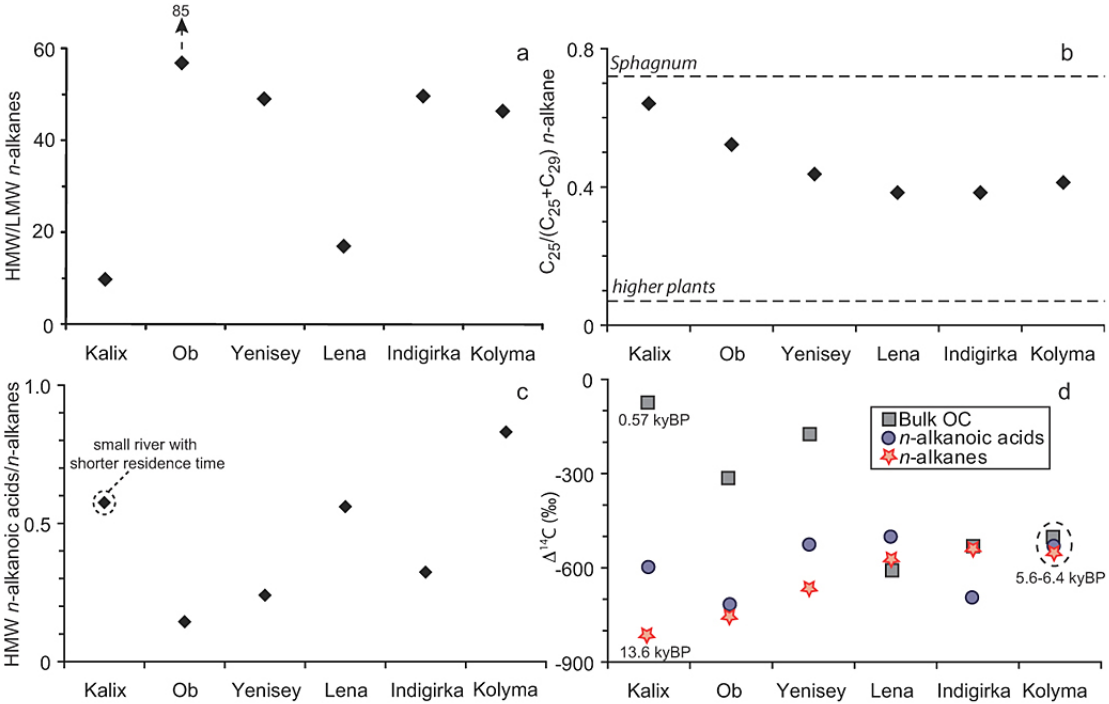

Fig. 2. Molecular and molecular-isotopic patterns of soil marker compounds from west-to-east across the Eurasian Arctic climosequence. **(a)** Ratio of terrigenous-to-aquatic derived *n*-alkanes (HMW *n*-alkanes to LMW *n*-alkanes). **(b)** $C_{25}/(C_{25}+C_{29})$ *n*-alkane ratio indicating contribution from *Sphagnum*-rich peatlands to the terrestrial OC (end-member values in Vonk et al., 2009). **(c)** Ratio of HMW *n*-alkanoic acids to HMW *n*-alkanes indicating an increased contribution of less degraded carbon eastward. **(d)** $\Delta^{14}C$ values of bulk OC, *n*-alkanoic acids ($C_{24}+C_{26}+C_{28}$) and *n*-alkanes ($C_{27}+C_{29}+C_{31}$) from estuarine surface sediments across the Eurasian Arctic. Uncertainties are smaller than symbols (see Table 2).

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Widespread release of old carbon across the Siberian Arctic

Ö. Gustafsson et al.