Characterization of particulate organic matter in the Lena River Delta and adjacent nearshore zone, NE Siberia. Part I: 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 one of the first radiocarbon (^{14}C) data sets for surface water POM within the Lena Delta sampled in summers 2009-2010 and spring 2011 (n=30 samples). The bulk $\Delta^{14}C$ values varied from –55 to –391‰ translating into ^{14}C ages of 395 to 3920 years BP. We further estimated the fraction of soil-derived POM to our samples based on 1) particulate organic carbon to particulate nitrogen ratios (POC:PN) and 2) on the stable carbon isotope ($\delta^{13}C$) composition of our samples. Assuming that this phytoplankton POM has a modern ^{14}C concentration we inferred the ^{14}C concentrations of the soil-derived POM fractions. The results ranged from –322 to –884 ‰ (i.e. 3.060 to 17,250 ^{14}C years BP) for the POC:PN-based scenario and from –261 to –944 ‰ (i.e. 2,370 to 23,100 ^{14}C years BP). Despite the limitations of our approach, the estimated $\Delta^{14}C$ values of the soil-derived POM fractions seem to reflect the heterogeneous ^{14}C concentrations of the Lena River catchment soils covering a range from Holocene to

Pleistocene ages better than the bulk POM $\Delta^{14}C$ values. We further used a dual-carbon isotope three end-member mixing model to distinguish between POM contributions from Holocene soils and Pleistocene Ice Complex deposits to our soil-derived POM fraction. Ice Complex contributions are comparatively low (mean of 0.14) compared to Holocene soils (mean of 0.32) and riverine phytoplankton (mean of 0.55), which could be explained with the restricted spatial distribution of Ice Complex deposits within the Lena catchment. Based on our newly calculated soil-derived POM $\Delta^{14}C$ values, we propose an isotopic range for the riverine soil-derived POM end-member with $\Delta^{14}C$ of –495 ± 153‰ deduced from our $\delta^{13}C$-based binary mixing model and $\delta^{13}C$ of –26.6 ± 1‰ deduced from our data of Lena Delta soils and literature values. These estimates 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.

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, increasing 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 the 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 ($\delta^{13}C$, $\Delta^{14}C$) of dissolved and particulate organic matter as well as individual biomarkers has helped characterizing and distinguishing these different carbon pools, e.g. 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) are only sparsely available. A recently published dataset from the Arctic Great Rivers Observatory (A-GRO, www.arcticgreatrivers.org, published 10 January 2015) provides POM ^{14}C data for the Lena River from Zhigansk, approximately 900km upstream of the delta. To our knowledge the only additional POM ^{14}C concentrations sampled directly in Siberian rivers are from the Lena River (unpublished in Vonk et al., 2010) and 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 of samples taken off the river mouths are already altered from the "original" river signal. In contrast to Siberia, there are detailed studies on riverine POM ^{14}C concentrations in the Yukon and Mackenzie Rivers in the North American Arctic (Goñi et al., 2005; Guo et al., 2007; Guo and Macdonald, 2006). Here, the POM was found to be significantly older than the dissolved organic matter (DOM) of these rivers and interpreted to be derived from riverbank erosion and thawing permafrost soils compared to a modern vegetation source of DOM (Guo et al., 2006, 2007). Against the backdrop of a warming Arctic (IPCC 2013, ACIA, 2005) and the projected release of old OM in the future presumably as POM rather than DOM (Guo et al., 2007), it is important to assess the age heterogeneity carried by riverine POM and sequestered in the nearshore zone today. This will be an important benchmark to distinguish catchment related changes caused by increasing temperatures from the natural variability of the river system.

In contrast to sedimentary OM, POM provides limited spatial and temporal snapshots of its OM properties. However, it has been shown that riverine POM in arctic rivers carries an integrated signal from their permafrost watersheds representing the watershed environmental characteristics (e.g. Goñi et al., 2000; Lobbes et al., 2000; Vonk and Gustafsson, 2009; Winterfeld et al., 2015). Despite the fact that Lena River POM flux is an order of magnitude smaller than its DOM flux it is more likely to transports the climate change signal from permafrost soils of the river catchment (Guo et al., 2007). Also, it has been proposed that the POM pool could be as important as the DOM pool for arctic carbon cycling, because of its possibly high degradation rates in the water column compared to DOM (Sanchéz-García et al., 2011; van Dongen et al., 2008).

Here we present the first part of a study on particulate OM in the Lena Delta, Siberia (Winterfeld et al. 2015, companion paper). Our POM samples taken in three consecutive years (2009-2011) in the spring and summer seasons add up to the existing data on elemental composition, stable carbon isotopes ($\delta^{13}C$), and radiocarbon (^{14}C) values as well as provide a first POM ^{14}C data set covering three consecutive years in the Lena Delta. Because riverine and marine POM concentrations are usually too low for source specific biomarker ^{14}C analysis, the available ^{14}C data is from bulk OM. This could result in a considerable age bias depending on the contribution of phytoplankton OM with a rather enriched (modern) ^{14}C concentration to the individual samples. We used different approaches to estimate the fraction of soil and plankton-derived OM in our POM samples and estimated the ^{14}C concentration for the soil-derived fraction of POM transported by the Lena River in summer and spring. Further, we used a dual-carbon isotope three end-member mixing model according to Vonk et al. (2010, 2012) to distinguish not only between plankton- and soil-derived OM, but also between OM from Holocene soils and Pleistocene Ice Complex deposits. The estimated ^{14}C concentrations of soil-derived POM will further help to define the typical isotopic signature of river POM more accurately for modeling riverine OM contributions to Laptev Sea shelf sediments.

2 Material and Methods

2.1 Study Area

A detailed description of the Lena River watershed and Lena Delta can for example be found in the companion paper of this study dealing with the lignin composition of OM in the Lena Delta (Winterfeld et al., 2015).

In short, the Lena River is one of the largest Russian Arctic rivers draining a watershed of ~2.46×10^6 km^2 in central Siberia into the Laptev Sea. Continuous permafrost makes up about 72-80% of the drainage area (Amon et al., 2012; Zhang et al., 2005) storing huge amounts of old OM. The permafrost acts as a water impermeable layer and thus affects the regional hydrology and hydrochemistry. The Lena River water discharge and related dissolved and particulate load discharge are highest during spring ice-breakup and snow melt in late May to June while summer and winter discharges are lower (Rachold et al., 2004). The mean annual water discharge is ~588 km^3 for the years 1999 to 2008 (Holmes et al., 2012). Corresponding annual sediment, dissolved organic carbon (DOC), and particulate organic carbon (POC) fluxes are 20.7 Tg/yr (Holmes et al., 2002), 5.7 Tg/yr (Holmes et al., 2012), and 1.2 Tg/yr respectively (Rachold and Hubberten, 1999). A second major source for terrigenous OM delivered to the Laptev Sea is the thermal erosion of ice- and OM-rich Pleistocene ice complex deposits along the coast (see Gustafsson et al., 2011; Mueller-Lupp et al., 2000; 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 ~0.66 Tg/yr (Günther et al., 2013).

The Lena River Delta is the largest Arctic delta (~32,000 km^2). It is characterized by a polygonal tundra landscape with active floodplains and it can be divided into three geomorphological terraces (Grigoriev, 1993; Schwamborn et al., 2002). The first terrace includes active floodplains that were formed during the Holocene and makes up about 55% of the total delta area (Morgenstern et al., 2008) covering the central and eastern part. Within the first delta terrace remains of a Pleistocene accumulation plain, also called Ice Complex or Yedoma deposits, form the third terrace covering about 6% of the total delta area (Morgenstern et al., 2008). Sandy islands forming the second terrace cover the rest of the delta in the west. 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. All soil samples were taken from riverbank bluffs of the first delta terrace (Holocene formation), which is elevated (5-16m) over the active floodplains. The soil profiles vary strongly in sediment composition and OM content. Within the profiles sandy layers derived from extreme flooding events (Schwamborn et al., 2002) and aeolian input (Kutzbach et al., 2004; Sanders, 2011) alternate with buried surface soil layers and peat layers rich in fibrous plant and root detritus in different stages of decomposition. The peat layers are either of autochthonous or of allochthonous origin. Allochthonous material is eroded from river banks further upstream and re-deposited in the delta. In order to obtain samples that reflect the original state of the frozen permafrost soils, we removed the thawed soil material from each riverbank bluff for its total height with a spade. Frozen pieces of soil were then 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) from the upper 0.5m of the water column. In the smaller delta channels the samples were taken approximately in the middle of the channel. Because the main delta channels can be >1km in width, we sampled a couple of hundred meters off the shore with water depth of >10m, but not in the middle of the channel. Three samples (19, 35, 36; Fig. 1B) were taken outside the delta in shallow water depth. We discuss these samples as “delta samples”, because the surface water at these sampling locations is outflowing Lena River water as shown by temperature and salinity profiles by Semiletov et al. (2011) and Kraberg et al. (2013). Between 1 and 30 L of water were filtered on pre-combusted (4.5h at 450°C) and pre-weighed glass fiber filters (GF/F Whatman, 0.7µm 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 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 this sample (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 (4.5h at 450°C) and GF/F filters were either stored in pre-combusted petri dishes (Ø 47mm) 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 24h.

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. Every 10 samples control standards were analyzed to constrain the analytical uncertainty of 0.1%.

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

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 and inorganic carbon was removed during sample preparation at NOSAMS. The radiocarbon analyses at NOSAMS were carried out using standard methods (McNichol et al., 1994). Results are reported as $\Delta^{14}C$, conventional radiocarbon ages (years BP), and fraction modern carbon (*fMC*) including the correction for isotope fractionation (Stuiver and Polach, 1977).

The stable carbon isotope composition ($\delta^{13}C$) was measured on splits of the CO_2 gas of the samples generated prior to graphite reduction at NOSAMS using a VG Optima IRMS. Results are reported in per mil (‰) relative to VPDB.

2.4 End-member modeling

The relative source contributions (phytoplankton and soil) to the POM samples are estimated by solving linear two end-member (using POC:PN and $\delta^{13}C$ as parameters) and dual-carbon isotope ($\delta^{13}C$, $\Delta^{14}C$) three end-member mixing models with end-members determined from own data and the literature. The end-member values used in the different modeling approaches are given in Table 4. Source data for the calculation of the POC:PN soil end-member as well as the $\delta^{13}C$ and $\Delta^{14}C$ end-members for Holocene soil are given in Tables S1-3 in the supplement.

To account for the uncertainty of the measured values, we assume independent, normally distributed uncertainties for the observed values with a standard deviation characterizing the measurement uncertainty. For the end-members we assume a normal distribution using the standard deviations from the observed samples on which the end-members are based on.

Uncertainty in the resulting contributions of phytoplankton and soil are estimated in a Monte Carlo approach similar to Vonk et al. (2010, 2012) and Karlsson et al. (2011). The source fractions are estimated 100,000 times, each time drawing the end-members as well as the observed values from their distributions. Solutions with negative fractions are omitted and the mean and the standard deviation are reported.

3 Results

3.1 Elemental composition

The organic carbon (OC) and total nitrogen (TN) concentrations of the first terrace soil samples can be found in Tables 3 and S3 in the companion paper Winterfeld et al. (2015).

The surface water POC concentrations within the delta showed a high spatial and interannual variability similar to the TSM concentrations of the respective samples. The mean POC concentration for August 2009 was 1.21 mg/L (n=21, range 0.35-7.24 mg/L) and corresponding POC content was 7.2 wt% (range 1.9-37.7 wt%, Table 2 and Table S1). The POC contents for July/August 2010 and June/July 2011 were lower than in 2009 with mean concentrations of 0.57 mg/L (n= 13, 0.15-1.30 mg/L) and 3.1 wt% (1.4-4.7 wt%) as well as 0.74 mg/L (n=9, 0.29-1.51 mg/L) and 4.3 wt% (3.2-5.0 wt%), respectively. The single sample from late May 2011 (sample ID 37) showed highest POC concentration per liter (8.2 mg/L) of all presented samples with a related POC content of 1.7 wt%. Our POC data are well within the range of values reported for the Lena Delta before (Cauwet and Sidorov, 1996; Rachold and Hubberten, 1999; Semiletov et al., 2011). The two samples from the Buor Khaya Bay surface waters showed the lowest POC concentrations per liter, i.e. 0.37 mg/L (sample 35) and 0.15 mg/L (sample 36) with corresponding 4.4 and 1.5 wt% POC, respectively (Table 2 and S4 supplement). The considerable drop of POC (and TSM) concentrations offshore the delta is due to flocculation and settling of TSM in the zone of fresh and salt water mixing (e.g. Lisitsyn, 1995) and was also observed during other years of sampling (e.g. Cauwet and Sidorov, 1996; Semiletov et al., 2011).

The atomic particulate organic carbon (POC) to particulate nitrogen (PN) ratios (POC:PN) of samples taken in summer 2009 were slightly higher than for summer 2010 samples with mean values of 9.6 (n=20, 6.8-19.3) and of 8.0 (n=13, 5.0-10.3, Table 2 and S4), respectively. The POC:PN ratios of samples taken in June/July 2011 were rather similar to the July/August samples with a mean of 7.8 (n=9, 5.9-9.7). The sample from late May 2011 had a POC:PN ratio of 7.5.

3.2 Carbon isotope inventories

The stable carbon ($\delta^{13}C$) and radiocarbon (^{14}C) results are shown in Table 3. The radiocarbon data presented here will predominantly be discussed in terms of $\Delta^{14}C$ in per mil (‰).

Additionally, the fraction modern carbon (*fMC*) and ^{14}C ages in years before present (yrs BP) are given in Table 3.

3.2.1 Stable carbon isotope composition ($\delta^{13}C$)

The two soil profiles from the first delta terrace showed $\delta^{13}C$ values between –27.0‰ and –25.1‰ with a mean of –26.1‰, (n = 7, Table 3), which is within the range observed for Holocene soils in the delta (e.g. Schirrmeister et al., 2011 and references therein). Similar to the POC contents, the Lena Delta surface water POM $\delta^{13}C$ values varied strongly spatially and annually (Fig. S1 supplement). POM from August 2009 showed more depleted $\delta^{13}C$ values compared to the other years, ranging from –34.2‰ to –28.8‰ (mean value = –30.4‰, n = 13). July/August 2010 and June/July 2011 POM isotopic compositions ranged from –30.4‰ to –28.3‰ (mean value = –29.3‰, n = 13) and from –29.3‰ to –28.3‰ (mean value = –28.7‰, n = 3), respectively. The isotopically most enriched POM $\delta^{13}C$ value of –26.5‰ was determined for the sample from late May 2011 (Table 3). The $\delta^{13}C$ of Buor Khaya Bay surface water POM in August 2010 could only be determined for one of the samples, i.e. sample 36 with –30.4‰.

3.2.2 Radiocarbon (^{14}C) concentration

The $\Delta^{14}C$ values 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 1,710 to 4,900 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 $\Delta^{14}C$ values reflect the late Holocene formation of these soils and fit within the range of ages determined for the Lena Delta first terrace (Bolshiyanov et al., 2015; Kuptsov and Lisitsin, 1996; Schwamborn et al., 2002).

As with other TSM parameters, the POM $\Delta^{14}C$ values showed strong spatial and interannual variability (Fig. 2, panels Q-T and Fig. S1). Lena Delta POM ^{14}C 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 ^{14}C concentration of –306‰ and the Buor Khaya Bay surface samples of –176‰ (sample 35) and –143‰ (sample 36, Table 3, Fig. 2R-S). Overall these ^{14}C concentrations covered a range of ^{14}C ages from

395 to 3,920 yrs BP. The samples with the lowest $\Delta^{14}C$ values of –262‰ (sample 1) and –391‰ (sample 31) were taken close to the Pleistocene Ice Complex deposits, which likely contributed to the local POM in the Lena River surface water.

4 Discussion

4.1 Origin of organic matter in the Lena Delta

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 (0-400m elevation, e.g. Grosse et al., 2013) and the Holocene permafrost soils. POC:PN ratios as well as $\delta^{13}C$ and $\Delta^{14}C$ values 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 and S4) similar to the TSM and lignin phenol concentrations sampled during the same field trips (Winterfeld et al. 2015, companion paper). 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 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 would suggest a considerable fraction of primary production OM to be present in our samples.

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 section 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).

Analogous to the POC:PN ratios, the $\delta^{13}C_{POM}$ values point to a considerable contribution of primary production OM to our samples from 2009-2011. The $\delta^{13}C_{POM}$ values of our samples taken in 2009-2011 vary over a broad range (–34.2 to –26.5‰, Table 3), which is similar to

$\delta^{13}C_{POM}$ 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 A-GRO (www.arcticgreatrivers.org) ~900km 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 $\delta^{13}C_{POM}$ 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 $\delta^{13}C_{POM}$ values (<–29‰) point to mixing with riverine plankton utilizing dissolved inorganic carbon (DIC) with depleted $\delta^{13}C$ signatures as suggested for the Lena River (Alling et al., 2012; Rachold and Hubberten, 1999).

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.

In chapter 4.3 we present the results of two binary and one three end-member mixing model to estimate the soil OM contribution to our POM samples and to further calculate the ^{14}C concentration of the soil fraction.

4.2 ^{14}C and age heterogeneity of POM ($^{14}C_{POM}$) 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 $^{14}CO_2$, 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 versus lignin) from arctic watersheds (e.g. Feng et al., 2013; Gustafsson et al., 2011).

The Lena Delta and Buor Khaya Bay surface water $^{14}C_{POM}$ concentrations presented here are depleted with respect to the current atmospheric $^{14}CO_2$ (Table 3). The $\Delta^{14}C$ values range from

–391 ‰ to –55 ‰ for 2009 to 2011 translating into ^{14}C ages >400 yrs and up to 3,920 yrs BP for samples taken close to the Pleistocene Ice Complex deposits of Kurungnakh Island (Fig. 2, panels Q-T, Table 3). These results are within the range of reported $\Delta^{14}C$ values 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 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 3,000 yrs BP in the Lena Delta (30 cm below surface, Höfle et al., 2013) or more than >10,000 yrs BP south of the delta (Kuptsov and Lisitsin, 1996). As shown by the lignin phenol composition of Lena Delta POM (Winterfeld et al., 2015, companion paper) approximately half of the surface water POM 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 soil profile, not only from the active layer.

One possible explanation for our relatively young POM ^{14}C ages could be the contribution of plankton-derived OM with a rather modern ^{14}C concentration concealing the "true" age of soil-derived OM. Phytoplankton-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. In the following section 4.3 we present results from two different approaches to quantify the soil OM fraction in our samples and further estimate its ^{14}C concentration based on the assumption of plankton-derived OM with modern $\Delta^{14}C$ values.

4.3 Quantitative POM source determination and soil ^{14}C concentrations

4.3.1 Binary mixing model scenario 1

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, as noted above (section 4.1) 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 also result in lower POC:PN ratios. 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 Sanchéz-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 versus PN content regression line at POC = 0 (Fig. S2). By subtracting these amounts from the analyzed total PN content of the respective samples, we could calculate new soil POC:PN ratios ($POC:PN_{NEW}$) for our samples (Table S4 supplement). Based on these $POC:PN_{NEW}$ ratios (and their uncertainty, derived from the standard error of the POC versus PN regressions), we calculated the soil-derived fraction within our POM samples using a simple two end-member mixing model as following:

$$POC:PN_{NEW} = f_{soil} \times POC:PN_{soil} + f_{plankton} \times POC:PN_{plankton} \quad (1)$$

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

where $POC:PN_{NEW}$ is the corrected value of the POM sample (Table S4). As the end-member for $POC:PN_{soil}$ we chose 23.7 ± 11 (Table 4 and S1 supplement), which is an average calculated from Lena Delta first terrace soils presented in Winterfeld et al. (2015, companion paper) and delta soils as well as soil from the Lena River watershed covering the taiga to tundra transition from Höfle et al. (2013), Zubrzycki (2013), and Zubrzycki e al. (2012, individual values in Table S1 supplement). The $POC:PN_{plankton}$ end-member value is 6 ± 1 (Table 4, e.g. Meyers, 1994).

The calculated soil-derived OM fractions of Lena Delta POM varied from 0.26 to 0.70 (mean of 0.35, n=21, Table 5) for summer 2009, from 0.26 to 0.44 (mean of 0.34, n=11) in summer

2010, and from 0.39 to 0.50 (mean of 0.43, n=9) for late June/early July 2011 (Table 5). We used the POC versus 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 0.39 (sample 35) and 0.12 (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 0.13 (Table 5). 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 POC:PN_{NEW} ratios (Table S4) might be highly over- and underestimating OM fractions in individual samples.

The calculated soil and plankton OM fractions (f_{soil} and $f_{plankton}$, Table 5) were further used in an isotopic mass balance to determine the ^{14}C concentration of the soil fraction assuming the plankton-derived OM is modern ($\Delta^{14}C_{plankton}$ = 41.9 ± 4.2‰, Table 4):

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

and $f_{POM} = f_{soil} + f_{plankton}$ (4)

where f_{POM}, f_{soil}, and $f_{plankton}$ are the fractions of POM, soil- and plankton-derived OM and $\Delta^{14}C_{POM}$, $\Delta^{14}C_{soil}$, and $\Delta^{14}C_{plankton}$ are the $\Delta^{14}C$ values of these sources. As noted above, 41.9 ± 4.2‰ is assumed as a maximum estimate for $\Delta^{14}C_{plankton}$ based on atmospheric CO_2 $\Delta^{14}C$ values for May to August 2009-2011 from the Schauinsland observatory, Germany.

The results of the newly calculated soil $\Delta^{14}C$ values using POC:PN_{NEW} ratios ($\Delta^{14}C_{POC:PN}$) to partition between soil- and plankton-derived OM are shown in Table 5. The soil $\Delta^{14}C_{POC:PN}$ values range from –884 ‰ to –322 ‰ for the sampling period of 2009-2011 translating into soil ^{14}C ages >3,000 yrs BP with an average of 6,700 yrs BP (Table 5). Analogous to the two comparatively old bulk $^{14}C_{POM}$ ages off Kurungnakh Island taken in 2009 and 2010 (Table 3), the calculated $^{14}C_{POC:PN}$ age for sample 1 was 17,250 yrs BP and yielded a fossil age (>50,000 years BP) for sample 31. 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 of soil and plankton OM fractions. These uncertainties are further affecting the calculation of the soil $\Delta^{14}C_{POC:PN}$ 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 Binary mixing model scenario 2

Similar to the approach discussed in section 4.3.1, we used a second scenario based on $\delta^{13}C$ values 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 $\delta^{13}C$ of around –25‰ to –27‰ (Rachold and Hubberten, 1999). This is also reflected in our $\delta^{13}C$ data from the first delta terrace soils with an average of –26.2‰ (n=7, Table 3). Bird et al. (2002) determined the $\delta^{13}C$ composition of taiga and tundra soils (excluding peatlands) along Yenisey River on a latitudinal transect. For the binary mixing model we used a soil OM end-member value –26.6 ± 1‰ (Table 4), which is a combination of $\delta^{13}C$ data from this study and the literature (Table S2, Bird et al., 2002; Pitkänen et al., 2002; Xu et al., 2009) covering tundra and taiga soils in Siberia and Alaska.

The $\delta^{13}C$ 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 $\delta^{13}C$ of bicarbonate represents the $\delta^{13}C$ of DIC. Sources for DIC are generally the CO_2 derived from soil OM degradation, CO_2 released during the dissolution of carbonates, and CO_2 from the atmosphere. The Lena River geochemistry is mainly influenced by carbonate weathering and groundwater in the summer season (Gordeev and Sidorov, 1993). Assuming a fractionation of –22.5 ± 2.5‰ between phytoplankton and DIC (Mook and Tan, 1991) and a $\delta^{13}C_{DIC}$ of –8‰ for the Lena Delta (Alling et al., 2012) a plankton $\delta^{13}C$ end-member value of –30.5 ± 2.5‰ would be expected. Similar or more depleted $\delta^{13}C$ 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 $\delta^{13}C_{POM}$ of –34.2‰ for a sample from the summer 2009 (Table 3) is even lower than the plankton end-member used here ($\delta^{13}C_{plankton}$ = –30.5‰). The sample location is offshore Muostakh Island (Fig. 1B) and influenced by mixing with marine waters, which complicates the DIC composition and processes affecting $\delta^{13}C$ of DIC (Alling et al., 2012).

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:

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

and $1 = f_{soil} + f_{plankton}$ (6)

where $\delta^{13}C_{POM}$ is the analyzed $\delta^{13}C$ value of our POM samples (Table 3), $\delta^{13}C_{soil}$ is the soil end-member value of –26.6 ± 1‰, and $\delta^{13}C_{plankton}$ is plankton end-member value of –30.5 ± 2.5‰ (Tables 4 and S3 supplement).

The calculated soil OM fractions varied from 0.12 to 0.51 in summer 2009 (mean of 0.32, n=12, Table 5), from 0.34 to 0.59 in summer 2010 (mean of 0.45, n=12), and from 0.44 to 0.59 in spring 2011 (mean of 0.53, n=3). The Buor Khaya Bay POM sample 21 showed a soil fraction of 0.75 and the spring freshet value (sample 37) a soil fraction of 0.80 (Table 5). As discussed above in Sect. 4.3.1, these soil OM fractions are rough estimates in the absence of direct plankton determination.

Furthermore, $\Delta^{14}C$ values of the soil-derived OM fraction were calculated for the contribution of modern plankton OM as described in Sect. 4.3.1 using the Eq. (3) and (4). Because the soil contributions calculated in this scenario are slightly higher than in the POC:PN-based scenario, the $\Delta^{14}C$ calculations resulted in less radiocarbon depleted estimates compared to the POC:PN scenario (table 5). Nonetheless, the $\Delta^{14}C$ values based on the $\delta^{13}C$ end-member model are considerably ^{14}C-depleted compared to bulk POM ^{14}C concentrations. The estimated $\Delta^{14}C$ values range from –944‰ to –495‰ for all samples 2009-2011 representing ^{14}C ages from 23,100 to 2,370 yrs BP (Table 5). The oldest samples are, again, the ones taken close to Pleistocene Ice Complex deposits (samples 1 and 31).

4.3.3 Comparison of scenario 1 and 2

Of all samples from 2009-2011 presented here, 28 samples have a soil fraction estimate from both scenarios. For these samples the POC:PN scenario gives a lower mean soil fraction of 0.33 compared to 0.43 from scenario 2 (based $\delta^{13}C$, Fig. S3). The comparison demonstrates that the error estimates determined in the end-member calculation are realistic, as only 6 (2) of 28 samples are outside one (two) standard deviations of the estimated error. As the uncertainty of the estimates is high, both estimates are only weakly correlated (removing the clear outlier sample 37 we get $R=0.3$, $p=0.1$).

Because we assumed a modern $\Delta^{14}C$ value for the phytoplankton OM, the estimated soil ^{14}C concentrations are related to the amount of soil OM calculated in both scenarios. The lower mean fraction in scenario 1 therefore resulted in a lower mean soil $\Delta^{14}C$ value of –569‰ (^{14}C age = 6,700 year BP, n= 26) compared to –495‰ (^{14}C age = 5,430 years BP, n= 28) for scenario 2. Besides the spring freshet sample from 2011, the two samples with the lowest bulk $\Delta^{14}C$ values (sample 1 & 31, Table 3) also showed the one of the lowest estimated soil $\Delta^{14}C$ values, namely –884‰ and radiocarbon-free (fossil) for scenario 1 as well as –944‰ and –

754‰ for scenario 2. The two samples were taken close to Ice Complex deposits within the delta, which illustrates the locally pronounced influence of these deposits. Another sample standing out is sample 19 taken outside the delta close to Muostakh Island, which is also composed of Ice Complex deposits (Fig. 1B). Its bulk POM Δ^{14}C value was highest (Δ^{14}C = –55‰, Table 3) pointing to comparatively young OM and its δ^{13}C value was lowest (–34.2‰) pointing to a rather high contribution of phytoplankton. This is in our line of argumentation assuming a contribution of plankton-derived OM with a modern Δ^{14}C value. Because of our chosen end-members, the soil fraction in sample 19 was rather small in the ^{13}C-based scenario and the resulting Δ^{14}C value of the soil fraction was with –774‰ (^{14}C age of 11,890 years BP) the second lowest.

In contrast to scenario 1, it is more obvious in scenario 2 that the POM samples taken in late June/early July 2011 are more enriched in ^{14}C (younger) than the POM samples taken later in the summer (Aug 2009 and July/Aug 2010). Similar observation were made for DOM Δ^{14}C concentrations of the Lena River at Zhigansk ~900km south of the delta (Raymond et al., 2007) and the Kolyma River in East Siberia (Neff et al., 2006). Their explanation was that due to the deepening of the active layer in summer older soil layers are accessible for melt water and groundwater contributing an older DOM signature to the river than in spring when most soil is still frozen. A comparable scenario could explain estimated Δ^{14}C concentration decreasing from spring freshet to summer. In addition to active layer deepening riverbank erosion is strongest during the summer and might contribute a considerable amount of soil-derived OM with a large ^{14}C age range to the Lena Delta surface water.

4.3.4 Holocene soil versus Pleistocene Ice Complex deposits

As an extension of the two end-member model, we applied a dual-carbon isotope (δ^{13}C, Δ^{14}C) three end-member model (after Karlsson et al., 2011; Vonk et al., 2010, 2012) to separate the source contributions of phytoplankton, Holocene soil and Pleistocene Ice Complex deposits (ICD) using the following equations:

$$\delta^{13}C_{POM} = f_{soil\_Holocene} \times \delta^{13}C_{soil} + f_{soil\_ICD} \times \delta^{13}C_{soil} + f_{plankton} \times \delta^{13}C_{plankton} \quad (7)$$

$$^{14}C_{POM} = f_{soil\_Holocene} \times {}^{14}C_{soil} + f_{soil\_ICD} \times {}^{14}C_{soil} + f_{plankton} \times {}^{14}C_{plankton} \quad (8)$$

and $1 = f_{soil\_Holocene} + f_{soil\_ICD} + f_{plankton}$ (9),

where $f_{soil\_Holocene}$, f_{soil_ICD}, and $f_{plankton}$ are the fractions of Holocene soil, Ice Complex deposits, and riverine plankton contributing to each POM sample. The end-member values chosen for Holocene soil OM were δ^{13}C –26.6 ± 1‰ and Δ^{14}C of –282 ± 133‰. As the two soil end-members have very similar $\delta^{13}C$ values (see Table 4, Holocene soil $\delta^{13}C = -26.6 \pm 1‰$, Ice Complex $\delta^{13}C = -26.3 \pm 0.67‰$), the $\delta^{13}C$ mainly determines the phytoplankton vs. total (Holocene + Pleistocene) soil fraction, whereas $\Delta^{14}C$ mainly determines the fraction of Holocene to Pleistocene soil. Therefore, the results concerning the total soil fraction are very similar to those derived from the two-end-member model (R^2=0.92, mean two end-member soil fraction is 0.43, mean three end-member is 0.46, Fig. S4) than derived with the two end-member model.

As shown in Table S5 and Figure 4, the POM source determinations resulted in relative fractions of 0.19-0.83, 0.11-0.65, and 0.07-0.34 for plankton, Holocene soil, and Ice Complex deposits, respectively. Again, sample 31 taken off Kurungnakh Island (Ice Complex) stands out with the highest contribution from ICD (fraction of 0.34) and sample 19 taken outside the delta with highest contribution of phytoplankton (fraction of 0.83). Overall, with the end-members chosen here and despite the high spatial variability within the delta, the riverine phytoplankton fraction contributes most (mean of 0.55) to the surface water POM compared to the Holocene soil OM (0.32) and Ice Complex deposits (0.14). The rather low OM contribution from Ice Complex deposits reflects the distribution of these deposits in the Lena watershed, where they are only locally concentrated within elevations up 400m (Grosse et al., 2013).

4.4 Implications of estimated soil-derived POM $\Delta^{14}C$

The two binary mixing models (scenario 1 & 2) discussed above allow an estimate of the soil $\Delta^{14}C$ values based on a contribution of modern phytoplankton-derived OM. Both scenarios show considerably ^{14}C-depleted soil-derived OM compared to the bulk $^{14}C_{POM}$ concentrations. This implies that the bulk POM ^{14}C age of samples taken in surface water during the summer, when the riverine primary production is high, likely underestimate the age of the soil-derived OM transported by the Lena River. The estimated soil $\Delta^{14}C$ values and ^{14}C ages in both scenarios give a more plausible picture for soil-derived POM in the Lena River watershed. In contrast to DOM that is restricted in its flow path to the unfrozen soil layers, POM is not exclusively derived from surface soils. It also originates from resuspension of accumulated pre-aged material along the river channels and from riverbank erosion. The latter contributes POM with ^{14}C concentrations representing the whole range covered by the respective riverbank bluffs. In the Lena Delta this is predominantly OM of late Holocene age with local inputs from ice complex deposits of Pleistocene age (e.g. Bolshiyanov et al., 2015; Schirrmeister et al., 2011; Schwamborn et al., 2002). About half of the POM in the Lena

Delta originates from the boreal forest hinterland south of the delta (Winterfeld et al. 2015, companion paper). In the hinterland the soils along the Lena River and its tributaries can be older than the delta soils, i.e. covering the whole age range from Holocene soils to Pleistocene Ice Complex deposits (e.g. Grosse et al., 2013; Kuptsov and Lisitsin, 1996). Our estimated soil ^{14}C ages of about 2,370 to 23,100 years (Table 5) for both scenarios therefore better reflect these hinterland deposits contributing a heterogeneous ^{14}C age mix to riverine POM than the bulk POM ^{14}C ages.

The soil POM ^{14}C estimates as well as the Lena Delta first terrace soil data ($\delta^{13}C$ and $\Delta^{14}C$) presented here improve our knowledge of the stable and radiocarbon isotopic range characteristic for soil-derived OM exported by the Lena River to the Laptev Sea. This information is critical for modeling the OM contribution from different terrestrial (fluvial vs. coastal erosion) and marine sources to Laptev Sea sediments and thus help characterizing and quantifying the OM pools released from permafrost thawing. Recent studies suggest that OM exported by arctic rivers and OM derived from erosion of ice complex coasts differ in their mineral and OM composition and thus show different potential for remineralization by microorganisms after thawing as well as different modes of transport and burial (e.g. Feng et al., 2013; Gustafsson et al., 2011; Knoblauch et al., 2013; Vonk et al., 2012). This has a direct impact on how to assess the possibility of a positive carbon-climate feedback from permafrost degradation, which has the potential to enhance global greenhouse warming by releasing huge amounts of previously frozen OM to the atmosphere.

A promising approach to distinguish OM sequestered in arctic sediments is the dual-carbon-isotope end-member simulation applied by Karlsson et al. (2011) and Vonk et al. (2012; 2010) to Laptev and East Siberian Sea sediments. The authors use the $\delta^{13}C$ and $\Delta^{14}C$ values of surface water suspended matter and surface sediments to quantify OM derived from Pleistocene ice complex deposits, soil/top-soil OM exported by Siberian rivers, and marine phytoplankton OM. Their end-member definitions 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 the $\delta^{13}C$ end-member chosen for surface soil including our first terrace soil data (Table S2) and our estimates for soil-derived POM (Table 5) provide new $\delta^{13}C$ and $\Delta^{14}C$ value ranges (Fig. 3B) for fluvially exported soil POM. Together with published $\delta^{13}C$ values from tundra and taiga soils in Siberia and Alaska (Bird et al., 2002; Pitkänen et al., 2002; Xu et al., 2009) we defined a Lena River soil OM end-member with a $\delta^{13}C$ value of –26.6 ± 1‰. Based on the $\delta^{13}C$ binary mixing model (scenario 2) the corresponding $\Delta^{14}C$ value is –495 ±

153 ‰ (Table 5). We favor scenario 2 over the POC:PN-based scenario, because it allows to calculate the soil fraction of each POM sample based on its bulk $\delta^{13}C$ value, 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 above proposed $\delta^{13}C$ and $\Delta^{14}C$ values for the soil OM end-member of the Lena River catchment make 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 $\delta^{13}C$ values of both end-members are almost indistinguishable. The $\Delta^{14}C$ range of our soil-derived POM estimates is lower than the end-members used by Karlsson et al. (2011) and Vonk et al. (2012, 2010). Furthermore, Höfle et al. (2013) have shown that OM within the first 30cm of a polygon rim of the Lena Delta first terrace can be 3,000 ^{14}C years 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 $\delta^{13}C$ and $\Delta^{14}C$ values 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 $\Delta^{14}C$ estimates discussed above (and in sections 4.3.1, 4.3.2). 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 (section 4.3.1) and based $\delta^{13}C$ values (section 4.3.2) can only be regarded as rough 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 $\Delta^{14}C$ values for soil-derived POM. An additional source of uncertainty is our assumption of a modern ^{14}C concentration of plankton OM. Without determining the ^{14}C concentration of the Lena River DIC, which is utilized by phytoplankton, we cannot be sure of a modern ^{14}C concentration. Lena River DIC is derived from several sources providing carbon with different ^{14}C concentrations. DIC derived from carbonate and silicate weathering is ^{14}C-depleted, DIC derived from soil respiration has a broad range of $\Delta^{14}C$ values due decomposition of soil OM pools of varying age, and DIC derived from exchange with the atmosphere has modern $\Delta^{14}C$ values. The resulting DIC $\Delta^{14}C$ value is mixture of these sources depending on the varying contribution from each source.

Furthermore, Tank et al. (2012) found that DIC yields are negatively correlated with continuous permafrost extent in the watersheds of the six large Arctic rivers including the Lena. This would imply that carbonate weathering is to some extent hampered by continuous permafrost making up 77% of the Lena catchment area (Tank et al., 2012). In summary, because there is no DIC $\Delta^{14}C$ value available for the Lena River and there are too many factors influencing the DIC $\Delta^{14}C$ value, we made the simplified assumption of modern $\Delta^{14}C$ of 41.9 ± 4.2 (average value of atmospheric CO_2 $\Delta^{14}C$ May-Aug 2009-2011 from Levin et al., 2013). Consequently, if true $\Delta^{14}C$ values of DIC in the Lena were depleted relative to the modern atmosphere, the soil-derived POM would be less ^{14}C depleted than estimated here. Thus, $\Delta^{14}C$ values for soils have to be considered minimum estimates or in other words, the estimated ^{14}C soil ages have to be considered maximum ages.

The best way to determine the $\Delta^{14}C$ value of riverine soil-derived OM would be a biomarker-specific radiocarbon analysis using source-specific compounds, e.g., short- and long-chain alkanoic 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 are only few data available on ^{14}C concentrations of POM from Lena River surface water, but regarding 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 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 one of the first data sets on surface water POM ^{14}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 on the one hand estimated using binary mixing models based on POC:PN ratios and $\delta^{13}C$ values, which allowed us to calculate the $\Delta^{14}C$ values of the soil-derived POM fraction. These soil $\Delta^{14}C$ estimates were low compared to the bulk POM $\Delta^{14}C$ values 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. Moreover, we applied a dual carbon-isotope three end-member model to further distinguish between OM contributions from Holocene soils and Pleistocene Ice Complex deposits to the soil fraction. Here, we could show that the overall contribution of Ice Complex deposits to surface water POM in the Lena Delta was relatively low, which reflects the restricted spatial distribution of these deposits within the Lena watershed. Only samples taken close to Ice Complex deposits exhibited higher contributions of this source in the model implying a small, locally pronounced influence on surface water POM before it becomes mixed with other soil-derived POM during fluvial transport.

Because of the limitations of our approach, particularly the assumption of modern phytoplankton OM without determining the ^{14}C concentration of the Lena River DIC utilized by phytoplankton, our ^{14}C estimates for the soil-derived fraction have to be considered minimum $\Delta^{14}C$ concentrations and maximum ^{14}C ages, respectively. Nonetheless, we propose average values for the soil POM isotopic composition based on our data and published values of $\delta^{13}C = -26.6 \pm 1$ ‰ and $\Delta^{14}C = -495 \pm 153$ ‰ (Tables 5 and S2), 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. Soil samples from riverbank bluffs and total suspended matter (TSM) samples presented in this study analyzed for particulate organic carbon, stable, and radiocarbon isotope composition. Latitude and longitude are given in decimal degrees (dec). Bluff height is given in meters above river level [m a.r.l.] measured in August 2009 and only applicable for the soil samples. The TSM samples were taken from the surface water layer at a sampling depth of ca. 0.5m.

Sample code	Sample & site description	Date of sampling	Lat.N [dec]	Long. E [dec]	Bluff height [m a.r.l.]
Lena Delta first terrace soil profiles					
L09-12	Samoylov Island, 5 depths sampled	18-Aug-2009	72.3775	126.4954	7.5
L09-28-2	Bykovskaya Channel, 2 depths sampled	21-Aug-2009	72.0586	128.6309	1.7
Lena Delta TSM					
1	Olenyokskaya Channel	14-Aug-2009	72.4772	125.2856	
2	Olenyokskaya Channel	14-Aug-2009	72.3598	125.6728	
3	Lena River main channel	16-Aug-2009	72.1526	126.9160	
4	Lena River main channel south of Tit Ari Island	16-Aug-2009	71.9040	127.2544	
5	Sardakhskaya/Trofimovskaya Channel	17-Aug-2009	72.5825	127.1891	
6	Sardakhskaya Channel	17-Aug-2009	72.7002	127.4930	
7	Sardakhskaya/Trofimovskaya Channel	17-Aug-2009	72.6268	127.3860	
8	near Kurungnakh Island	18-Aug-2009	72.2904	126.0909	
9	Lena River main channel	19-Aug-2009	72.2987	126.7080	
10	Lena River main channel	19-Aug-2009	72.2760	126.9041	
11	Lena River main channel	19-Aug-2009	72.5159	126.7142	
12	Bykovskaya Channel	20-Aug-2009	72.4140	126.9124	
13	Lena River Bykovskaya Channel	20-Aug-2009	72.2352	127.9619	
14	Lena River Bykovskaya Channel	20-Aug-2009	72.0341	128.5232	
15	Lena River Bykovskaya Channel	21-Aug-2009	72.0354	128.0974	
16	Lena River Bykovskaya Channel	21-Aug-2009	72.0586	128.6309	
17	offshore Bykovsky Peninsula	22-Aug-2010	71.7889	129.4189	
18	NE of Muostakh Island	22-Aug-2010	71.6761	130.1728	
19	NE of Muostakh Island	22-Aug-2010	71.7062	130.2900	
20	W of Muostakh Island	23-Aug-2010	71.6088	129.9393	
21	close to Muostakh Island shoreline	23-Aug-2010	71.5750	129.8200	
22	close to Samoylov Island	30-Jul-2010	72.3650	126.4628	
24	Sardakhskaya/Trofimovskaya Channel	31-Jul-2010	72.5343	126.8794	
25	Lena River Trofimoskaya Channel	31-Jul-2010	72.4764	126.6250	
26	Lena River Trofimoskaya Channel	31-Jul-2010	72.4764	126.8588	
27	Lena River main channel south of Samoylov	1-Aug-2010	72.3776	126.7478	
28	Lena River main channel north of Tit Ari Island	1-Aug-2010	72.2102	126.9423	
29	Lena River main channel south of Tit Ari Island	1-Aug-2010	71.9514	127.2582	

30	Lena River main channel off Kurungnakh	2-Aug-2010	72.2808	126.2091	
31	Lena River main channel	2-Aug-2010	72.3567	126.3521	
32	Lena River Bykovskaya Channel	3-Aug-2010	72.3604	127.6761	
33	Bykovskaya Channel	3-Aug-2010	72.3604	127.6765	
34	off Bykovsky Peninsula	4-Aug-2010	71.7015	129.7523	
35	offshore Lena Delta	6-Aug-2010	72.0379	129.7694	
36	offshore Lena Delta	6-Aug-2010	72.2753	129.8248	
37	Lena River main channel off Samoylov Island	29-May-2011	72.3651	126.4757	
38	Lena River main channel, south of Stolb	26-Jun-2011	72.3705	126.6538	
39	off Kurungnakh	26-Jun-2011	72.3334	126.2914	
40	close to Samoylov Island	29-Jun-2011	72.3681	126.4738	
41	close to Samoylov Island	29-Jun-2011	72.3681	126.4738	
42	close to Samoylov Island	30-Jun-2011	72.3681	126.4738	
43	close to Samoylov Island	30-Jun-2011	72.3681	126.4738	
44	close to Samoylov Island	1-Jul-2011	72.3681	126.4738	
45	close to Samoylov Island	1-Jul-2011	72.3681	126.4738	
46	close to Samoylov Island	2-Jul-2011	72.3681	126.4738	
47	close to Samoylov Island	2-Jul-2011	72.3681	126.4738	

Table 2. Particulate organic carbon (POC) contents in Lena Delta surface water (2009 to 2011) given in milligram per liter (mg/L) 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 S4). Note that for Aug 2009 there are only n=20 samples for POC (wt%), because the total suspended matter concentration was not determined for sample 19.

		POC [mg/L]	POC [wt%]	atomic POC:PN
Lena Delta TSM 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 TSM Jul/Aug 2010		*n=13*	*n=13*	*n=13*
	mean	0.57	3.05	7.6
	median	0.47	3.05	7.8
	min	0.15	1.42	3.7
	max	1.30	4.74	10.3
Lena Delta TSM late May 2011				
sample code:	37	8.20	1.66	7.5
Lena Delta TSM 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

Table 3. Stable carbon isotope ($\delta^{13}C$) and radiocarbon composition (^{14}C) of Lena Delta first terrace soil profiles and surface water particulate organic matter (2009-2011). Soil profile samples are given in meters below surface (m b.s.). Not determined is denoted by *n.d.*

Sample code	$\delta^{13}C$ [‰ VPDB]	*fMC*[1]	1σ *fMC*	$\Delta^{14}C$ [‰]	conv. ^{14}C age[2] [yrs BP]	1σ ^{14}C age [yrs BP]	Lab ID
Lena Delta first terrace soils							
L09-12, 0.45m b.s.	-26.8	0.8084	0.0039	-197	1710	40	OS-84097
L09-12, 1.35m b.s.	-26.3	0.7311	0.0029	-274	2510	30	OS-84073
L09-12, 2.50m b.s.	-25.2	0.7023	0.0025	-303	2840	30	OS-84072
L09-12, 4.70m b.s.	-27.0	0.5708	0.0024	-433	4500	35	OS-84071
L09-12, 5.80m b.s.	- 25.1	0.6275	0.0027	-377	3740	35	OS-84070
L09-28, 030m b.s.	-26.1	0.8015	0.0026	-204	1780	25	OS-84074
L09-28, 070m 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	-55	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 June/early July 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	

[1]*fMC*=fraction modern carbon, [2]conv. age=conventional radiocarbon age in years before present [yrs BP], i.e. before 1950

Table 4. End-member values used for the binary mixing models based on POC:PN and $\delta^{13}C$, and for dual-carbon isotope three end-member mixing model. If not directly taken from the literature, individual values from the literature and this study used to calculate the end-members can be found in Tables S1-S3 in the supplement.

	Scenario 1 POC:PN		Scenario 2 $\delta^{13}C$		Dual-carbon isotope three end-member		
	riverine phytoplankton	soil	riverine phytoplankton	soil	riverine phytoplankton	Holocene soils	Ice Complex deposits
POC:PN	6 ±1[1]	23.7 ± 11	–	–	–	–	–
$\delta^{13}C$ [‰ vs. VPDB]	–	–	30.5 ± 2.5	–26.6 ± 1.0	30.5 ± 2.5	26.6 ± 1.0	–26.3 ± 0.67[3]
$\Delta^{14}C$ [‰]	–	–	–	–	41.9 ± 4.2[2]	–282 ± 133	–940 ± 84[3]

[1]Meyers (1994) and references therein

[2]average and standard deviation of atmospheric $\Delta^{14}C$ values from May-Aug 2009-2011 from Levin et al. (2013)

[3]from Vonk et al. (2012)

Table 5 Soil fractions calculated based on POC:PN ratios and $\delta^{13}C$ values and associated estimated ^{14}C concentrations of the soil-derived POM fraction. Soil fractions are given with standard deviation (1σ). $\Delta^{14}C$ values are given with lower quantile (q16) and upper quantile (q84) representing the 68% confidence interval (1σ for normal distributions). $\Delta^{14}C$ values lower than -1000‰ were marked as 'fossil'. Not determined denoted by *n.d.*

	POC:PN based estimates					plankton $\delta^{13}C$-based estimates				
sample code	fraction soil $POM_{POC:PN}$	$\Delta^{14}C_{POC:PN}$ [‰]	$\Delta^{14}C_{POC:PN}$ q16 [‰]	$\Delta^{14}C_{POC:PN}$ q84 [‰]	conv. $^{14}C_{POC:PN}$ age [yrs BP]	fraction soil $POM_{\delta13C}$	$\Delta^{14}C_{\delta13C}$ [‰]	$\Delta^{14}C_{\delta13C}$ q16 [‰]	$\Delta^{14}C_{\delta13C}$ q84 [‰]	conv. $^{14}C_{\delta13C}$ age [yrs BP]
Lena Delta TSM Aug 2009										
1	0.33 ± 0.18	–884	fossil	–550	17,250	0.31 ± 017	–944	fossil	–585	23,100
2	0.32 ± 0.18	–688	fossil	–416	9,300	0.17 ± 0.12	fossil	–	–	fossil
3	0.30 ± 0.18	–535	fossil	–313	6,100	0.28 ± 0.16	–591	fossil	–354	7,120
4	0.26 ± 0.17	–811	fossil	–465	13,320	0.40 ± 0.20	–511	–967	–327	5,690
5	0.31 ±0.18	–528	fossil	–314	5,970	0.24 ± 0.15	–681	fossil	–399	9,120
6	0.36 ±0.19	–506	–998	–492	5,600	0.31 ± 0.17	–594	fossil	–363	7,180
7	0.27 ± 0.18	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
8	0.29 ± 0.18	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
9	0.38 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
10	0.38 ± 0.19	–506	–985	–322	5,600	0.38 ± 0.19	–500	–953	–317	5,510
11	0.32 ± 0.18	-595	fossil	–359	7,200	0.50 ± 0.21	–364	–655	–245	3,580
12	0.42 ± 0.20	–332	–625	–213	3,190	0.30 ± 0.17	–483	–981	–291	5,240
13	0.43 ± 0.20	–382	–709	–249	3,810	0.37 ± 0.19	–451	–899	–282	4,760
14	0.34 ± 0.19	–704	fossil	–434	9,720	0.51 ± 0.22	–447	–793	–307	4,700
15	0.38 ± 0.20	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
16	0.70 ± 0.16	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
17	0.36 ± 0.18	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
18	0.38 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
19	0.27 ± 0.17	–322	–718	–174	3,060	0.12 ± 0.09	–774	fossil	–407	11,890
20	0.38 ± 0.20	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
21	0.30 ± 0.20	–724	fossil	–413	10,280	0.75 ± 0.20	–261	–377	–214	2,370
Lena Delta TSM Jul/Aug 2010										
22	0.30 ± 0.18	–675	fossil	–402	8,970	0.39 ± 0.20	–505	–976	–322	5,590
24	0.30 ± 0.18	–708	fossil	–424	9810	0.43 ± 0.20	–491	–914	–320	5,370
25	0.37 ± 0.19	–544	fossil	–349	5100	0.59 ± 0.22	–330	–553	–237	3,160
26	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	0.50 ± 0.21	–456	–807	–308	4,830
27	0.44 ± 0.20	–431	–787	–284	3760	0.50 ± 0.21	–456	–679	–256	3,780
28	0.41 ± 0.20	–544	fossil	–355	5360	0.51 ± 0.21	–380	–756	–296	4,490
29	0.35 ± 0.18	–570	fossil	–353	5890	0.50 ± 0.21	–384	–690	–258	3,830
30	0.26 ± 0.17	–764	fossil	–437	11700	0.34 ± 0.18	–581	fossil	–360	6,930
31	0.31 ± 0.18	fossil	–	–	fossil	0.54 ± 0.22	–754	fossil	–536	11,210
32	0.34 ± 0.19	–564	fossil	–349	6190	0.40 ± 0.20	–476	–903	–306	5,130
33	0.32 ± 0.18	–694	fossil	–419	8890	0.38 ± 0.19	–579	fossil	–366	6,890

34	0.36 ± 0.21	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
35	0.39 ± 0.20	515	–997	–329	5190	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
36	0.12 ± 0.13	fossil			*n.d.*	0.34 ± 0.18	–510	fossil	–312	5,670
Lena Delta TSM late May 2011										
37	0.13 ± 0.13	fossil			*n.d.*	0.80 ± 0.19	–393	–539	–341	3,950
Lena Delta TSM late Jun/early Jul 2011										
38	0.43 ± 0.19	–391	–704	–258	2870	0.59 ± 0.22	–274	–460	–196	2,510
39	0.50 ± 0.20	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
41	0.39 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
42	0.41 ± 0.19	–429	–798	–280	3340	0.44 ± 0.21	–394	–738	–256	3,970
43	0.43 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
44	0.41 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
45	0.43 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
46	0.44 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
47	0.40 ± 0.19	–451	–848	–290	3620	0.56 ± 0.22	–310	–534	–215	2,920

Figure captions

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).

Figure 2. Spatial distribution of organic carbon concentrations, POC:PN ratios and stable and radiocarbon isotopic composition in surface water total suspended matter (TSM) samples from 2009-2011. TSM concentrations of the years 2009 to late May 2011 (panels A-C) taken from Winterfeld et al. (2015, companion paper).

Figure 3. Stable and radiocarbon isotopic values of Lena Delta surface water POM of A) this study and literature data as well as B) estimated isotopic data of soil-derived POM based on POC:PN and $\delta^{13}C$ ratios. Ranges for different end-members for topsoil, ice complex, and marine are taken from the literature.

Figure 4. Results of dual carbon-isotope ($\delta^{13}C$, $\Delta^{14}C$) three end-member mixing model showing the fractions for riverine phytoplankton, Holocene soils, and Pleistocene Ice Complex deposits contributing to individual surface water POM samples. The end-member used in the model can be found in Table 4. Individual values comprising the Holocene soil $\delta^{13}C$ and $\Delta^{14}C$ end-member are given in Tables S2 and S3 in the supplement. Note the bulk POM $\Delta^{14}C$ values are ordered from the lowest to the highest and the respective sample IDs are given on top axis.

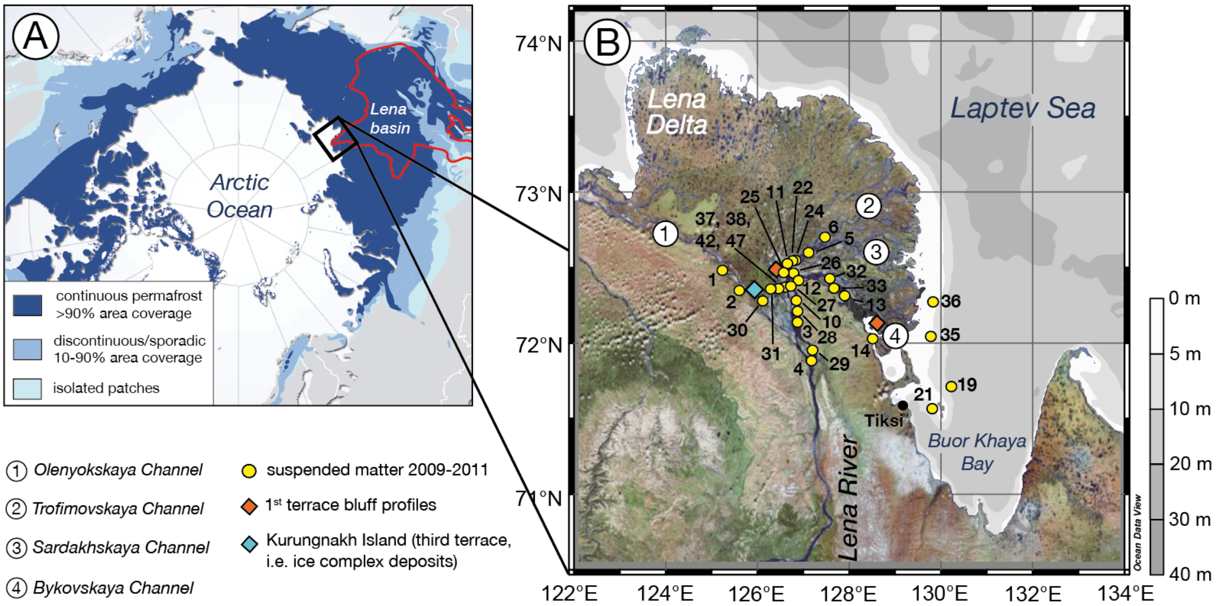
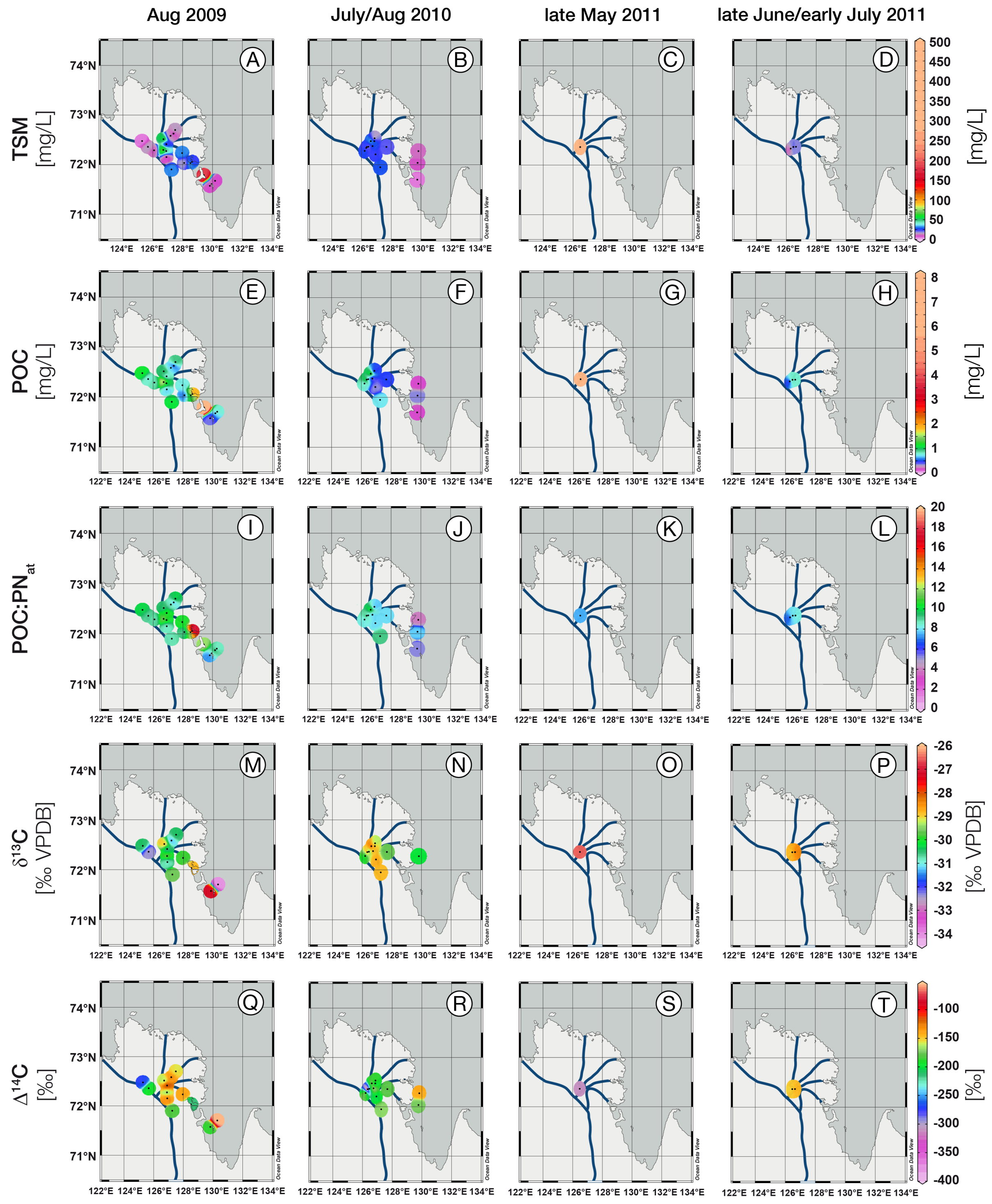

A
Lena basin
Arctic Ocean
continuous permafrost >90% area coverage
discontinuous/sporadic 10-90% area coverage
isolated patches
B
Lena Delta
Laptev Sea
Buor Khaya Bay
Tiksi
Lena River
74°N
73°N
72°N
71°N
122°E
124°E
126°E
128°E
130°E
132°E
134°E
0 m
5 m
10 m
20 m
30 m
40 m
Ocean Data View
① Olenyokskaya Channel
② Trofimovskaya Channel
③ Sardakhskaya Channel
④ Bykovskaya Channel
suspended matter 2009-2011
1st terrace bluff profiles
Kurungnakh Island (third terrace, i.e. ice complex deposits)

Figure 1.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July/Aug 2010
late May 2011
late June/early July 2011
TSM
[mg/L]
POC
[mg/L]
POC:PN$_{at}$
δ^{13}C
[‰ VPDB]
Δ^{14}C
[‰]
A
B
C
D
E
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M
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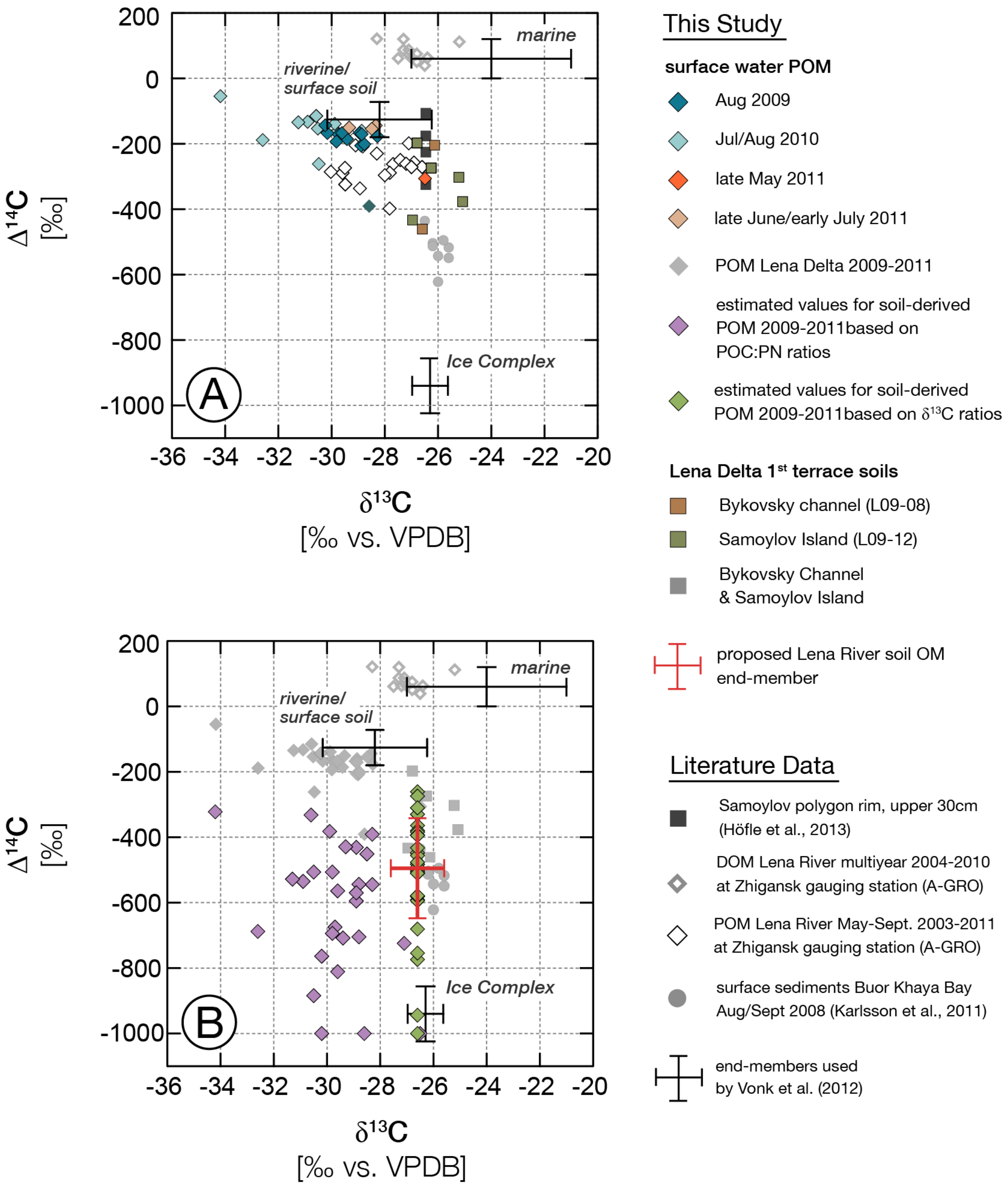
1

2 Figure 2.

3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0
-200
-400
-600
-800
-1000
-36 -34 -32 -30 -28 -26 -24 -22 -20
Δ14C [‰]
δ13C [‰ vs. VPDB]
marine
riverine/ surface soil
Ice Complex
A
B
This Study
surface water POM
Aug 2009
Jul/Aug 2010
late May 2011
late June/early July 2011
POM Lena Delta 2009-2011
estimated values for soil-derived POM 2009-2011based on POC:PN ratios
estimated values for soil-derived POM 2009-2011based on δ13C ratios
Lena Delta 1st terrace soils
Bykovsky channel (L09-08)
Samoylov Island (L09-12)
Bykovsky Channel & Samoylov Island
proposed Lena River soil OM end-member
Literature Data
Samoylov polygon rim, upper 30cm (Höfle et al., 2013)
DOM Lena River multiyear 2004-2010 at Zhigansk gauging station (A-GRO)
POM Lena River May-Sept. 2003-2011 at Zhigansk gauging station (A-GRO)
surface sediments Buor Khaya Bay Aug/Sept 2008 (Karlsson et al., 2011)
end-members used by Vonk et al. (2012)

Figure 3.

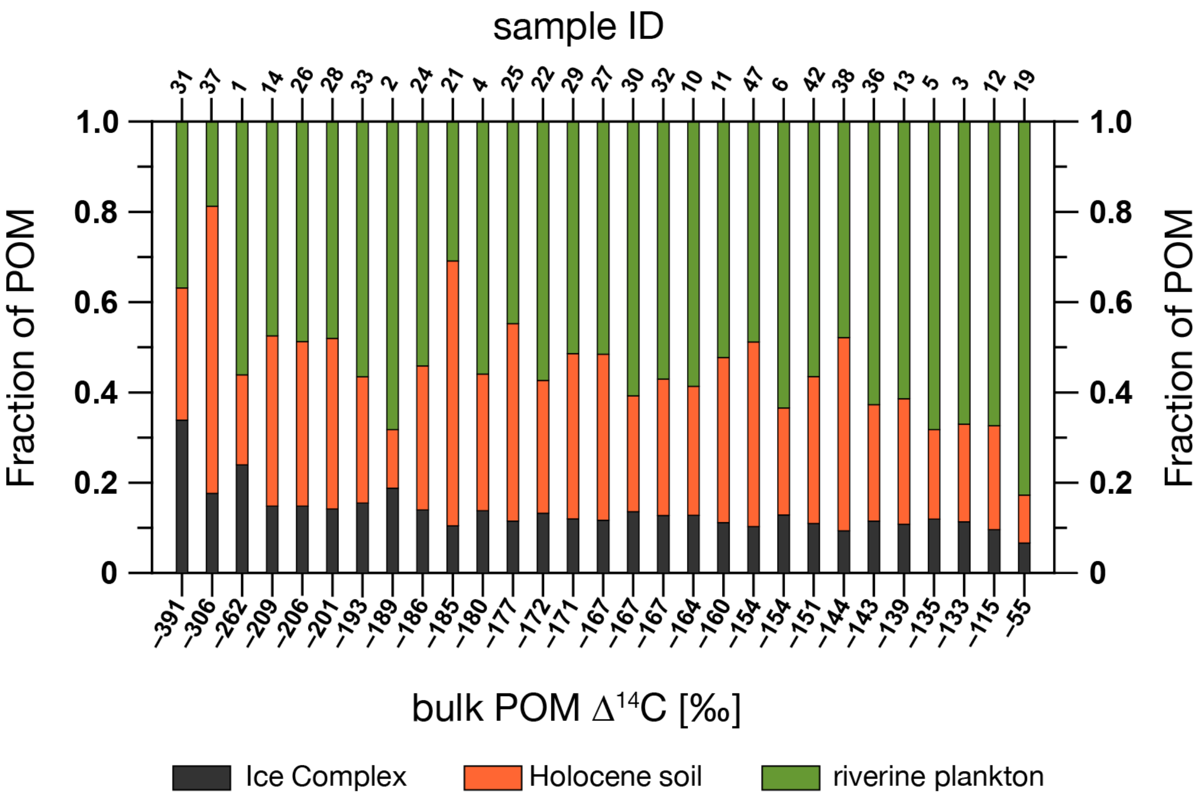
sample ID
31 37 1 14 26 28 33 2 24 21 4 25 22 29 27 30 32 10 11 47 6 42 38 36 13 5 3 12 19
Fraction of POM
1.0
0.8
0.6
0.4
0.2
0
–391 –306 –262 –209 –206 –201 –193 –189 –186 –185 –180 –177 –172 –171 –167 –167 –167 –164 –160 –154 –154 –151 –144 –143 –139 –135 –133 –115 –55
bulk POM Δ14C [‰]
Ice Complex
Holocene soil
riverine plankton

Figure 4.