

**Characterization of
particulate organic
matter – Part 2**

M. Winterfeld and
G. Mollenhauer

Characterization of particulate organic matter in the Lena River Delta and adjacent nearshore zone, NE Siberia – Part 2: Radiocarbon inventories

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Abstract

Particulate organic matter (POM) derived from permafrost soils and transported by the Lena River represents a quantitatively important terrestrial carbon pool exported to Laptev Sea sediments (next to POM derived from coastal erosion). Its fate in a future warming Arctic, i.e. its remobilization and remineralization after permafrost thawing as well as its transport pathways to and sequestration in marine sediments is currently under debate. We present the first radiocarbon (^{14}C) data set of surface water POM within the Lena Delta sampled in summers 2009–2010 and spring 2011 ($n = 30$ samples). The bulk $\Delta^{14}\text{C}$ concentrations varied from -55 to -391 ‰ translating into ^{14}C ages of 395 to 3920 yr BP. We further estimated the fraction of phytoplankton-derived POM to our samples based on (1) particulate organic carbon to particulate nitrogen ratios (POC : PN) and (2) on the stable carbon isotope ($\Delta^{13}\text{C}$) composition of our samples. Assuming that this phytoplankton POM has a modern ^{14}C signature we inferred the ^{14}C concentrations of the soil-derived POM fractions. The results ranged from -258 to -768 ‰ (i.e. 2340 to 11 700 ^{14}C yr BP) for the POC : PN-based scenario and from -191 to -704 ‰ (i.e. 1640 to 9720 ^{14}C yrs BP). Despite the limitations of our approach, the estimated $\Delta^{14}\text{C}$ concentrations of the soil-derived POM fractions seem to reflect the heterogeneous ^{14}C signal of the Lena River catchment soils covering a range from Holocene to Pleistocene ages. We therefore propose a typical isotopic signature of riverine soil-derived POM with a $\Delta^{13}\text{C}$ of -26.6 ± 1.1 ‰ deduced from our data of Lena Delta soils and published values, and a $\Delta^{14}\text{C}$ concentration of -362 ± 123 ‰ deduced from our $\Delta^{13}\text{C}$ -based estimates. These data can help to improve the dual-carbon-isotope simulations used to quantify contributions from riverine soil POM, Pleistocene ice complex POM from coastal erosion, and marine POM in Siberian shelf sediments.

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1 Introduction

Huge amounts of soil organic carbon are currently stored frozen in permafrost soils of the high northern latitudes (e.g. Tarnocai et al., 2009; Zimov et al., 2009) and excluded from biogeochemical cycling. Due to recent observed and projected amplified warming of the Arctic (ACIA, 2005; Serreze et al., 2000) carbon cycling and the fate of organic carbon released from permafrost soils have received growing attention (e.g. Guo et al., 2007; McGuire et al., 2009; Schuur et al., 2008, 2009; Zimov et al., 2006).

Increasing permafrost temperatures, thaw depth in summer (active layer depth), increasing river runoff (Boike et al., 2013; McClelland et al., 2012; Peterson et al., 2002), and increasing length of open water season (Markus et al., 2009) affecting coastal erosion of permafrost deposits will likely lead to enhanced mobilization and export of old, previously frozen organic matter (OM) to the Arctic shelf seas. The understanding of the different terrestrial OM sources (e.g. fresh vegetation, surface soils, Pleistocene ice complex), their age and quality has significantly improved over the last decade (e.g. Guo et al., 2007; Vonk et al., 2010). The use of carbon isotopes ($\delta^{13}\text{C}$, $\Delta^{14}\text{C}$) of dissolved and particulate organic matter as well as individual biomarkers has helped characterizing and distinguishing these different carbon pools, e.g. the old, yet little degraded Pleistocene ice complex-derived OM and comparatively younger and more degraded fluvial OM reaching the Siberian shelf seas (Feng et al., 2013; Guo et al., 2004; Gustafsson et al., 2011; Karlsson et al., 2011; Vonk et al., 2012, 2010).

However, particularly in Siberia ^{14}C data on riverine suspended particulate organic matter (POM) is only very sparsely available. To our knowledge there are no POM ^{14}C concentrations published which were sampled directly in Siberian rivers besides a few samples from the Kolyma River (supplementary data of Vonk et al., 2012). Available POM ^{14}C concentrations for Siberia are from offshore the deltas of the Lena (Karlsson et al., 2011) and Kolyma Rivers (Vonk et al., 2010) and inferred from a sediment core taken in a floodplain lake of the Ob' River (Dickens et al., 2011). Due to settling of POM and marine primary production fueled by the riverine nutrients, the ^{14}C concentrations

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Rachold and Hubberten, 1999; Rachold et al., 2004). Recently, it has been shown that the annual supply of total organic carbon from ice complex deposits along the Laptev Sea coast by erosion is $\sim 0.66 \text{ Tg yr}^{-1}$ (Günther et al., 2013).

The Lena River Delta is the largest arctic delta ($\sim 32000 \text{ km}^2$). It is characterized by a polygonal tundra landscape with active floodplains. Water and sediment discharge are not equally distributed through the several delta channels. Approximately 80–90 % of the total water and up to 85 % of the sediment discharge are delivered through the three main eastern channels to the Buor Khaya Bay east of the delta (Fig. 1b), i.e. through the Sardakhsko-Trofimovskaya channel system (60–75 % water, 70 % sediment) and the Bykovskaya channel (20–25 % water, 15 % sediment). Only a minor portion is discharged to the north and west through the Tumatskaya and Olenyokskaya channels (5–10 % water, 10 % sediment) (Charkin et al., 2011 and references therein; Ivanov and Piskun, 1999).

2.2 Sampling

The sampling sites presented in this study are located in the eastern part of the Lena Delta and adjacent Buor Khaya Bay (Table 1, Fig. 1b) in and along the channels of highest discharge. Permafrost soil samples from the first delta and surface water total suspended matter (TSM) were collected during three expeditions in August 2009, July/August 2010, and in late May and late June/early July 2011. In order to obtain samples that reflect the original state of the frozen permafrost soils, thawed material was removed with a spade for the total height of each bluff. Frozen pieces of peat were excavated at different depths using hatchet and hammer.

Suspended particulate matter of Lena River surface water was sampled at different stations in the main river channels of the delta on the Russian vessel Puteyski 405 (Fig. 1b, Table 1). Between 1 and 30 L of water were filtered on pre-combusted (4.5 h at 450°C) and pre-weighed glass fiber filters (GF/F Whatman, $0.45 \mu\text{m}$ membrane) for particulate organic carbon (POC) and nitrogen (PN) analysis as well as carbon isotope analysis. Additionally, one water sample of 20 L from the spring freshet in 2011 was

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stored cooled in opaque canisters for several days to allow for the suspended matter to settle. Before decanting the supernatant water it was filtered on pre-combusted and pre-weighed GF/F filters to check for the SPM remaining in suspension. For the sample presented here (sample ID 37) the SPM of the supernatant water represented 0.1 % of the settled material on a dry weight basis and therefore the loss of material in suspension can be neglected.

The soil samples were stored in pre-combusted glass jars (4.5 h at 450 °C) and GF/F filters were either stored in pre-combusted petri dishes (\emptyset 47 mm) or wrapped in pre-combusted aluminum foil. All samples were kept frozen at -20°C during storage and transport until analysis.

2.3 Laboratory analyses

Soil and sediment samples were freeze-dried, homogenized, and subsampled for elemental analysis. All filters were oven-dried at 40°C for 24 h.

2.3.1 Elemental analyses

Weight percent organic carbon (OC) and total nitrogen (TN) content of soil samples were determined by high temperature combustion after removal of carbonates as described by Goñi et al. (2003). TSM samples were analyzed for OC and TN at the Alfred Wegener Institute in Bremerhaven, Germany, using the same protocol.

2.3.2 Carbon isotope analysis ($\Delta^{14}\text{C}$, $\delta^{13}\text{C}$)

Samples were radiocarbon-dated at the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility at Woods Hole Oceanographic Institution, USA. Bulk sediment samples and filters with TSM were submitted unprocessed. The Radiocarbon analysis at NOSAMS were carried out using standard methods (McNichol et al., 1994). Results are reported as $\Delta^{14}\text{C}$, conventional radiocarbon ages (years BP), and fraction

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3.2.2 Radiocarbon (^{14}C) composition

The $\Delta^{14}\text{C}$ concentrations of the two soil profiles decreased with depth from -197 to -377‰ for the riverbank profile L09-12 and from -204 to -466‰ for profile L09-28 (Table 3, Fig. 3a). The corresponding ^{14}C ages for both profiles together ranged from 1710 to 4900 yrs BP. Within profile L09-12 on Samoylov Island an age reversal was observed for the two oldest samples (Table 3), which most likely is due to allochthonous material that was transported to the delta from an upstream location. The same age reversal on Samoylov Island was also observed by Kuptsov and Lisitsin (1996). Overall, our $\Delta^{14}\text{C}$ concentrations reflect the late Holocene formation of these soils and fits within the range of ages determined for the Lena Delta first terrace (Bolshiyarov et al., 2014; Kuptsov and Lisitsin, 1996; Schwamborn et al., 2002).

As with other TSM parameters, the POM $\Delta^{14}\text{C}$ concentrations showed strong spatial and interannual variability. Lena Delta POM concentrations varied from -262‰ to -55‰ in August 2009 (mean of -160‰ , $n = 13$), from -391‰ to -143‰ in July/August 2010 (mean of -194‰ , $n = 13$), and from -154‰ to -144‰ in June/July 2011 (mean of -150‰ , $n = 3$). The sample from late May 2011 showed a $\Delta^{14}\text{C}$ concentration of -306‰ and the Buor Khaya Bay surface samples of -176‰ (sample 35) and -143‰ (sample 36, Table 3, Fig. 2r and s). Overall these ^{14}C compositions covered a range of ^{14}C ages from 395 to 3920 yrs BP. The samples with the most depleted $\Delta^{14}\text{C}$ values of -262‰ (sample 1) and -391‰ (sample 31) were taken close to the Pleistocene ice complex deposits of Kurungnakh Island (Fig. 1b), which likely contributed to the local POM in the Lena River surface water.

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4 Discussion

4.1 Origin of organic matter in the Lena Delta

4.1.1 Particulate carbon to nitrogen ratios (POC : PN)

Riverine particulate organic matter consists of a heterogeneous mixture derived from two major sources, i.e. terrestrial (e.g. fresh vegetation and litter, surface and deep soils horizons) and aquatic (phytoplankton/bacterial primary production). The terrestrial OM in the Lena River catchment can further be differentiated into two pools of different age: the late Pleistocene organic-rich ice complex or Yedoma deposits, particularly in the lowlands (0–400 m, e.g. Grosse et al., 2013) and the Holocene permafrost soils. POC : PN ratios as well as $\delta^{13}\text{C}$ and ^{14}C signatures of bulk OM can be used to estimate terrestrial and aquatic contributions (e.g. Hedges and Oades, 1997). However, due to overlaps in soil/plant and algal/bacterial signatures it might be difficult to unambiguously differentiate between terrestrial and aquatic sources.

Our POC : PN ratios from the summers 2009 and 2010 as well as spring 2011 vary largely throughout the delta (Table 2, Fig. S2) similar to the TSM and lignin phenol concentrations sampled during the same field trips (Winterfeld et al., 2014). The ratios range from 3.7 to 19.3 for all samples and seasons with mean atomic POC : PN ratios of 9.6 (2009), 7.6 (2010), and 7.8 (2011). Low POC : PN ratios, i.e. ~ 6 generally indicate high contributions from phytoplankton and or⁻¹ bacterial primary production while ratios > 20 are indicative of soil and plant contributions (e.g. Hedges et al., 1997; Meyers and Lallier-vergés, 1999). Based on these end-members, our data from the summer and spring seasons suggest a considerable fraction of primary production OM to be present in our samples. Using a simple two end-member mixing model with POC : PN = 6 for primary production OM and 20.2 for soil-derived OM mean of OC : TN ratios from the first delta terrace of this study and delta soils as well as tundra and taiga soils south of the delta from Zubrzycki (2013), our soil fractions vary from 0.06 to 0.94 (mean 0.26)

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in 2009, from 0.10 to 0.30 (mean 0.16) in 2010, and from 0.08 to 0.26 (mean 0.14) in 2011.

The total flux of primary production OM in the Lena River is thought to be negligible due to low light penetration in the turbid waters (Cauwet and Sidorov, 1996; Sorokin and Sorokin, 1996). Yet, the surface water layer from which we took our TSM samples is characterized by an abundance of riverine plankton (Kraberg et al., 2013; Sorokin and Sorokin, 1996), which could explain the small soil-derived OM fraction.

However, due to possible sorption of inorganic nitrogen to clay minerals (Schubert and Calvert, 2001) our calculated POC : PN ratios might be too low, underestimating the soil-derived OM fraction. This will be discussed in more detail in Sect. 4.3.1. Another possibility for lower, but mainly soil derived POC : PN ratios could be the selective degradation of labile OM compared to total nitrogen (Kuhry and Vitt, 1996).

4.1.2 Stable carbon isotopes ($\delta^{13}\text{C}$)

The $\delta^{13}\text{C}_{\text{POM}}$ composition of our samples taken in 2009–2011 varies over a broad range (–34.2 to –26.5‰, Table 3), which is similar to $\delta^{13}\text{C}_{\text{POM}}$ values previously published by Rachold and Hubberten (1999) for the Lena Delta and Lena River upstream the delta (–31.3 to –25.7‰, July/August 1994/95), by ArcticGRO (www.arcticgreativers.org) ~ 900 km upstream the Lena Delta at Zhigansk (–30.3 to –25.2‰, May–August 2004–2010), and by Semiletov et al. (2011) for the Lena Delta and Lena River (–30.0 to –25.0‰, August–September 1995–2008).

In general, the more enriched $\delta^{13}\text{C}_{\text{POM}}$ values (around –27‰) reflect the dominant contribution from C3 plants and soils (e.g. Hedges et al., 1997) from the river catchment. The more depleted $\delta^{13}\text{C}_{\text{POM}}$ values (< –29‰) point to mixing with riverine plankton utilizing dissolved inorganic carbon (DIC) with depleted $\delta^{13}\text{C}$ signatures as suggested for the Lena River (Alling et al., 2012; Rachold and Hubberten, 1999). Analogous to the POC : PN ratios, the $\delta^{13}\text{C}_{\text{POM}}$ compositions point to a considerable contribution of primary production OM to our samples from 2009–2011. Possible end-

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member calculation and implications for the ^{14}C concentration of the POM samples will be further discussed below in Sect. 4.3.2.

4.2 Radiocarbon composition of POM ($^{14}\text{C}_{\text{POM}}$) in the Lena Delta

Terrestrial POM enters a river predominantly by physical weathering of adjacent soils (Raymond and Bauer, 2001 and references therein). Compared to vegetation utilizing modern atmospheric $^{14}\text{CO}_2$, the bulk POM of soil is pre-aged. The specific residence time of POM in the soil before entering the river depends on various environmental factors like humidity, temperature, topography, soil type, size and topography of the catchment area (e.g. Kusch et al., 2010; Oades, 1988; Raymond and Bauer, 2001; Trumbore, 2009, 1993). Furthermore, the fluvial transport of POM (and TSM) is governed by hydrological characteristics like runoff, discharge and flow velocity, sedimentation along riverbanks and floodplains as well as resuspension of deposited material. Therefore, the age of terrestrial POM is a combination of its residence time within the soil plus its residence time within the river basin, which can differ substantially for different terrestrial POM fractions (lipids vs. lignin) from arctic watersheds (e.g. Feng et al., 2013; Gustafsson et al., 2011). In permafrost affected watersheds with soils storing up to Pleistocene aged OM, the residence time within the watershed after entering the river might have only a minor effect.

The Lena Delta and Buor Khaya Bay surface water $^{14}\text{C}_{\text{POM}}$ concentrations presented here are depleted with respect to the current atmospheric $^{14}\text{CO}_2$ (Table 3). The $\Delta^{14}\text{C}$ concentrations ranged from -391‰ to -115‰ for 2009 to 2011 translating into ^{14}C ages > 1200 yrs and up to 3920 yrs BP for samples taken close to the Pleistocene ice complex deposits of Kurungnakh Island (Figs. 1b and 2q–t, Table 3). These results are within the range of reported $\Delta^{14}\text{C}$ concentrations for surface water POM offshore the Lena Delta and influenced by Lena River outflow (Karlsson et al., 2011). Based on these results it seems reasonable to assume that a large fraction of Lena Delta POM

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originates from physical weathering of relatively young Holocene soils (active layer) of the first delta terrace and south of the delta.

However, soils outcropping along the Lena River south of the delta can be substantially older than late Holocene ages (Kuptsov and Lisitsin, 1996) including some areas of Pleistocene ice complex deposits (Grosse et al., 2013). Also, OM within the active layer and shallow permafrost table can be as old as 3000 yrs BP in the Lena Delta 30 cm below surface, (Höfle et al., 2013) or more than > 10000 yrs BP south of the delta (Kuptsov and Lisitsin, 1996). As shown by the lignin phenol composition of Lena Delta POM (Winterfeld et al., 2014) approximately half of the surface water particulate organic matter is derived from the catchment south of the delta with considerable contributions from delta soils, particularly in the summer season when riverbank erosion is strongest. Considering that, we would expect generally older POM ^{14}C ages. Additionally, riverbank erosion contributes POM covering the ^{14}C age range of the whole bluff profile, not only from the active layer.

One possible explanation for our relatively young POM ^{14}C ages could be the contribution of algal-derived OM with a rather modern ^{14}C signature concealing the “true” age of soil-derived OM. Algal-derived OM contribution was also suggested to be the reason for relatively young POM from the Ob’ River inferred from a core of a floodplain lake (Dickens et al., 2011). Although the overall contribution and flux of autochthonous phytoplankton OM in the Lena River is rather small or even negligible due to the high turbidity and low light penetration (Cauwet and Sidorov, 1996; Sorokin and Sorokin, 1996), phytoplankton can be quite abundant in the surface water (Kraberg et al., 2013; Sorokin and Sorokin, 1996), which we sampled for our POM analyses. However, without directly determining the phytoplankton OM by microscopic counts or determining the chlorophyll-*a* content for each radiocarbon-dated sample, indirect evaluations have to be considered estimates. In the following chapter we describe three different scenarios for possible ^{14}C corrections of POM in the Lena Delta to assess the potential range of $\Delta^{14}\text{C}$ concentrations and ^{14}C ages of soil-derived particles. In these scenarios we assume that the algal-derived OM is of modern ^{14}C age ($\Delta^{14}\text{C} \sim 49\%$ and)

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although this might not be true. DIC $\Delta^{14}\text{C}$ concentrations are not available for the Lena River. The DIC utilized by the phytoplankton is predominantly derived from the carbonate weathering within the Lena watershed and from soils, both, providing a depleted ^{14}C carbon signature and only slowly exchanging with the atmosphere. Therefore, the following calculated $\Delta^{14}\text{C}$ concentrations of surface water POM from the Lena River have to be considered minimum, i.e. maximum ^{14}C ages.

4.3 ^{14}C estimates of soil-derived POM for algal OM contribution

4.3.1 Scenario 1: Soil-derived OM ^{14}C estimates based on POC : PN ratios

POC : PN ratios around 6 are usually associated with OM derived from algal/bacterial primary production and higher ratios of > 20 with OM derived from soils (Hedges et al., 1997; Meyers and Lallier-vergés, 1999 and references therein). However, adsorption of inorganic nitrogen derived from OM decomposition (e.g. ammonium) to clay minerals (Schubert and Calvert, 2001), which is not accounted for when determining the total PN content, may additionally affect the POC : PN ratio in TSM leading to lower values than would be expected from mixing of the two end-members alone. Likewise, selective degradation of labile organic carbon (Kuhry and Vitt, 1996) would have the same effect. Both processes would lead to low estimates of the soil-derived OM contribution and suggest a higher contribution from plankton. Both explanations were also suggested by Sánchez-García et al. (2011) for POM in the Laptev Sea offshore the Lena Delta with unusually low POC : PN ratios.

In order to estimate the inorganic nitrogen content for our sample sets from each year, we used the intercept of the POC vs. PN content regression line at zero POC (Fig. S2 Supplement). By subtracting these amounts from the analyzed total PN content of the respective samples, we could calculate soil POC : PN ratios (POC : PN) for our samples (Table S2 Supplement). Based on these POC : PN_{NEW} ratios, we calculated the soil-derived fraction within our POM samples using a simple two end-member

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mixing model as following:

$$\text{POC} : \text{PN}_{\text{POM}} = f_{\text{soil}} \times \text{POC} : \text{PN}_{\text{soil}} + f_{\text{plankton}} \times \text{POC} : \text{PN}_{\text{plankton}} \quad (1)$$

and

$$1 = f_{\text{soil}} + f_{\text{plankton}} \quad (2)$$

where $\text{POC} : \text{PN}_{\text{POM}}$ is the measured value in the POM sample (Table S1). As the end-member for $\text{POC} : \text{PN}_{\text{soil}}$ we assumed a value of 20.2, which is an average calculated from Lena Delta first terrace soils presented in Winterfeld et al. (2014) and delta soils as well as soil from the Lena River watershed covering the taiga to tundra transition from Zubrzycki (2013). The $\text{POC} : \text{PN}_{\text{plankton}}$ endmember value is 6 (e.g. Meyers, 1994 and references therein).

The calculated soil-derived OM fractions of Lena Delta POM varied from 0.28 to 0.55 (mean of 0.41, $n = 11$) for summer 2009, from 0.26 to 0.51 (mean of 0.43, $n = 10$) in summer 2010, and from 0.48 to 0.69 (mean of 0.54, $n = 3$) for late June/early July 2011 (Table 3). We used the POC vs. PN (wt%) regression line from the summer 2010 samples within the delta to correct the two samples taken outside of the delta in the Buor Khaya Bay. They had calculated soil fractions of 0.43 (sample 35) and 0.03 (sample 36). The same was done for the single sample from late May 2011. We used the regression line from the samples taken 4 weeks later in June/July and the soil fraction of sample 37 was 0.25 (Table 3). Note that these soil- and plankton-derived OM fractions can only serve as rough estimates. Without determining the particulate organic nitrogen directly for every sample our $\text{POC} : \text{PN}_{\text{NEW}}$ ratios might be highly over- and underestimating OM fractions in individual samples.

The so calculated soil and plankton OM fractions (f_{soil} and f_{plankton} , Table 3 Supplement) were further used in an isotopic mass balance to determine the $\Delta^{14}\text{C}$ concentration of the soil fraction assuming the plankton-derived OM is modern ($\Delta^{14}\text{C}_{\text{plankton}} = 49\text{‰}$):

$$f_{\text{POM}} \times \Delta^{14}\text{C}_{\text{POM}} = f_{\text{soil}} \times \Delta^{14}\text{C}_{\text{soil}} + f_{\text{plankton}} \times \Delta^{14}\text{C}_{\text{plankton}} \quad (3)$$

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and

$$f_{\text{POM}} = f_{\text{soil}} + f_{\text{plankton}} \quad (4)$$

where f_{POM} , f_{soil} , and f_{plankton} are the fractions of POM, soil- and plankton-derived OM and $\Delta^{14}\text{C}_{\text{POM}}$, $\Delta^{14}\text{C}_{\text{soil}}$, and $\Delta^{14}\text{C}_{\text{plankton}}$ are the $\Delta^{14}\text{C}$ concentrations of these sources. As mentioned above, 49‰ is assumed as a maximum estimate for $\Delta^{14}\text{C}_{\text{plankton}}$.

The results of the newly calculated $\Delta^{14}\text{C}_{\text{soil}}$ concentrations using POC:PN_{corr} ratios to partition between soil- and plankton-derived OM are shown in Table 4. The soil $\Delta^{14}\text{C}_{\text{POC:PN}}$ concentrations range from -786‰ to -258‰ for the sampling period of 2009–2011 translating into $^{14}\text{C}_{\text{soil}}$ ages > 2300 yrs BP with an average of 6200 yrs BP (Table 4). Analogous to the two comparatively old $^{14}\text{C}_{\text{POM}}$ ages off Kurungnakh Island taken in 2009 and 2010 (Table 3), the calculated $^{14}\text{C}_{\text{POC:PN}}$ ages for these samples were with ~ 11 600 yrs BP the oldest. Considering that the riverbank outcrops of Kurungnakh Island cover an age range of approximately 100 kyrs with organic-rich ice complex deposits of about 50 kyrs (Wetterich et al., 2008) these ^{14}C ages seem to be realistic.

Again, the uncertainties associated with the contribution of inorganic nitrogen to our total PN contents are rather high resulting in a relatively rough estimation soil and plankton OM fractions. These uncertainties are further affecting the calculation of the soil $\Delta^{14}\text{C}_{\text{POC:PN}}$ concentrations. That means the calculated ^{14}C ages are estimates. Yet, they demonstrate that a possible underestimation of soil-derived OM ages can be considerable.

4.3.2 Scenario 2: Soil-derived OM ^{14}C estimates based on $\delta^{13}\text{C}$

Similar to the correction approach discussed above, we used a second scenario based on $\delta^{13}\text{C}$ compositions to distinguish between soil- and plankton-derived OM in our Lena Delta POM samples. The vegetation in the Lena River catchment (taiga and

tundra) is dominated by C3 plants with a $\delta^{13}\text{C}$ of around -25% to -27% (Rachold and Hubberten, 1999) also reflected in our $\delta^{13}\text{C}$ data from the first delta terrace soils with an average of -26.2% ($n = 7$, Table 3). Bird et al. (2002) determined the $\delta^{13}\text{C}$ composition of taiga and tundra soils (excluding peatlands) along Yenisey River on a latitudinal transect. Their data suggests an average $\delta^{13}\text{C}$ concentration of -26.9% for tundra and taiga soils combined, which we use as the soil OM end-member in our two end-member model.

The $\delta^{13}\text{C}$ composition of riverine plankton POM depends on the fractionation between phytoplankton and dissolved inorganic carbon (DIC). The distribution of the different DIC fractions dissolved CO_2 , bicarbonate (HCO_3^-), and carbonate ion (CO_3^{2-}) varies depending on temperature and pH. In the lower reaches of the Lena River and the Lena Delta $> 90\%$ of the DIC are made up of bicarbonate (Alling et al., 2012), i.e. the $\delta^{13}\text{C}$ of bicarbonate represents the $\delta^{13}\text{C}$ of DIC. Sources for DIC are generally the CO_2 derived from soil OM degradation and CO_2 released during the dissolution of carbonates. The Lena River geochemistry is mainly influenced by carbonate weathering and groundwater in the summer season (Gordeev and Sidorov, 1993). Assuming a fractionation of -25% between phytoplankton and DIC these high latitude waters and a $\delta^{13}\text{C}_{\text{DIC}}$ of -8% for the Lena Delta (Alling et al., 2012) a plankton $\delta^{13}\text{C}$ end-member of -33% would be expected. Similar or more depleted $\delta^{13}\text{C}$ values of bicarbonate and phytoplankton POM were also determined in the Yenisey and Ob' Rivers (Galimov et al., 2006) and in temperate estuaries (e.g. Ahad et al., 2008; Chanton and Lewis, 2002). Our most depleted $\delta^{13}\text{C}_{\text{POM}}$ of -34.2% for a sample from the summer 2009 (Table 3) is even lower than the plankton end-member used here ($\delta^{13}\text{C}_{\text{plankton}} = -33\%$). The sample location is offshore Muostakh Island (Fig. 1b) and influenced by mixing with marine waters, which complicates the DIC composition and processes affecting $\delta^{13}\text{C}$ of DIC (Alling et al., 2012).

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We used the following two end-member model to estimate the soil- and plankton-derived OM fractions (f_{soil} , f_{plankton}) to our POM samples:

$$\delta^{13}\text{C}_{\text{POM}} = f_{\text{soil}} \times \delta^{13}\text{C}_{\text{soil}} + f_{\text{plankton}} \times \delta^{13}\text{C}_{\text{plankton}} \quad (5)$$

5 and

$$1 = f_{\text{soil}} + f_{\text{plankton}} \quad (6)$$

where $\delta^{13}\text{C}_{\text{POM}}$ is the analyzed $\delta^{13}\text{C}$ composition of our POM samples (Table 3), $\delta^{13}\text{C}_{\text{soil}}$ is the soil end-member value of -26.9‰ , and $\delta^{13}\text{C}_{\text{plankton}}$ is plankton end-member value of -33‰ .

The calculated soil OM fractions varied from 0.07 to 0.68 in summer 2009 (mean of 0.44, $n = 11$), from 0.47 to 0.77 in summer 2010 (mean of 0.62, $n = 10$), and from 0.60 to 0.76 in spring 2011 (mean of 0.70, $n = 3$). The Buor Khaya Bay POM sample 21 showed a soil fraction of 0.45. Two samples are not considered in this end-member model, because their $\delta^{13}\text{C}$ values are outside the range of the end-member values chosen here (sample 37 and 19, see Table 3). This illustrates the limitations of our approach. Constraining a model with appropriate end-members strongly influences the quality of the model output and here we were not able to account for the whole range of natural variability observed in the Lena River. As discussed above in Sect. 4.3.1, these soil OM fractions are rough estimates in the absence of direct plankton determination.

The $\Delta^{14}\text{C}$ concentrations are further corrected for the contribution of modern plankton OM as described in Sect. 4.3.1 using the Eqs. (3) and (4). Because the soil contributions calculated in this scenario are slightly higher than in the POC:PN-based scenario, the $\Delta^{14}\text{C}$ calculations resulted in less radiocarbon depleted estimates compared to the POC:PN scenario (Table 4). Nonetheless, the $\Delta^{14}\text{C}$ concentrations based on the $\delta^{13}\text{C}$ end-member model are considerably ^{14}C -depleted compared to bulk POM $\Delta^{14}\text{C}$ concentrations. The $\Delta^{14}\text{C}_{\text{corr}}$ concentrations range from -704‰ to -191‰ for all samples 2009–2011 representing ^{14}C ages from 9720 to 1640 yrs BP (Table 4).

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nitions for ice complex deposits and marine primary production are rather well constrained. In contrast, the soil/topsoil end-member is more difficult to define, particularly when using indirect parameters such as riverine DOM and POM. Here our first terrace soil data and estimates for soil-derived POM (Table 4) provide new $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ concentration ranges (Fig. 3b) for fluvially exported soil POM. Together with published $\delta^{13}\text{C}$ concentrations from tundra and taiga soils in Siberia and Alaska (Bird et al., 2002; Pitkänen et al., 2002; Xu et al., 2009) we can define a Lena River soil OM end-member with a $\delta^{13}\text{C}$ value of $-26.6 \pm 1.1\text{‰}$. The corresponding $\Delta^{14}\text{C}$ concentration is $-362 \pm 123\text{‰}$ is taken from the second scenario discussed above (Table 4). The $\delta^{13}\text{C}$ -based scenario is favored here over the POC:PN-based scenario, because it allows to calculate the soil fraction of each POM sample based on its bulk $\delta^{13}\text{C}$ composition, which is a mixture of soil and phytoplankton OM. In contrast, in the POC:PN-based scenario a constant value for particulate inorganic nitrogen is subtracted, which does not necessarily represent the actual inorganic nitrogen content of the particular sample. Moreover, it does not account for selective degradation of labile carbon, which could result in overestimating the phytoplankton contribution. The uncertainties associated with the $\delta^{13}\text{C}$ -based estimates are smaller, as the $\delta^{13}\text{C}$ end-member for soil-derived and phytoplankton-derived OM are fairly well constrained (see also Sect. 4.3.2).

However, our soil OM end-member for the Lena River catchment makes it more complicated to distinguish between soil/top-soil derived OM from the river and ice complex deposits from coastal erosion (see Fig. 3b). The $\delta^{13}\text{C}$ signatures of both end-members are almost indistinguishable. The $\Delta^{14}\text{C}$ range of our soil-derived POM estimates is more depleted than the end-member used by Karlsson et al. (2011) and Vonk et al. (2012, 2010). Furthermore, Höfle et al. (2013) have shown that OM within the first 30 cm of a polygon rim of the Lena Delta first terrace can be 3000 ^{14}C yrs old, which make the end-members chosen for fluvial exported soil/top-soil OM by Karlsson et al. (2011) and Vonk et al. (2012, 2010) appear too young. Using the bulk surface water POM and DOM $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ compositions as end-member could therefore highly over- or underestimate the soil OM contribution from permafrost watersheds

and in turn highly over- or underestimate OM contribution from ice complex to marine sediments.

However, we are aware of the limitations and uncertainties associated with the soil $\Delta^{14}\text{C}$ estimates discussed above. Without determining the phytoplankton biomass of each sample by microscopic counting or from chlorophyll *a* analysis, the plankton OM fraction calculated based on POC:PN ratios (Sect. 4.3.1) and based $\delta^{13}\text{C}$ composition (Sect. 4.3.2) can only be regarded as estimates. These estimates give orders of magnitude of OM contribution from the individual sources rather than exact values, which in turn provide a possible range of $\Delta^{14}\text{C}$ concentrations for soil-derived POM. An additional source of uncertainty is our assumption of a modern $\Delta^{14}\text{C}$ concentration of plankton OM. Without determining the $\Delta^{14}\text{C}$ concentration of the Lena River DIC, which is utilized by phytoplankton, we cannot be sure of a modern ^{14}C signature. Lena River geochemistry and DIC are dominated by dissolution of carbonates in the watershed contributing fossil ^{14}C to the DIC pool, leading likely to ^{14}C -depletion in river phytoplankton. The true contribution of modern phytoplankton OM is thus likely smaller than estimated above. Consequently, the soil-derived POM would be less ^{14}C depleted than calculated here. Therefore, our estimated ^{14}C soil ages have to be considered maximum ages.

The best way to determine the $\Delta^{14}\text{C}$ concentration of riverine soil-derived OM would be a biomarker-specific radiocarbon analysis using source-specific compounds, e.g., short- and long-chain fatty acids for plankton- and terrestrial-derived OM, respectively. However, for these analyses large samples of POM are needed. The samples analyzed during our study were too small to allow for compound-specific dating.

5 Conclusions

There is only scarce data available on ^{14}C contents of POM from Lena River surface water, but within the likely positive carbon-climate feedback to greenhouse warming the quality and fate of this permafrost OM pool in the coastal waters of the Laptev Sea is

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currently under debate (e.g. Feng et al., 2013; Gustafsson et al., 2011; Karlsson et al., 2011; Vonk et al., 2012). With this study we provide the first data set on surface water POM $\Delta^{14}\text{C}$ concentrations from the Lena Delta sampled during the summers 2009 and 2010 and during spring 2011 ($n = 30$ samples). The contribution of modern phytoplankton POM to these samples was estimated based on POC : PN ratios and $\delta^{13}\text{C}$ values to allow for calculation of the $\Delta^{14}\text{C}$ concentrations of the soil-derived POM fraction. These soil $\Delta^{14}\text{C}$ estimates are depleted compared to the bulk POM $\Delta^{14}\text{C}$ concentrations and therefore seem to represent the heterogeneous ^{14}C mix of soil OM ranging from Holocene to Pleistocene age (e.g. ice complex deposits) in the Lena River watershed more accurately. Because of the limitations of our approach, particularly the assumption of modern phytoplankton OM without determining the $\Delta^{14}\text{C}$ concentration of the Lena River DIC utilized by phytoplankton, our ^{14}C estimates for the soil-derived fraction have to be considered minimum $\Delta^{14}\text{C}$ concentrations and maximum ^{14}C ages, respectively. Nonetheless, we propose average values for soil POM isotopic composition based on our data and published values ($\delta^{13}\text{C} = -26.6 \pm 1.1 \text{‰}$; $\Delta^{14}\text{C} = -362 \pm 123 \text{‰}$), which will be useful for dual-carbon-isotope simulations focusing on unraveling the OM contributed by different terrigenous (fluvial vs. coastal erosion) and marine sources to arctic sediments.

The complete data set presented here can also be found in PANGAEA (www.pangaea.de).

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Table 1. Samples presented in this study analyzed for stable and radiocarbon isotope composition. Information on additional samples analyzed for organic carbon content can be found in Winterfeld et al. (2014, submitted as companion paper) in Tables 1 and S1. Bluff height is given in meters above river level (m a.r.l.) measured in August 2009. Not applicable denoted by n.a.

Sample code	Sample and site description	Date of sampling	Latitude (dec)	Longitude (dec)	Bluff height (m a.r.l.)	Water depth (m)
L09-12	Samoylov Island, 5 depths sampled	18 Aug 2009	72.3775	126.4954	7.5	n.a.
L09-28-2	Bykovskaya Channel, 2 depths sampled	21 Aug 2009	72.0586	128.6309	1.7	n.a.
1	Olenyokskaya Channel	14 Aug 2009	72.4772	125.2856	n.a.	0.5
2	Olenyokskaya Channel	14 Aug 2009	72.3598	125.6728	n.a.	0.5
3	Lena River main channel	16 Aug 2009	72.1526	126.9160	n.a.	0.5
4	Lena River main channel south of Tit Ari Island	16 Aug 2009	71.9040	127.2544	n.a.	0.5
5	Sardakhskaya/Trofimovskaya Channel	17 Aug 2009	72.5825	127.1891	n.a.	0.5
6	Sardakhskaya Channel	17 Aug 2009	72.7002	127.4930	n.a.	0.5
10	Lena River main channel	19 Aug 2009	72.2760	126.9041	n.a.	0.5
11	Lena River main channel	19 Aug 2009	72.5159	126.7142	n.a.	0.5
12	Bykovskaya Channel	20 Aug 2009	72.4140	126.9124	n.a.	0.5
13	Lena River Bykovskaya Channel	20 Aug 2009	72.2352	127.9619	n.a.	0.5
14	Lena River Bykovskaya Channel	20 Aug 2009	72.0341	128.5232	n.a.	0.5
19	NE of Muostakh Island	22 Aug 2010	71.7062	130.2900	n.a.	0.5
21	close to Muostakh Island shoreline	23 Aug 2010	71.5750	129.8200	n.a.	0.5
22	close to Samoylov Island	30 Jul 2010	72.3650	126.4628	n.a.	0.5
24	Sardakhskaya/Trofimovskaya Channel	31 Jul 2010	72.5343	126.8794	n.a.	0.5
25	Lena River Trofimoskaya Channel	31 Jul 2010	72.4764	126.6250	n.a.	0.5
26	Lena River Trofimoskaya Channel	31 Jul 2010	72.4764	126.8588	n.a.	0.5
27	Lena River main channel south of Samoylov	1 Aug 2010	72.3776	126.7478	n.a.	0.5
28	Lena River main channel north of Tit Ari Island	1 Aug 2010	72.2102	126.9423	n.a.	0.5
29	Lena River main channel south of Tit Ari Island	1 Aug 2010	71.9514	127.2582	n.a.	0.5
30	Lena River main channel off Kurungnakh	2 Aug 2010	72.2808	126.2091	n.a.	0.5
31	Lena River main channel	2 Aug 2010	72.3567	126.3521	n.a.	0.5
32	Lena River Bykovskaya Channel	3 Aug 2010	72.3604	127.6761	n.a.	0.5
33	Bykovskaya Channel	3 Aug 2010	72.3604	127.6765	n.a.	0.5
35	offshore Lena Delta	6 Aug 2010	72.0379	129.7694	n.a.	0.5
36	offshore Lena Delta	6 Aug 2010	72.2753	129.8248	n.a.	0.5
37	Lena River main channel off Samoylov Island	29 May 2011	72.3651	126.4757	n.a.	0.5
38	Lena River main channel, south of Stolb	26 Jun 2011	72.3705	126.6538	n.a.	0.5
42	close to Samoylov Island	30 Jun 2011	72.3681	126.4738	n.a.	0.5
47	close to Samoylov Island	2 Jul 2011	72.3681	126.4738	n.a.	0.5

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Table 2. Particulate organic carbon (POC) contents in Lena Delta surface water (2009 to 2011) given in milligram per liter (mg L^{-1}) and percent based on sediment dry weight (wt%) as well as atomic particulate organic carbon (POC) to particulate total nitrogen (PN) ratios. Data on individual samples can be found in the Supplement (Table S2).

	POC (mg L^{-1})	POC (wt%)	atomic POC : PN
Lena Delta Aug 2009	$n = 21$	$n = 20$	$n = 21$
mean	1.21	7.2	9.6
median	0.83	4.7	9.2
min	0.35	1.9	6.8
max	7.24	37.7	19.3
Lena Delta Jul/Aug 2010	$n = 13$	$n = 13$	$n = 13$
mean	0.57	3.05	7.6
median	0.47	3.25	3.05
min	0.15	1.42	3.7
max	1.30	4.74	10.3
Lena Delta late May 2011 sample code: 37	8.20	1.66	7.5
Lena Delta late Jun/early Jul 2011	$n = 9$	$n = 9$	$n = 9$
mean	0.74	4.32	7.8
median	0.69	4.61	7.8
min	0.29	3.20	5.9
max	1.51	4.99	9.7

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Table 3. Stable carbon isotope ($\delta^{13}\text{C}$) and radiocarbon composition (^{14}C) of Lena Delta first terrace bluff profiles and surface water particulate organic matter (2009–2011). Bluff profile samples are given in meters below bluff surface (m b.s.).

Sample code	$\delta^{13}\text{C}$ (‰ VPDB)	fMC ^a	1 σ fMC	$\Delta^{14}\text{C}$ (‰)	conv. ^{14}C age ^b (yrs BP)	1 $\sigma^{14}\text{C}$ age (yrs BP)	Lab ID
Lena Delta first terrace							
L09-12, 0.45 m.b.s.	-26.8	0.8084	0.0039	-197	1710	40	OS-84097
L09-12, 1.35 m.b.s.	-26.3	0.7311	0.0029	-274	2510	30	OS-84073
L09-12, 2.50 m.b.s.	-25.2	0.7023	0.0025	-303	2840	30	OS-84072
L09-12, 4.70 m.b.s.	-27.0	0.5708	0.0024	-433	4500	35	OS-84071
L09-12, 5.80 m.b.s.	-25.1	0.6275	0.0027	-377	3740	35	OS-84070
L09-28, 0.30 m.b.s.	-26.1	0.8015	0.0026	-204	1780	25	OS-84074
L09-28, 0.70 m.b.s.	-26.6	0.5430	0.0028	-461	4900	40	OS-84087
Lena Delta Aug 2009							
1	-30.5	0.7436	0.0029	-262	2380	30	OS-84096
2	-32.6	0.8173	0.0034	-189	1620	35	OS-84090
3	-30.9	0.8735	0.0031	-133	1090	30	OS-84093
4	-29.6	0.8259	0.0029	-180	1540	25	OS-84091
5	-31.3	0.8717	0.0031	-134	1100	30	OS-84098
6	-30.5	0.8524	0.0032	-154	1280	30	OS-84127
10	-29.8	0.8419	0.0032	-164	1380	30	OS-84101
11	-28.9	0.8458	0.0031	-160	1340	30	OS-84100
12	-30.6	0.8913	0.0031	-115	925	25	OS-84102
13	-29.9	0.8672	0.0036	-139	1140	30	OS-84133
14	-28.8	0.7971	0.0031	-209	1820	30	OS-84099
19	-34.2	0.9522	0.0042	-65	395	35	OS-84086
21	-27.1	0.8210	0.0028	-185	1580	25	OS-84088
mean	-30.4	0.8462	0.0032	-160	1353	30	
standard deviation	1.7	0.0480	0.0003	48	457	3	
Lena Delta TSM Jul/Aug 2010							
22	-29.7	0.8344	0.0029	-172	1450	30	OS-95088
24	-29.4	0.8201	0.0034	-186	1590	35	OS-95100
25	-28.3	0.8288	0.0030	-177	1510	30	OS-95266
26	-28.9	0.8001	0.0028	-206	1790	30	OS-95382
27	-28.9	0.8386	0.0034	-167	1410	30	OS-95268
28	-28.8	0.8046	0.0035	-201	1750	35	OS-95380
29	-28.9	0.8353	0.0117	-171	1440	110	OS-94853
30	-30.2	0.8389	0.0033	-167	1410	30	OS-95238
31	-28.6	0.6139	0.0021	-391	3920	25	OS-95239
32	-29.6	0.8390	0.0033	-167	1410	30	OS-95267
33	-29.8	0.8125	0.0144	-193	1670	140	OS-94857
35	n.d.	0.8299	0.0035	-176	1500	35	OS-95377
36	-30.4	0.8628	0.0150	-143	1180	140	OS-94858
mean	-29.3	0.8122	0.0056	-194	1695	54	
standard deviation	0.6	0.0594	0.0045	59	661	42	
Lena Delta TSM late May 2011							
37	-26.5	0.6988	0.0028	-306	2880	30	OS-94760
Lena Delta TSM late Jun/early Jul 2011							
38	-28.3	0.8623	0.0030	-144	1190	30	OS-95378
42	-29.3	0.8558	0.0033	-151	1250	30	OS-95384
47	-28.5	0.8519	0.0117	-154	1290	110	OS-94854
mean	-28.7	0.8567	0.0060	-150	1243	57	
standard deviation	0.4	0.0043	0.0040	4	41	38	

^a fMC = fraction modern carbon, ^b conv. age = conventional radiocarbon age in years before present (yrs BP), i.e. before 1950.

Table 4. Estimated ^{14}C concentrations of soil-derived POM fraction using POC : PN ratios and $\delta^{13}\text{C}$ ratios to estimate amount of soil-derived fraction. Not determined denoted by n.d.

Sample code	POC : PN based estimates			plankton $\delta^{13}\text{C}$ -based estimates		
	Fraction soil POM _{POC:PN}	$\Delta^{14}\text{C}_{\text{POC:PN}}$ (‰)	conv. $^{14}\text{C}_{\text{POC:PN}}$ age (yrs BP)	Fraction soil POM _{$\delta^{13}\text{C}$}	$\Delta^{14}\text{C}_{\delta^{13}\text{C}}$ (‰)	conv. $^{14}\text{C}_{\delta^{13}\text{C}}$ age (yrs BP)
Lena Delta Aug 2009						
1	0.38	-768	11 700	0.41	-704	9720
2	0.36	-604	7380	0.07	n.d.	n.d.
3	0.35	-475	5120	0.34	-480	5190
4	0.28	-757	11 300	0.55	-367	3620
5	0.36	-467	5000	0.29	-590	7140
6	0.43	-427	4420	0.41	-450	4750
10	0.46	-417	4270	0.52	-358	3500
11	0.36	-526	5930	0.68	-260	2360
12	0.53	-258	2340	0.40	-365	3590
13	0.55	-292	2720	0.51	-319	3030
14	0.39	-607	7450	0.68	-328	3140
19	0.29	-306	2870	n.d.	n.d.	n.d.
21	0.34	-634	8020	0.98	-191	1640
Lena Delta Jul/Aug 2010						
22	0.31	-670	8840	0.53	-364	3580
24	0.31	-707	9810	0.59	-351	3420
25	0.43	-474	5100	0.77	-243	2180
27	0.51	-378	3760	0.67	-276	2540
28	0.46	-491	5360	0.69	-312	2950
29	0.38	-523	5890	0.68	-276	2530
30	0.26	-768	11 700	0.47	-416	4260
31	0.33	n.d.	n.d.	0.72	-559	6520
32	0.37	-541	6190	0.56	-340	3280
33	0.34	-672	8890	0.52	-416	4270
35	0.43	-480	5190	n.d.	n.d.	n.d.
36	0.03	n.d.	n.d.	0.45	-377	3740
late May 2011						
37	0.25	n.d.	n.d.	n.d.	n.d.	n.d.
late Jun/early Jul 2011						
38	0.54	-306	2870	0.76	-204	1770
42	0.51	-345	3340	0.60	-284	2620
47	0.49	-367	3620	0.74	-226	2000

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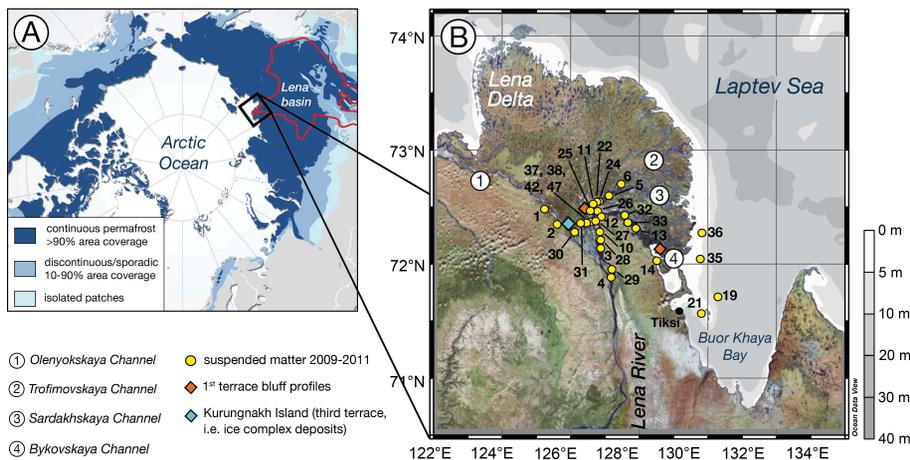


Figure 1. (A) Lena River catchment area within the Northern Hemisphere permafrost zone (map by Hugo Ahlenius, UNEP/GRID-Arendal, see www.grida.no/graphicslib/detail/permafrost-extent-in-the-northern-hemisphere_1266; source data from Brown et al., 1998), (B) Lena Delta and Buor Khaya Bay sampling sites from 2009 to 2011 analyzed for radiocarbon content (see also Table 1).

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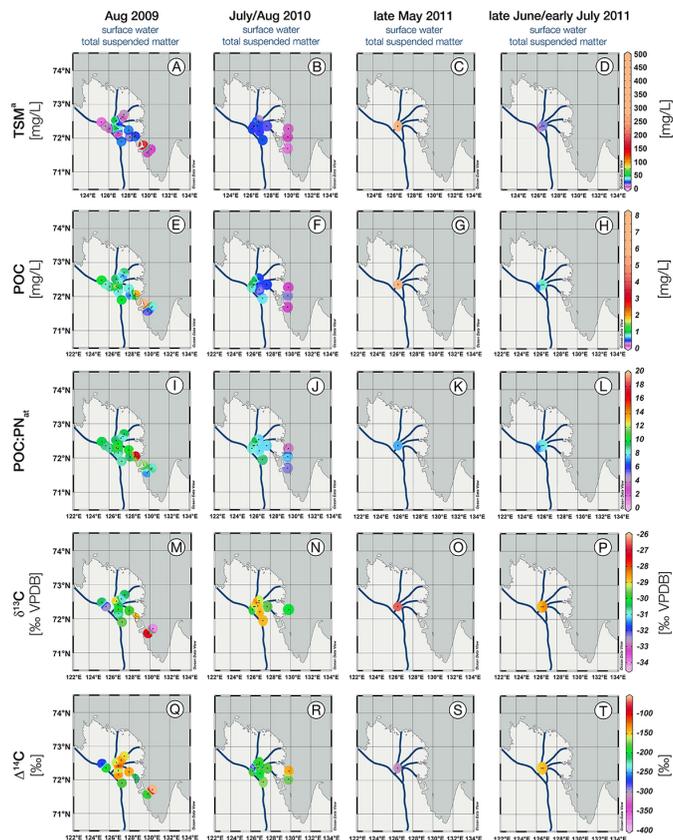


Figure 2. Spatial distribution of organic carbon concentrations, POC : PN ratios and stable and radiocarbon isotopic composition in surface water from 2009–2011. TSM concentrations of the years 2009 to late May 2011 (A–C) taken from Winterfeld et al. (2014), submitted as companion paper.

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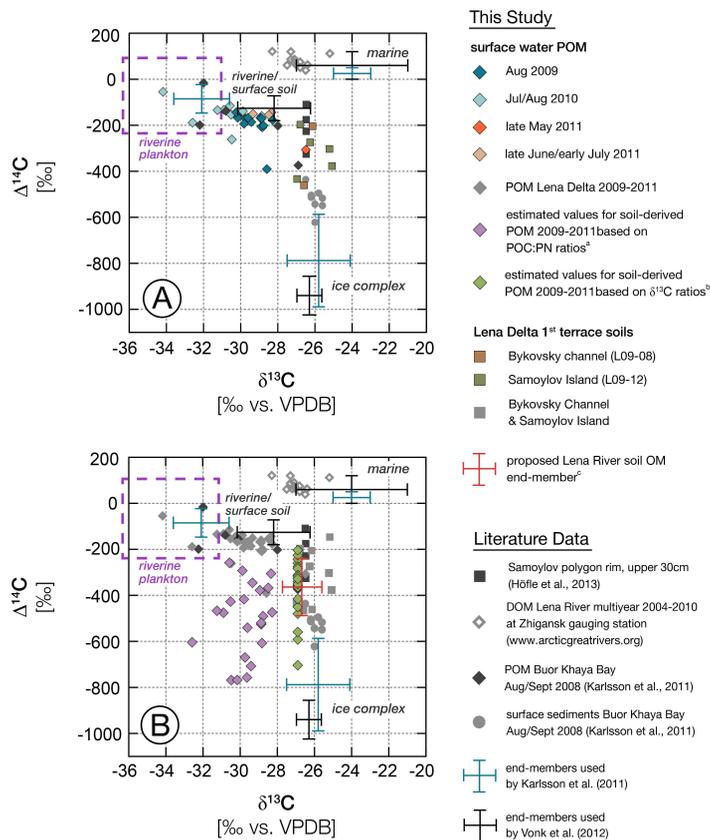
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