

RESEARCH ARTICLE

10.1002/2017JG003858

Key Points:

- Spatial differences in OC concentrations and export due to catchment topography and regional hydroclimate in Lena River basin
- Sources of DOM were fresh plant material and SOM with seasonal patterns reflecting differences in water flow pathways
- Sources of POM were aquatic primary production and SOM and were related to topography

Supporting Information:

- Supporting Information S1
- Table S1
- Table S2

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Citation:

Kutscher, L., C.-M. Mörtz, D. Porcelli, C. Hirst, T. C. Maximov, R. E. Petrov, and P. S. Andersson (2017), Spatial variation in concentration and sources of organic carbon in the Lena River, Siberia, *J. Geophys. Res. Biogeosci.*, 122, 1999–2016, doi:10.1002/2017JG003858.

Received 23 MAR 2017

Accepted 21 JUL 2017

Accepted article online 28 JUL 2017

Published online 10 AUG 2017

Spatial variation in concentration and sources of organic carbon in the Lena River, Siberia

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Abstract Global warming in permafrost areas is expected to change fluxes of riverine organic carbon (OC) to the Arctic Ocean. Here OC concentrations, stable carbon isotope signatures ($\delta^{13}\text{C}$), and carbon-nitrogen ratios (C/N) are presented from 22 sampling stations in the Lena River and 40 of its tributaries. Sampling was conducted during two expeditions: the first in July 2012 in the south and southeastern region and the second in June 2013 in the northern region of the Lena basin. The data showed significant spatial differences in concentrations and major sources of OC. Mean subcatchment slopes were correlated with OC concentrations, implying that mountainous areas in general had lower concentrations than lowland areas. $\delta^{13}\text{C}$ and C/N data from tributaries originating in mountainous areas indicated that both dissolved and particulate OC (DOC and POC) were mainly derived from soil organic matter (SOM). In contrast, tributaries originating in lowland areas had larger contributions from fresh vegetation to DOC, while aquatically produced OC was the major source of POC. We suggest that these differences in dominant sources indicated differences in dominant flow pathways. Tributaries with larger influence of fresh vegetation probably had surficial flow pathways, while tributaries with more SOM influence had deeper water flow pathways. Thus, the future export of OC to the Arctic Ocean will likely be controlled by changes in spatial patterns in hydroclimatology and the depth of the active layers influencing the dominant water flow pathways in Arctic river basins.

1. Introduction

Permafrost soils are considered to be among the largest terrestrial reservoirs of carbon [Hugelius *et al.*, 2014]. During the last century, large areas underlain by permafrost in the Northern Hemisphere have experienced environmental changes due to climate warming. Permafrost temperatures have risen [Romanovsky *et al.*, 2010], the active layer has become deeper during summer months [Frey and McClelland, 2009; Zhang *et al.*, 2005], and freshwater runoff from rivers to the Arctic Ocean has increased [Costard *et al.*, 2007; Peterson *et al.*, 2002; Rachold *et al.*, 1996; Shiklomanov and Lammers, 2009]. Some of the largest Arctic Rivers have also had increased sediment export [Rachold *et al.*, 1996]. According to climate models, climate warming in permafrost regions will continue during the next century and the Arctic is predicted to experience one of the largest temperature increases in the world [Collins *et al.*, 2013]. It is expected that permafrost thawing will change fluxes of natural organic matter (NOM) from the terrestrial environment to freshwater and marine systems due to changes in discharge, transport pathways, residence times in soils and water, plant species composition, and increased erosion [Costard *et al.*, 2007; Frey and Smith, 2005; Frey and McClelland, 2009; Guo *et al.*, 2007; Neff and Hooper, 2002; Striegl *et al.*, 2005]. However, there is no consensus regarding the direction of these changes [Frey and McClelland, 2009]. Whatever the direction of changes, it will ultimately have implications for the carbon cycling and ecosystem functioning in Arctic freshwater systems as well as in the Arctic Ocean [Frey and McClelland, 2009; Keller *et al.*, 2010], since NOM plays an important role in the aquatic food web as nutrient and energy source as well as transport media of essential metals and dissolved nutrients [Jansson *et al.*, 2007; Tipping, 2002].

Riverine NOM is a terrestrially (allochthonous) or aquatically (autochthonous) produced heterogeneous mixture of organic compounds with a generally complex structure containing carbon as its major constituent [Finlay and Kendall, 2008; Lam *et al.*, 2007]. Due to the high content of carbon, NOM is often equated with organic carbon (OC) and can be divided into two size fractions: dissolved organic carbon (DOC) and

particulate organic carbon (POC). DOC in stream ecosystems is generally derived from terrestrial sources, e.g., litterfall and soil leachate, while POC primarily is derived from eroded soil and sedimentary rocks as well as from autochthonous sources, e.g., macrophytes, algae, and heterotrophic bacteria [Galy *et al.*, 2015; Meybeck, 1993]. However, the division between DOC and POC is arbitrary, since DOC can flocculate and form POC; also, both DOC and POC can be mineralized along river networks [del Giorgio and Pace, 2008; Drake *et al.*, 2015; Kerner *et al.*, 2003; Meybeck, 1993].

Several different factors controlling OC concentrations in rivers have been suggested. Mean OC concentrations have been correlated to spatial variables such as topography and landscape characteristics, e.g., amount of upstream wetland cover, lake cover, and average channel slope [Eckhardt and Moore, 1990; Mulholland, 1997, 2003]. Factors controlling temporal DOC dynamics have been shown to include discharge [Eckhardt and Moore, 1990; Winterdahl *et al.*, 2014], soil temperature, local climate (air temperature and precipitation), and antecedent flow [Winterdahl *et al.*, 2011]. OC concentrations in rivers often peak in connection with high-discharge events; this is commonly explained to be an effect of shifting flow pathways from deeper soil horizons to shallower organic-rich soil layers [Mulholland, 2003]. For example, in rivers in permafrost areas and in many boreal rivers, OC concentrations often increase during snowmelt and decrease with decreasing flow so that the lowest concentrations commonly occur during base flow [Holmes *et al.*, 2012; Le Fouest *et al.*, 2013; Raymond *et al.*, 2007; Winterdahl *et al.*, 2014]. A warming climate is expected to shift dominating flow paths to deeper soil layers [Frey and McClelland, 2009]. However, the subsurface hydrology in permafrost is not fully understood [Watson *et al.*, 2013; Woo *et al.*, 2008] and limits our understanding of element transport from areas underlain by permafrost.

For a better understanding of the influence of permafrost thawing on export and fluxes of riverine carbon it is necessary to unravel the main contributing areas and sources of OC in permafrost regions. Among the world's deepest layers of permafrost are found in the Lena River drainage basin, in central Siberia, where the major part of the basin is underlain by continuous permafrost [Chevychev and Bosikov, 2010]. It is the ninth largest drainage basin in the world and has the largest transport of OC to the Arctic Ocean [Holmes *et al.*, 2012; Le Fouest *et al.*, 2013]. Holmes *et al.* [2012] estimate that the annual export from the six largest Arctic rivers draining directly into the Arctic Ocean is 25 Tg DOC year⁻¹, of which the Lena River contributes approximately 5.8 Tg DOC year⁻¹ and 0.8 Tg POC year⁻¹ [Holmes *et al.*, 2012; Le Fouest *et al.*, 2013; McClelland *et al.*, 2016; Raymond *et al.*, 2007]. The export of OC from the Lena varies seasonally and OC concentrations peak during snowmelt in the beginning of June and then decrease to reach annual lows during winter base flow [Amon *et al.*, 2012; Holmes *et al.*, 2012; Le Fouest *et al.*, 2013; McClelland *et al.*, 2016; Raymond *et al.*, 2007]. Previous studies in the Lena River indicate that the dominant sources of DOC in the main channel are relatively young soil and plant material [Amon *et al.*, 2012; Lara *et al.*, 1998; Lobbes *et al.*, 2000]. POC on the other hand is suggested to be older degraded soil organic matter (SOM) [Lobbes *et al.*, 2000; McClelland *et al.*, 2016]. The amount of aquatically produced OC in the Lena River is believed to be low [Lobbes *et al.*, 2000; McClelland *et al.*, 2016; Sorokin and Sorokin, 1996]. The contribution of old, highly biologically available OC from thawing permafrost to fluvial networks increase aquatic microbial metabolism and CO₂ emissions within Arctic drainage basins [Mann *et al.*, 2015]. This permafrost-derived OC is also suggested to result in loss of ancient OC in river networks and a selective export of younger OC to the oceans [Mann *et al.*, 2015; Vonk *et al.*, 2010].

Stable carbon isotopes values ($\delta^{13}\text{C}$) and carbon/nitrogen atomic ratios (C/N) can be used to identify sources of freshwater OC, since these could be indicative of different sources [Cleveland and Liptzin, 2007; Elser *et al.*, 2000; Finlay and Kendall, 2008; McGroddy *et al.*, 2004; Sterner and Elser, 2002]. Freshwater primary producers assimilate carbon from CO₂, which can originate from a variety of sources and result in a wide range in $\delta^{13}\text{C}$ signatures of aquatically produced OC (−47 to −8‰) [Farquhar *et al.*, 1989; Finlay and Kendall, 2008]. In addition, the isotope fractionation factor of freshwater primary production is more varied than in terrestrial systems [O'Leary, 1988]. C/N ratios in aquatically produced material are often narrow in range (4–15) [Elser *et al.*, 2000; Finlay and Kendall, 2008], which could be due to the low supply of nitrogen in aquatic systems [Sardans *et al.*, 2012]. In contrast, terrestrial sources generally have a narrower range in $\delta^{13}\text{C}$ and a wider range in C/N [Elser *et al.*, 2000; Finlay and Kendall, 2008; McGroddy *et al.*, 2004; O'Leary, 1988]. Terrestrial plants mainly assimilate carbon from the atmosphere, which is a well-mixed reservoir with a $\delta^{13}\text{C}$ of −8‰. The isotope fractionation factor in C3 plants, the most common plant type [Ridge, 2002], is about −20‰, resulting in an average $\delta^{13}\text{C}$ of approximately −28‰ in C3 plants [Farquhar *et al.*, 1989; Finlay and Kendall, 2008; O'Leary, 1988].

C/N in plants and litter worldwide varies substantially and could range from around 15 to several hundreds, but reported mean values of C/N are between 36 and 66 [Elser *et al.*, 2000; Finlay and Kendall, 2008; McGroddy *et al.*, 2004]. It is commonly observed that $\delta^{13}\text{C}$ increase while C/N decrease with depth in mineral soils in boreal and arctic ecosystems [Boström *et al.*, 2007; Cleveland and Liptzin, 2007; Kaiser *et al.*, 2007; McGroddy *et al.*, 2004; Palmtag *et al.*, 2015; Zech *et al.*, 2007] and is believed to be due to isotope fractionation and selective removal during SOM decomposition [Boström *et al.*, 2007; Ehleringer *et al.*, 2000; Palmtag *et al.*, 2015].

Previous studies of OC in the Lena River have mainly focused on the main channel of the river and the delta [Amon *et al.*, 2012; Cooper *et al.*, 2008; Dudarev *et al.*, 2006; Holmes *et al.*, 2012; Karlsson, 2015; Lara *et al.*, 1998; Le Fouest *et al.*, 2013; Lobbes *et al.*, 2000; Pipko *et al.*, 2010; Raymond *et al.*, 2007; Semiletov *et al.*, 2011]. OC data from tributaries of the Lena River are scarce and include only the largest subbasins and then only for a few occasions [Kuzmin *et al.*, 2009; Lara *et al.*, 1998]. Spatially defined OC fluxes from the terrestrial environment to the Lena River are therefore not well understood, limiting our ability to adequately predict future changes in OC concentrations and fluxes to the Arctic Ocean.

In this study, we present a spatial data set of OC concentrations, C/N ratios, and $\delta^{13}\text{C}$ values of DOC and POC, from the Lena River main channel and 40 of its tributaries sampled during two field expeditions conducted in July 2012 and June 2013. These catchments cover different landscape types with varying vegetation, lithology, and hydrology, as well as permafrost depth and distribution. The specific objectives of this study were to enhance our understanding of spatial variability in OC concentration and sources in the Lena basin by (i) quantifying spatial patterns in OC concentrations and composition, (ii) making a preliminary spatial export estimate of OC from the largest subbasins of Lena, and (iii) identifying major sources of OC in rivers in the Lena basin.

2. Methods

2.1. Study Area

The area of the Lena River basin is about $2.4 \times 10^6 \text{ km}^2$ and extends from its southernmost headwater at latitude $52\text{--}53^\circ\text{N}$ in the mountain areas of Baikal, Stanovoi, and Dzhugdzhur to its delta at the shallow shelf in the Laptev Sea at 73°N (Figure 1a). The eastern part of the drainage basin is dominated by the mountain ranges Verkhoyansk and Suntar-Khaiata, with its easternmost headwaters at longitude 141°E . The central and northwestern areas are characterized by plains and plateaus, which make the western water divide diffuse in comparison to the mountain ranges in the south and the east. The westernmost headwaters extend to the Viliuskoe plateau as far west as longitude 104°E .

The Lena River basin is characterized by a continental climate, with long, cold winters and short, warm summers [Chevychev and Bosikov, 2010]. The mean annual air temperatures are in general low and decrease with increasing latitudes [Fedorov *et al.*, 2014], from approximately -5°C in the southern parts of the Lena basin, -9°C at Viliusk in the central part of the basin, and around -13°C in the north at Kiuisur [Fedorov *et al.*, 2014]. The major part of the Lena basin is underlain by continuous permafrost, except for patches of discontinuous and sporadic permafrost in the southern parts of the basin and some areas along the main channel of the Lena River [Chevychev and Bosikov, 2010]. In the central part of the Lena basin, the Central Siberian Plateau (CSP), the deepest layers of permafrost are found [Chevychev and Bosikov, 2010]. Both air and permafrost temperatures in the basin show increasing trends since the late 1950s [Romanovsky *et al.*, 2007], which have resulted in an estimated deepening of the active layer of 0.35 cm/year [Frey and McClelland, 2009].

The permafrost is primarily overlaid by podzols and leptosols in the south, cambisols in the central basin, and gleysols in the northernmost region [Stolbovoi and McCallum, 2002]. The vegetation is divided into three zones, where the most productive forest is found in the southernmost area. Most of the basin is dominated by larch forest, while alpine regions have bare rock terrain [Chevychev and Bosikov, 2010].

The mean annual precipitation in the southern and southwestern part of the Lena basin is $\sim 350\text{--}500 \text{ mm/year}$ and $\sim 200\text{--}250 \text{ mm/year}$ elsewhere [Chevychev and Bosikov, 2010]. Monthly discharge has been measured at several hydrologic stations since the 1930s by the Russian Hydrometeorological Services. These data show a strong seasonal flow regime in the Lena River and its tributaries, with the highest discharge peak at snowmelt in May to June (Figure 1c) [Ye *et al.*, 2009]. Discharge gradually decreases after spring flood and

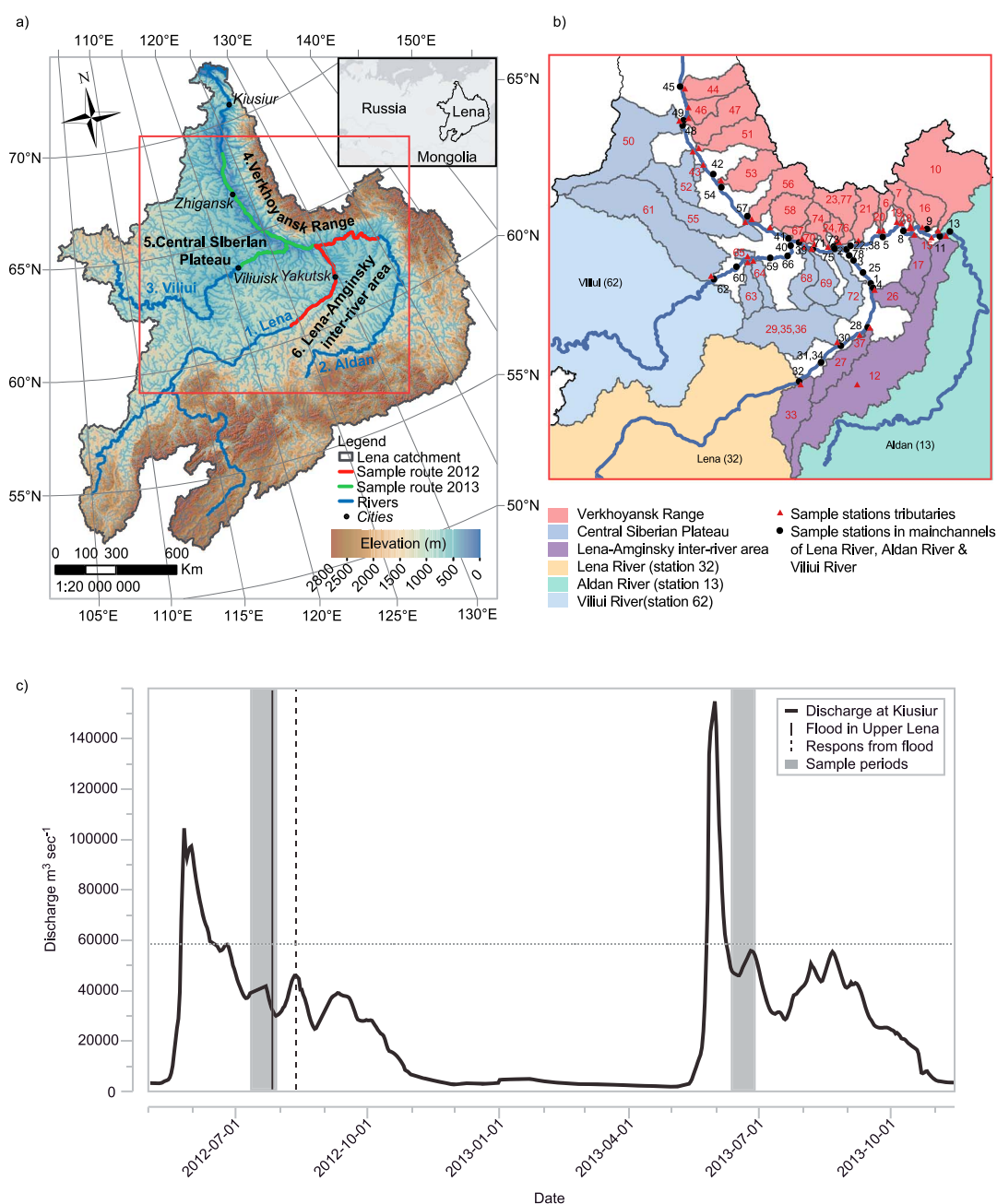


Figure 1. (a) Map of the Lena River basin and sample routes during 2012 (red) and 2013 (green). Number 1–6 indicate separation of collected samples in main channels (1–3) and tributaries (4–6). (b) Black dots show sample stations along the main channels and red triangles show sample stations in tributaries. Colored areas indicate the catchment area of sampled tributaries. (c) Lena River discharge hydrograph at the hydrologic gauging station at Kiuisur during 2012 and 2013. Grey areas show the sampling periods in 2012 and 2013. The black vertical line indicates the first sign of the flood close to the city of Sinsk in the Upper Lena and the dashed line shows the highest discharge peak from the flood at Kiuisur 17 days later.

reaches the annual low during winter. The average in-stream water travel time is 1–2 months from the uppermost regions of the basin to the outflow of Lena [Smith and Pavelsky, 2008; Ye *et al.*, 2003].

2.2. Field Methods and Sampling

River water samples were collected at 77 locations across the Lena River basin during two field expeditions conducted with the ships R/V Geokryolog and R/V Akademik in July 2012 and with R/V Merelotov in June 2013 (Figure 1 and supporting information Table S1). The first expedition covered the south and

southeastern region of the Lena basin, upstream of the confluence of Lena and Aldan rivers, and the second expedition covered the region downstream of Yakutsk. The samples were collected at 22 stations in the Lena River main channel, between the Toulba River (60.6°N) and Dzhardzhan River (68.7°N), and at 55 stations in tributaries (Figure 1 and Table S1). In most tributaries the outflow of the rivers were sampled, but in the largest tributaries (the Aldan and Viliui Rivers) additional samples were taken along their main channels. In total 40 different tributaries were sampled. Sampling was repeated in the second expedition in the outflow of Aldan and some of the other tributaries by the confluence of Aldan and Lena River as well as in the Lena River between Yakutsk and the outflow of Aldan. In addition to water samples, plant samples were collected at three different locations along the Lena River. At the end of the 2012 expedition a flood took place in the upstream areas of the Lena basin resulting in elevated water levels and large amounts of debris in the Lena River. The average discharge in the end of July at the gauging station Tabaga (close to Yakutsk) is generally around $10000 \text{ m}^3 \text{ s}^{-1}$, but at the flood in 2012 the discharge rose to almost $40,000 \text{ m}^3 \text{ s}^{-1}$, which is almost as high as the 2012 spring flood [Tananaev, 2016].

At each sample station, surface water was collected in 10 L low-density polyethylene (LDPE) bottles by grab sampling. Water temperature, pH, and conductivity were measured in situ with an YSI 556 multiprobe system, with accuracies at $\pm 0.15^\circ\text{C}$, ± 0.2 pH units, and $\pm 0.5\%$, respectively. These variables were also measured along a transect from sample station 73 (Figure 1b) to the eastern shoreline of the Lena main channel in 2013 to investigate mixing patterns within the river. Positions were recorded with a handheld GPS receiver.

Water samples were filtered through 25 mm precombusted (at 450°C for 2 h) glass fiber filters (Whatman® GF/F) with $0.7 \mu\text{m}$ pore size using a portable peristaltic pump. The mass of water pumped through the filters was recorded. The GF/F filters were then folded into packets of aluminum foil and stored in freezers until analysis of particulate carbon and nitrogen. Filtered water was stored in dark high-density polyethylene bottles for analyses of DOC and dissolved nitrogen. All bottles, vials, and filter equipment used during field work were prewashed with 10% HNO_3 and distilled water (MilliQ $0.1 \mu\text{m}$ filter). The filtered water samples collected in July 2012 were preserved with phosphoric acid (87%) and stored cool, while samples collected in June 2013 were stored in freezers until analyses. Plant samples were dried and packed in LDPE bags.

2.3. Analytical Methods

Concentrations of DOC were determined within 2 months by high-temperature catalytic oxidation (Shimadzu TOC-VCPH) at the Department of Environmental Science and Analytical Chemistry, Stockholm University (SU). Two internal controls were used: phenanthroline and humic lake water with known carbon concentrations (18–19 mg/l). The total uncertainty interval based on laboratory intercomparisons was less than $\pm 10\%$.

Dissolved water samples were freeze-dried at -110°C and then weighed and put into tin (2012) or silver capsules (2013). A small aliquot of 2 M HCl (PA grade) was added to remove any inorganic carbon in the samples, and thereafter the samples were left in an oven at 60°C for 1 h to allow evaporation of HCl. GF/F filters containing the POC and particulate organic nitrogen (assuming that inorganic N was low and negligible) were placed in desiccators together with a beaker with HCl (35%) overnight to remove inorganic carbon. The filters were then dried at 60°C and packed in silver capsules. The dried plant material was also placed into silver capsules. These prepared samples were measured with an isotope ratio mass spectrometer (IRMS) (Finnigan® Delta V advantage) at the stable isotope laboratory at SU. The mass spectrometer was connected to a CarloErba® NC2500 elemental analyzer through a ConFloIV open split interface.

The organic carbon and total nitrogen mass received from the IRMS measurements were used to calculate POC concentrations and C/N atomic ratios. The concentrations of POC were calculated from the mass of particulate carbon on the filters and the weighted water pumped through the GF/F filters. For the C/N ratios it was assumed that the total N was organic nitrogen. The total accuracy and precision of the C/N atomic ratios and POC concentrations were lower than 3%.

Isotope values of DOC, POC, and plant material were reported in per mil (‰), $\delta^{13}\text{C}$, relative Pee Dee Belemnite (PDB):

$$\delta = \left(\frac{R_{\text{Sample}}}{R_{\text{Std}}} - 1 \right) \cdot 1000, \quad (1)$$

where R_{Sample} and R_{Std} are the $^{13}\text{C}/^{12}\text{C}$ ratios in the sample and in the standard, respectively. The standards

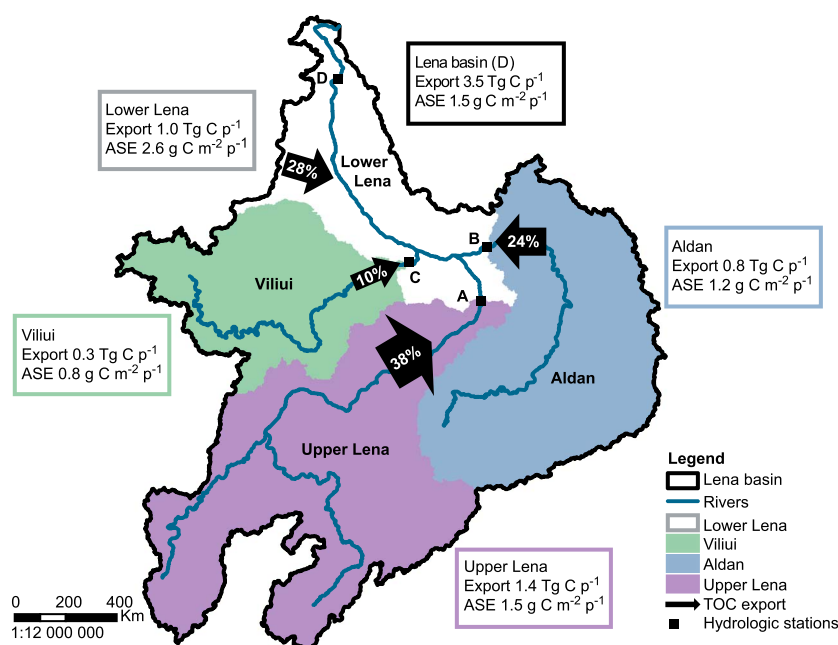


Figure 2. Export and area specific export (ASE) of total organic carbon (TOC) for the period June and July (p) from the Upper Lena, the Aldan River, the Viliui River, and the Lower Lena based on carbon concentrations and discharge data from the hydrologic gauging stations at Lena at Tabaga (A), Aldan at Verkhoyanski' Perevos (B), Viliui at Hatyrik-Homo (C), and Lena at Kiusiur (D). Black arrows indicate the fractional export from each subbasin of the total export from the Lena River outflow at station Kiusiur (D).

used for carbon and nitrogen calibrations in the IRMS were internal standards with known isotope compositions relative to PDB and AIR. The accuracy and precision of $\delta^{13}\text{C}$ measurements were less than $\pm 0.2\text{‰}$.

2.4. Export Calculations

A first preliminary estimate of the export and area specific export of carbon for the period June to July from different source areas was calculated from concentrations of DOC and POC from the main channels of Upper Lena, Aldan River, Viliui River and Lower Lena, and long-term monthly discharge data from the hydrologic stations Lena at Tabaga (A), Aldan at Verkhoyanski' Perevos (B), Viliui at Hatyrik-Homo (C) and Lena at Kiusiur (D) (Figure 2 and Table 1). The discharge data used in the calculations were downloaded from the Russian Hydrometeorological Services—R-ArcticNet (<http://www.r-arcticnet.sr.unh.edu/v4.0/index.html>), ArcticRIMS (<http://rims.unh.edu/data.shtml>), and Arctic Great Rivers Observatory (NSF-1107774) (<http://arctic.greaterrivers.org/data.html>), which only consist of discharge data from the Lena main channel and a few of the sampled tributaries. The hydrologic station in Kiusiur was taken to represent the outflow of the Lena basin, and the mean OC concentrations from the two northernmost sample stations 45 and 48 (Table S1) were used to estimate the total organic carbon (TOC) export and area specific export in June and July to the Arctic Ocean. The average TOC concentration for these two samples (990 μM) was similar to the annual mean value of the Arctic-GRO data (970 μM) collected between 2003 and 2013 at Zhigansk. In export calculations from subbasins, mean OC concentrations from samples collected in the main channels of the Aldan River (station 8, 13, and 38 from July 2012 and June 2013) and the Viliui River (station 59, 60, 62, and 66 from June 2013) were used. In the Upper Lena, the mean OC concentrations from stations 28, 30, and 31 collected in July 2012 were used (Table S1). Samples collected after the flood in the Upper Lena were excluded in the export estimations, since the flood in 2012 was substantially larger than average discharge for July [Tananaev, 2016], and we do not have data to assess how exceptional floods of this magnitude are in this area in July. The remaining TOC export and area specific export were assigned to the northern region of the basin, here called the Lower Lena (Figure 2).

POC concentrations commonly increase with depth in rivers [McClelland *et al.*, 2016]. In this study only surface water was sampled and the TOC concentrations and export could therefore have been

Table 1. Carbon Export and Area Specific Export (ASE) Over the Period June and July From Areas Upstream of Hydrologic Stations in the Tributaries Aldan River and Viliui River As Well As in the Upstream and Downstream Lena Main Channel^a

	Upper Lena	Aldan	Viliui	Lena at Kiusiur	Lower Lena
Name hydrologic station	Lena at Tabaga	Aldan at Verkhoyanski' Perevos	Viliui at Hatyrik-Homo ^b	Lena at Kiusiur	Remaining area
Hydrologic station code ^c	3,042/6,147	3,229/6,236	3,329/6,266	3,821/6,342	
Latitude (DD)	61.83	63.32	63.95	70.68	
Longitude (DD)	129.6	132.02	124.83	127.39	
Data period discharge	1936–2011	1942–1999	1936–1998	1936–2015	
Drainage area (km ²)	897,000	696,000	452,000	2,430,000	385,000
Drainage area (%)	37	29	19	100	15
Mean Q (m ³ s ⁻¹) ^d	19,355	14,921	4,950	56,259	17,033
Mean Q (%) ^d	34	27	9	100	30
Specific Q (l s ⁻¹ km ⁻²) ^d	22.6	21.4	11.0	23.2	44.2
E _{TOC} (Tg period ⁻¹) ^e	1.35 ± 0.4	0.84 ± 0.2	0.34 ± 0.09	3.54 ± 0.5	1.01 ± 0.3
E _{TOC} (%)	38.1	23.8	9.6	100	28.5
E _{DOC} (Tg period ⁻¹) ^e	1.20	0.79	0.33	3.46	1.14
E _{POC} (Tg period ⁻¹) ^e	0.15	0.05	0.01	0.08	−0.13
ASE _{TOC} (g m ⁻² period ⁻¹) ^f	1.5	1.2	0.8	1.5	2.6

^aStations are listed from upstream to downstream.^bReanalyzed Q at this site by sources.^cStation ArcticRIMS code/R-ArcticNetID.^dData based on discharge (Q) downloaded from the webpages of ArcticRIMS, R-ArcticNET, and Arctic Great Rivers Observatory (NSF-1107774).^e $E = Q \times C$, where E represents the export of OC, Q is the water discharge, and C is the concentrations OC. The data are based on mean monthly Q in June and July for above mentioned data periods and concentration for the months June 2012 and July 2013. The uncertainty in the export is estimated from a Monte Carlo simulation given as range; see section 2.^f $ASE_{TOC} = E_{TOC} A^{-1}$, where the ASE_{TOC} is the area specific export of TOC, i.e., the export per unit area (A).

underestimated. What the differences are between the surface and deeper waters in the rivers is not clear and so are the uncertainties related to that. Other uncertainties associated with the export calculations were sampling in different regions during different months and years, which means that most of the stations were only sampled once. A Monte-Carlo analysis was therefore applied to estimate the uncertainties in the TOC export from the export equation in Table 1. Random numbers (200,000 normally distributed) for TOC concentrations and discharge values were generated assuming a total uncertainty of $\pm 20\%$ for both TOC concentrations and measured discharge. The error estimates are reported in Table 1.

2.5. Slope Calculation

The catchment specific mean slope for each tributary and stations 32, 13, and 62 (representing the upper regions of the Lena, Aldan, and Viliui catchments) (Figure 1b) was estimated with the Slope tool in the Spatial Analyst toolbox in ArcGIS 10, based on the digital elevation model (DEM) GTOPO30 (<https://lta.cr.usgs.gov/GTOPO30>) with horizontal grid spacing of 30 arc second.

2.6. Grouping of Data

Spatial patterns of OC were studied by dividing the samples into groups based on geographic region (Figure 1a and Table S1): (1) the main channel of the Lena River, (2) the main channel of the Aldan River, (3) the main channel of the Viliui River, (4) tributaries in the mountain range of Verkhoyansk (VER), (5) tributaries in the lowland area of Central Siberian Plateau (CSP), and (6) tributaries in the Lena-Amginsky interriver area (LAIRA). The Aldan and Viliui Rivers were treated as separate groups due to their large catchment sizes and runoff to the Lena.

3. Results

3.1. Organic Carbon Concentrations

The total range in DOC concentrations for all sample stations was 90 to 3400 μM , with a median concentration of 900 μM (Table 2). We observed spatial variations in OC concentrations, where tributaries with headwaters in VER had much lower median DOC concentrations, 260 μM , compared to the rest of the study area (Table 2 and Figure 3a). Conversely, tributaries with headwaters in the CSP and the main channel of the Viliui

Table 2. Distribution of Stream Water Chemistry Divided Into Groups and Plant Sample Data

	Quantiles	T_{Air} (°C)	$T_{\text{H}_2\text{O}}$ (°C)	pH	Cond ($\mu\text{S}/\text{cm}^\text{C}$)	DOC (μM)	$\delta^{13}\text{C}_{\text{DOM}}$ (‰)	C/N _{DOM} (Atomic)	POC (μM)	$\delta^{13}\text{C}_{\text{POM}}$ (‰)	C/N _{POM} (Atomic)
Water samples											
All water samples	Median	23	17.9	7.2	112	900	−27.2	28	38	−29.2	10
	Q ₁	21	15.9	7	87	540	−27.5	18	22	−32.6	9
	Q ₂	25	19.3	7.5	159	1100	−26.6	34	70	−27.1	11
	Minimum	13	8.6	6.3	42	90	−28.4	4	6	−36.1	5
	Maximum	31	23.8	9.4	346	3400	−25.7	41	640	−24.3	17
Lena main channel	Median	24	16.5	7.2	101	970	−27.1	35	42	−27.1	11
	Q ₁	22	15.8	6.9	85	840	−27.4	20	20	−30.9	9
	Q ₂	26	18.2	7.5	140	1000	−26.6	38	77	−26.0	12
	Minimum	13	12.0	6.5	69	590	−27.8	11	10	−34.0	7
	Maximum	31	19.8	7.8	174	1300	−26.2	41	640	−25.3	15
Aldan main channel	Median	20	18.4	7.3	92	730	−26.8	21	33	−27.6	10
	Q ₁	19	17.9	7.1	86	700	−27.1	17	24	−29.0	7
	Q ₂	24	19.1	7.5	99	790	−26.4	30	64	−26.3	14
	Minimum	18	10.9	6.8	86	670	−27.2	17	19	−30.0	5
	Maximum	25	19.2	7.7	105	1100	−26.3	40	94	−25.3	17
Viliui main channel	Median	24	19.4	7.2	111	1000	−27.9	34	44	−33.4	10
	Q ₁	23	18.6	7.2	110	1000	−28.2	34	33	−34.3	9
	Q ₂	25	20.0	7.4	126	1100	−27.7	35	77	−32.0	11
	Minimum	21	17.9	7.2	110	1000	−28.4	34	31	−34.6	8
	Maximum	25	20.0	7.4	140	1200	−27.6	35	84	−31.1	12
Verkhoyansk tributaries (VER)	Median	23	16.3	7.1	111	260	−27.1	19	17	−27.6	10
	Q ₁	20	15.1	6.9	71	170	−27.5	15	9	−29.3	9
	Q ₂	25	18.3	7.4	172	450	−26.5	29	29	−27.0	11
	Minimum	16	11.2	6.3	49	90	−27.6	4	6	−32.9	8
	Maximum	28	22.3	7.8	244	1900	−25.7	38	116	−24.3	13
Central Siberian Plateau tributaries (CSP)	Median	23	19.4	7.4	147	1200	−27.6	29	61	−32.5	10
	Q ₁	22	18.2	7.1	57	930	−28.2	25	45	−34.6	7
	Q ₂	25	22.4	8.4	179	1800	−27.2	31	141	−30.8	11
	Minimum	17	16.2	6.7	42	780	−28.4	19	40	−36.1	5
	Maximum	30	23.8	9.4	346	2100	−26.6	39	215	−27.1	12
Lena-Amginsky interriver area tributaries (LAIRA)	Median	20	19.0	7.4	299	1100	−26.6	19	58	−33.1	8
	Q ₁	19	12.5	7.2	133	580	−26.8	16	39	−33.9	6
	Q ₂	23	21.6	8.2	310	1800	−26.4	28	100	−29.6	9
	Minimum	18	8.6	6.5	106	570	−27.5	13	38	−34.0	6
	Maximum	24	20.7	8.2	336	3400	−25.8	33	108	−28.5	9
Plant samples											
Plant material	Median									−29.6	36
	Betula									−25.4	42
	Carex									−29.6	26
	Larch									−29.9	36
	Salix 1									−29.8	39
	Salix 2									−27.0	33

River had substantially higher median concentrations, 1200 μM and 1000 μM , respectively (Table 2 and Figure 3a). We found a significant negative correlation between mean catchment slope and DOC concentrations with the catchments with higher mean slope having the lowest DOC concentrations (Figure 4a).

In samples collected in the Lena main channel, concentrations of DOC varied between 590 and 1300 μM , with a median concentration of 970 μM (Table 2 and Figure S1a). Samples collected between the outflow of the Toulba River (station 33) and Aldan River, i.e., in the upstream part of the Lena River, had a narrower range (840 to 1000 μM) compared to the samples collected further downstream (590 to 1300 μM) (Figure S1a). The large variations in DOC concentrations downstream from the confluence of Aldan River showed lower concentrations on the eastern side, the Verkhoyansk side, compared to those collected on the western side, the Central Siberian Plateau side (Figure S2a). The same pattern was also seen in conductivity, with higher conductivity on the CSP side (Figure S2b). In addition, observations from the transect across the Lena River

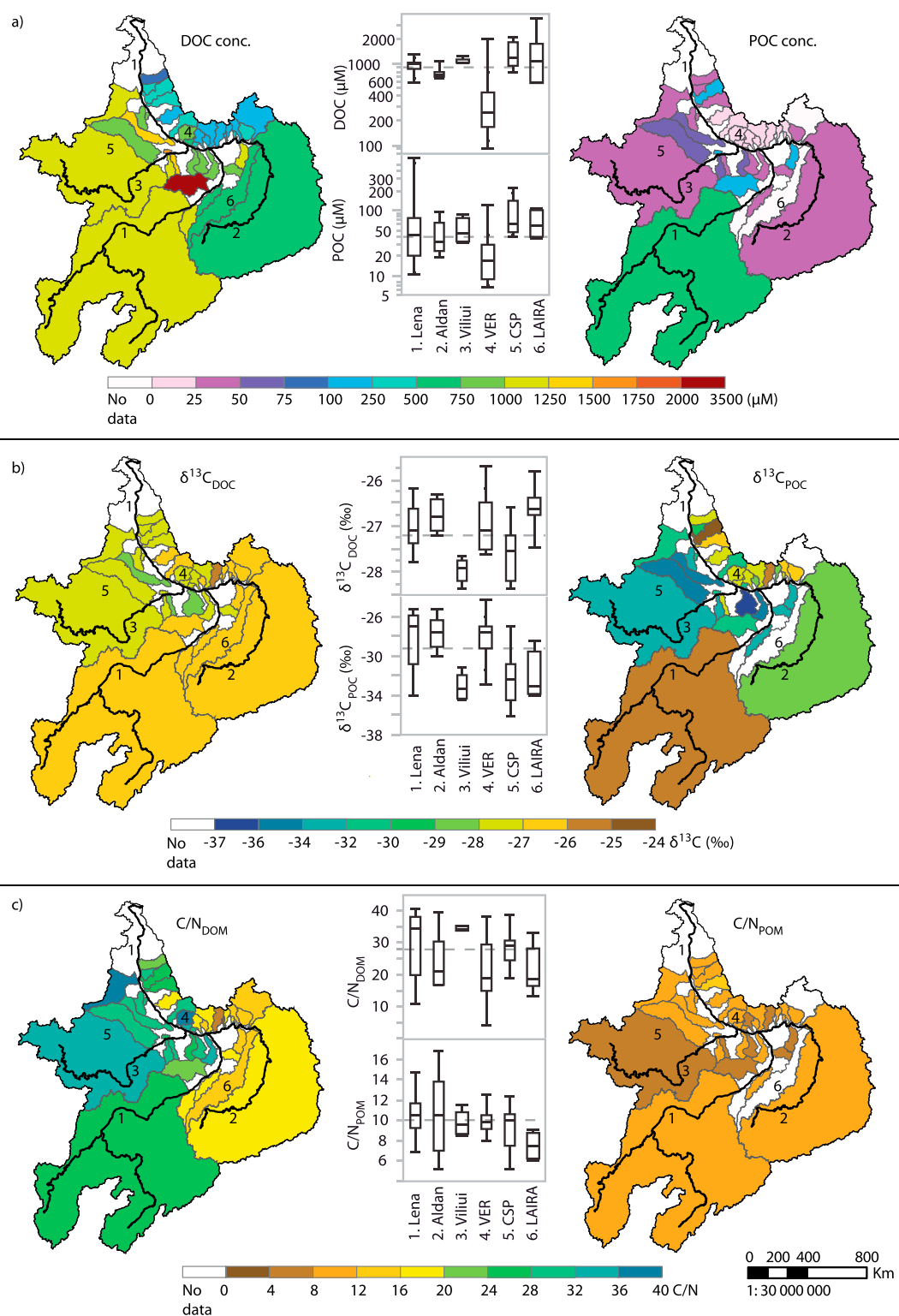


Figure 3. Maps showing (a) concentrations of DOC and POC, (b) $\delta^{13}\text{C}_{\text{DOC}}$ and $\delta^{13}\text{C}_{\text{POC}}$ values, and (c) $\text{C}/\text{N}_{\text{DOM}}$ and $\text{C}/\text{N}_{\text{POM}}$ atomic ratios at the outflow of sampled tributaries and the most upstream sample stations of the Lena River, Aldan River, and Viliui River. Boxplots show the distribution of DOC and POC (Figure 3a), $\delta^{13}\text{C}_{\text{DOC}}$ and $\delta^{13}\text{C}_{\text{POC}}$ (Figure 3b), and $\text{C}/\text{N}_{\text{DOM}}$ and $\text{C}/\text{N}_{\text{POM}}$ (Figure 3c) divided into source areas where 1–3 are the main channels of the Lena River, Aldan River, and Viliui River and 4–6 are groups of tributaries originating in Verkhoyansk (VER), Central Siberian Plateau (CSP), and the Lena-Amginsky interriver area (LAIRA). Dashed lines in boxplots indicate median values of all samples.

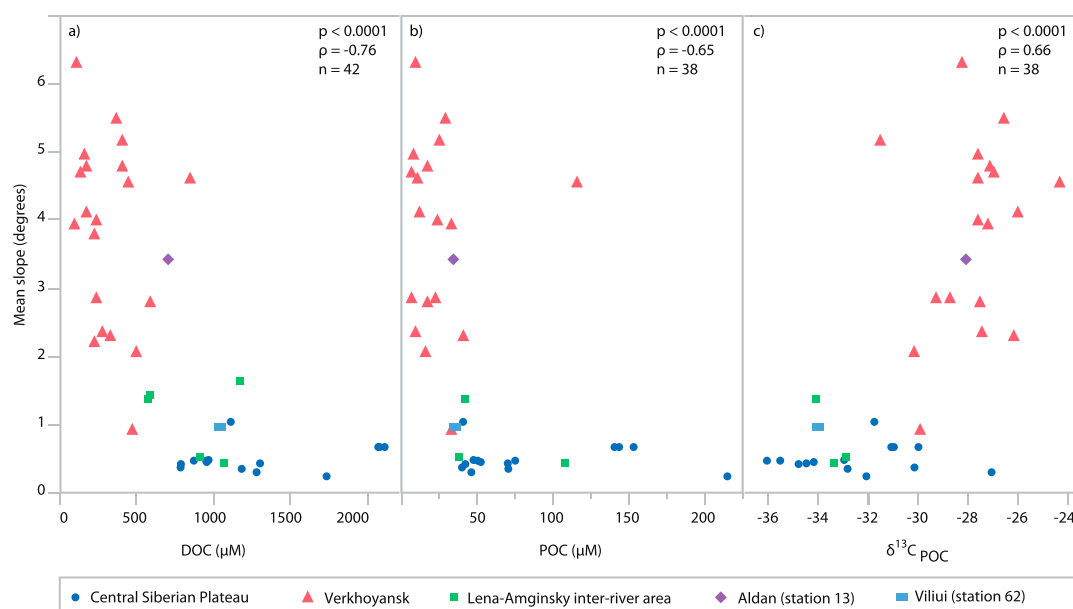


Figure 4. Mean catchment slope against (a) DOC concentrations and (b) POC concentrations and (c) $\delta^{13}\text{C}_{\text{POC}}$ in tributaries. Colors indicate the different source areas.

showed a gradient in conductivity, temperature, and pH in the main channel (Figure S3). Values for all three variables were higher on the western side than on the eastern side of the river.

The concentrations of POC were consistently much lower than those of DOC in the entire basin (Table 2 and Figure 3a) and were on average 5% of the TOC. The median POC concentration for all samples stations was $38 \mu\text{M}$ with a total range of 6 to $640 \mu\text{M}$ (Table 2). The POC concentrations in the main channel of Lena River varied between 10 and $640 \mu\text{M}$, with a median value of $42 \mu\text{M}$ (Table 2 and Figure S1b). This large variation was caused by two outliers (sample 32 and 34) collected after the flooding in the southern part of Lena River at the end of the expedition in July 2012. During the flood, POC concentrations increased with approximately $500 \mu\text{M}$ in the main channel of Lena River (Figure S1b). There was a significant difference in POC concentrations among groups, with a similar pattern as for DOC concentrations (Figure 3a), and a similar correlation between POC and slope (Figure 4b).

3.2. Transport and Export of TOC

The export of TOC in June and July from the Lena basin was estimated to be 3.54 Tg TOC , where 3.46 Tg was DOC (98%) and 0.08 Tg POC (2%) (Table 1 and Figure 2). The upstream areas from the hydrologic stations Lena at Tabaga (Upper Lena), Aldan at Verkhoyanski' Perevos, and Viliui at Hatyrik-Homo were estimated to contribute 1.35, 0.84, and 0.34 Tg TOC for the period, respectively. The remaining contribution from the lower northern region of the Lena River basin was 1.01 Tg TOC . The area specific export rate in June and July in the Lena River basin, at Kiusiur, was estimated to be 1.5 g m^{-2} . The rate from the Upper Lena was the same as in Kiusiur, while the rate from the Aldan River was lower, (1.2 g m^{-2}) and the Lower Lena was higher (2.6 g m^{-2}). The area specific export rate in Viliui River was considerably lower for the period compared to all other source regions (0.8 g m^{-2}).

3.3. $\delta^{13}\text{C}$ Values

The $\delta^{13}\text{C}$ values for DOC, $\delta^{13}\text{C}_{\text{DOC}}$, had a median value of -27.2‰ with a range of -28.4‰ to -25.7‰ (Table 2 and Figure 3b). There was a significant difference in $\delta^{13}\text{C}_{\text{DOC}}$ between sampling years, with lower $\delta^{13}\text{C}_{\text{DOC}}$ in the samples from June 2012 compared to samples from July 2013 (Figure 5a). There were also significant geographical differences in $\delta^{13}\text{C}_{\text{DOC}}$ among groups (Figure 3b). A comparison for each pair of groups showed that the Viliui River and the tributaries with headwater in CSP had lower $\delta^{13}\text{C}_{\text{DOC}}$ compared to other regions and main channels (Figure 3b). A negative correlation was found between $\delta^{13}\text{C}_{\text{DOC}}$ and

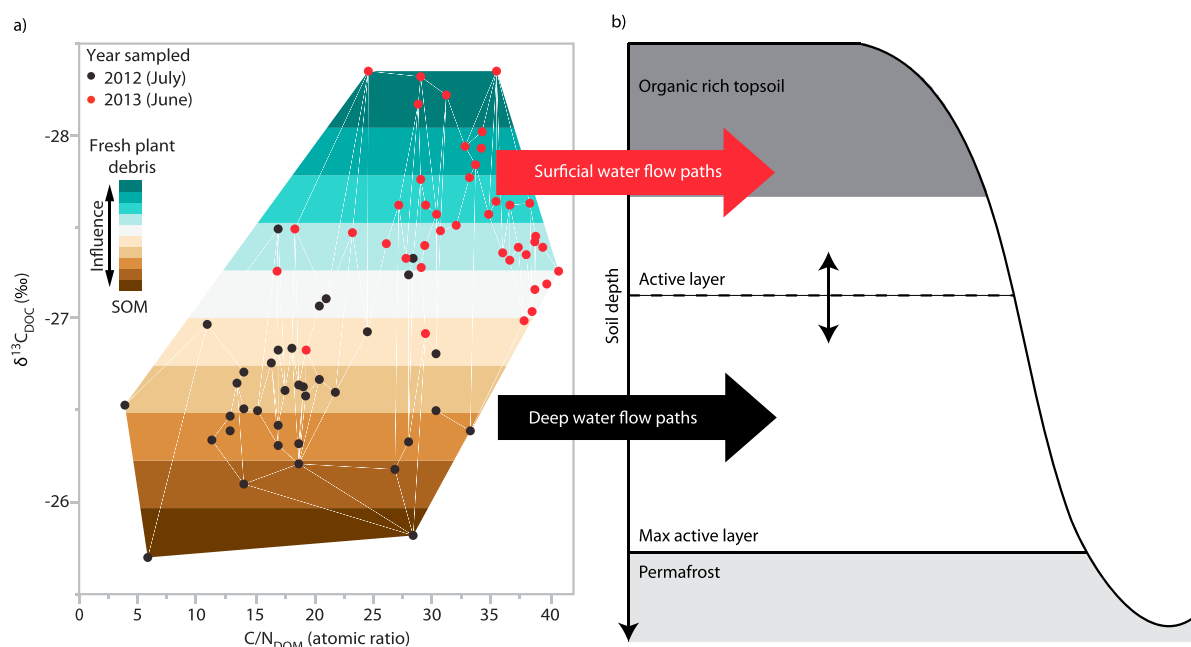


Figure 5. (a) Correlation between $\delta^{13}\text{C}_{\text{DOC}}$ and $\text{C}/\text{N}_{\text{DOM}}$, where red and black circles indicate the significant differences in $\delta^{13}\text{C}_{\text{DOC}}$ ($p < 0.0001$, $\chi^2 = 49.1$, $n = 77$) and $\text{C}/\text{N}_{\text{DOM}}$ ($p < 0.0001$, $\chi^2 = 39.7$, $n = 77$) between samples collected early in the summer (June 2013) and later in the summer (July 2012). Color range indicates gradient in primary organic matter source with relative importance of fresh plant material and SOM. (b) Schematic showing the influence of shifting active layer on water flow pathways.

latitude ($p < 0.0001$, $\rho = -0.66$), and positive correlation between $\delta^{13}\text{C}_{\text{DOC}}$ and longitude ($p < 0.0001$, $\rho = 0.52$). The combined data show that samples in the north and west of the study had the lowest $\delta^{13}\text{C}_{\text{DOC}}$.

The range in $\delta^{13}\text{C}$ values for POC, $\delta^{13}\text{C}_{\text{POC}}$ (-36.1‰ to -24.3‰) was larger than for $\delta^{13}\text{C}_{\text{DOC}}$ while the median value was lower (-29.2‰) (Table 2 and Figure 3b). There were significant differences in $\delta^{13}\text{C}_{\text{POC}}$ among geographical areas and the comparison for each pair showed that the Viliui River, CSP, and LAIRA tributaries had significantly lower $\delta^{13}\text{C}_{\text{POC}}$ compared to other groups (Figure 3b). The $\delta^{13}\text{C}_{\text{POC}}$ were lower in tributaries with low mean catchment slope compared to catchment with higher mean slope (Figure 4c). Additionally, a significant correlation was found between $\delta^{13}\text{C}_{\text{POC}}$ and water temperature (Figure S4). Water temperatures were also correlated with mean catchment slope ($p < 0.0003$, $\rho = -0.52$). Tributaries with lower mean catchment slope thus generally had lower $\delta^{13}\text{C}_{\text{POC}}$ and higher water temperatures, while tributaries originating in mountainous areas had higher $\delta^{13}\text{C}_{\text{POC}}$ and lower water temperatures.

The $\delta^{13}\text{C}$ values in plant samples, $\delta^{13}\text{C}_{\text{plant}}$, of *Carex*, *Betula*, *Larix*, and *Salix* varied between -29.9‰ and -25.4‰ , with a median value of -29.6‰ .

3.4. C/N Atomic Ratios

The median C/N ratios of the dissolved fraction of organic matter, $\text{C}/\text{N}_{\text{DOM}}$, was 28 and the range was large (4 to 41) with significantly lower values in samples collected in July 2012 compared to samples collected in June 2013 (Table 2 and Figure 5a). The same pattern was shown in the main channel of the Lena, with lower $\text{C}/\text{N}_{\text{DOM}}$ in the upstream area which was sampled in 2012 and higher $\text{C}/\text{N}_{\text{DOM}}$ in the downstream area that was sampled in 2013 (Figure S1e). There was also a significant difference among all groups, with higher median values in the Viliui (34) and CSP (29) compared to the other groups (19–21) (Table 1 and Figure 3c). Where $\text{C}/\text{N}_{\text{DOM}}$ were high, there was in general lower values of $\delta^{13}\text{C}_{\text{DOC}}$ ($p < 0.0001$, $\rho = -0.58$). There was positive correlation between $\text{C}/\text{N}_{\text{DOM}}$ and latitude ($p < 0.0001$, $\rho = 0.59$), and negative correlation between $\text{C}/\text{N}_{\text{DOM}}$ and longitude ($p < 0.0001$, $\rho = -0.55$). This means that the farther north and west the samples were collected in the Lena basin, the higher was the $\text{C}/\text{N}_{\text{DOM}}$, while samples collected in the southern and eastern regions had lower $\text{C}/\text{N}_{\text{DOM}}$.

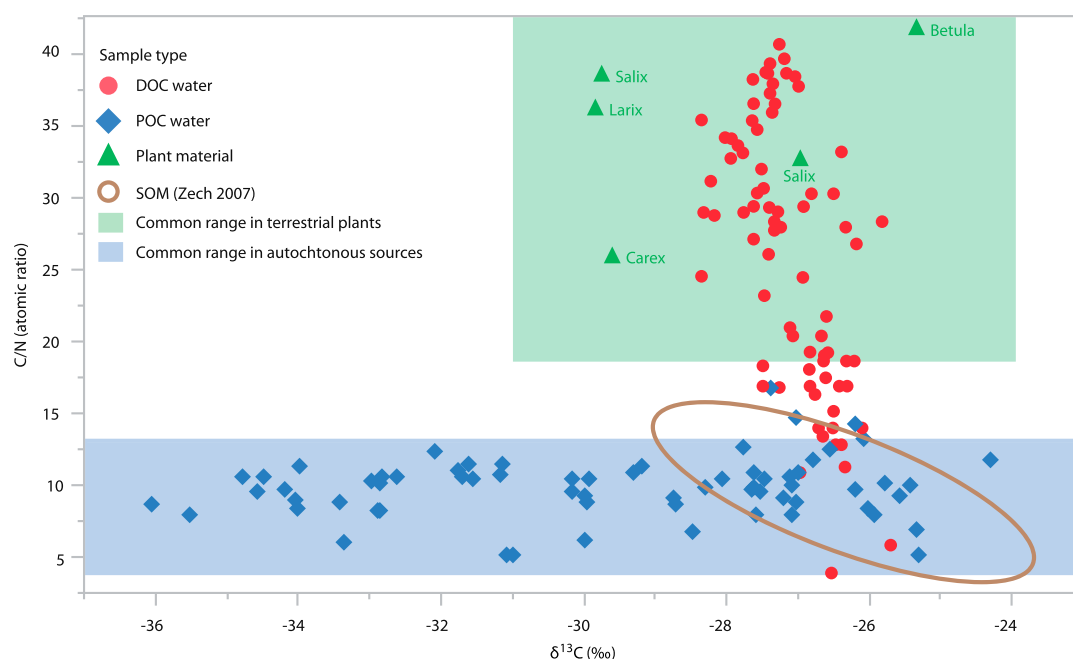


Figure 6. Plot of C/N and $\delta^{13}\text{C}$ in DOC and POC. The brown ellipse shows the range in C/N and $\delta^{13}\text{C}$ of soil samples collected in a 15 m soil profile in the catchment Tumara by Zech et al. [2007]. The ranges in C/N ratios and $\delta^{13}\text{C}$ values in autochthonous material and terrestrial plants are from Cleveland and Liptzin [2007], Finlay and Kendall [2008], Finlay et al. [1999], O'Leary [1988], Sterner and Elser [2002], and Vuorio et al. [2006].

The range in C/N ratios in the particulate organic matter, C/N_{POM} , was not as large as the range in C/N_{DOM} . Observed values were from 5 to 17 with a median of 10 (Table 2 and Figure 3c). Median C/N_{POM} were largest in the main channels of the Lena River and the Aldan River. There were no significant differences among geographical areas.

C/N ratios for plant samples, $\text{C/N}_{\text{plant}}$, ranged between 26 and 42, with a median of 36 (Table 2 and Figure 6).

4. Discussion

4.1. Organic Carbon Concentrations

DOC concentrations in the main channel of the Lena River (590–1300 μM) were similar to previous observations in the Lena River (170–1230 μM) and comparable to the range in other Arctic Rivers [Amon et al., 2012; Cauwet and Sidorov, 1996; Cooper et al., 2008; Kuzmin et al., 2009; Lara et al., 1998; Le Fouest et al., 2013; Lobbes et al., 2000]. However, DOC data from the tributaries had an even larger range (Figure 3a). The range in POC concentrations was larger (6–640 μM) compared to other studies in the Lena River (2.5–430 μM), but similar to the range in other Arctic Rivers [Cauwet and Sidorov, 1996; Guo et al., 2007; Kuzmin et al., 2009; Le Fouest et al., 2013; Lobbes et al., 2000; McClelland et al., 2016]. The large range in POC was primarily due to the flood event in the Upper Lena in 2012. The flood increased POC concentration by approximately 500% with a simultaneous increase in discharge (Figure S1b). At Tabaga, discharge increased to about 40,000 $\text{m}^3 \text{s}^{-1}$, which is almost 30,000 $\text{m}^3 \text{s}^{-1}$ more than the average flow in the end of July and of the same magnitude as spring floods [Tananaev, 2016]. At Kiusiur discharge increased from 30,000 to 46,000 $\text{m}^3 \text{s}^{-1}$ as observed 17 days later, which was the approximate in-river water travel time at this period (Figure S1). Notably, there was no concurrent change in DOC concentrations in the main channel of Lena River following the flood.

The observed differences in OC concentrations, conductivity, water temperatures, and pH across the Lena main channel suggest incomplete mixing in the main channel of the Lower Lena. Downstream the outflow of Aldan River, the Lena River main channel was wide and had a meandering deep channel with patches of islands in the channel that could act as barriers restricting mixing of the water. The incomplete mixing of Lena River might also have been a result of density differences between water from the CSP and the VER tributaries due to different sediment loads.

A correlation between mean slope and both DOC and POC concentrations, seen in the Lena tributaries (Figures 4a and 4b), has previously been reported by Mulholland [1997], who observed a strong correlation between average channel slope and DOC concentration in data from 33 North American catchments, along a climatic gradient from Puerto Rico to Alaska including streams with stream orders from 1 to 7. Recently, a study by Jasechko *et al.* [2016], comparing global watersheds that included permafrost dominated regions, showed that the stream water in mountainous catchments in steep terrain generally have a higher proportion of older water ($>2.3 \pm 0.8$ month old) compared to lower relief landscapes. As the proportion of young versus old water is a proxy for shifting pathways in streamflow and travel times, it suggests that areas with steeper relief have deeper vertical infiltration and are more influenced by groundwater [Jasechko *et al.*, 2016; McGuire *et al.*, 2005]. In permafrost areas, the permafrost could restrict deeper percolation by acting as semiimpermeable aquitards. When hydrologic flow paths are forced to surface layers of soils due to restrictions in vertical drainage, the interaction with organic-rich surface soil layers lead to higher DOC concentrations [Laudon *et al.*, 2011, 2012; Mulholland, 1997]. As the distribution of permafrost in steep terrain varies depending on aspect, elevation, and regional hydroclimate, a small-scale mosaic of permafrost and permafrost-free areas could shift the hydrology on local scale [Woo *et al.*, 2008] and thus the OC concentrations. Our results are consistent with deeper flow pathways dominating mountainous catchment drainage compared to that of catchments with lower relief. Deeper flow pathways will likely transfer water through mineral soils allowing substantial adsorption and degradation of organic matter and consequently lower stream DOC and POC concentrations [Kaiser and Kalbitz, 2012; Klaminder *et al.*, 2011]. In the VER region, headwaters are located in alpine areas but reach lowland areas with wetland type soils before discharging into the Lena River. Wetland areas are generally key contributors of DOC to aquatic systems due to the high content of SOM [Eckhardt and Moore, 1990; Laudon *et al.*, 2011; Mulholland, 1997]. However, the wetland areas did not seem to have a major influence on DOC concentrations in the VER tributaries, which remain low (Figure 3a). Similarly, tributaries to the Aldan River with headwaters primarily in mountainous areas, which have steeper slopes, had lower DOC and POC concentrations compared to the other stations in the Aldan catchment (Figure 3a). In addition, lower OC concentrations in rivers draining mountainous areas could be due to less soil organic carbon in these regions and less extensive tree cover at high altitudes [Chevychev and Bosikov, 2010; Hugelius *et al.*, 2014].

The Viliui River and its tributaries, which drain the flat lowland area of CSP, had high concentrations of DOC and POC (Figure 3a). The CSP is mainly underlain by continuous permafrost [Chevychev and Bosikov, 2010] and may have hydrologic flow path restricted to more organic-rich superficial soil layers. Catchment with large riparian wetlands that in general have low slopes and broad floodplains, commonly have high concentrations of DOC [Mulholland, 2003]. A recent study by Boike *et al.* [2016] reported that the area close to the Viliui River in generally became wetter and increased its lake distribution between 2002 and 2009, even though some parts had a shrinking lake distribution. An increase in wetlands may result in even greater OC concentrations in CSP tributaries.

4.2. Export of OC

The calculated export of TOC in June and July from the Lena River to the Arctic Ocean, 3.5 Tg TOC (Figure 2 and Table 1), was more than half of the annual TOC export estimated by previous studies (6.6 Tg TOC year⁻¹) [Le Fouest *et al.*, 2013]. DOC in June and July dominated the organic carbon transport; TOC consisted of 98% DOC, which is a common pattern for Arctic and boreal rivers [Gordeev and Kravchishina, 2009; Le Fouest *et al.*, 2013; Lobbis *et al.*, 2000; Meybeck, 1982].

The TOC export from the Upper Lena and the Aldan and Viliui Rivers showed that these areas contributed 34%, 27%, and 10% of the TOC, respectively (Figure 2). The Lower Lena contributed 29% of the TOC transported from the Lena River to the Arctic Ocean. Since discharge varies by several orders of magnitude (both spatially and temporally) while OC concentrations rarely vary by more than a factor of 10 [Godsey *et al.*, 2009], export estimates were probably dominated by variations in discharge (Table 1), and the higher contribution to TOC from the Upper Lena (Figure 2) could be explained by a higher mean precipitation in this region [Chevychev and Bosikov, 2010]. Compared to the other subbasins the Upper Lena also has more sporadic and discontinuous permafrost, which have been shown to contribute with higher fluxes and concentrations of river OC [Frey and McClelland, 2009]. This area also has variable soil and topographical characteristics, the mildest climate and the most productive forest in the Lena basin [Kuznetsova *et al.*, 2010]. These features may

influence OC dynamics both through variable OC sources and through its influence on water flow pathways, which have been shown to partly control OC concentrations in streams and rivers [Mulholland, 2003; Winterdahl *et al.*, 2011]. In contrast, the export of TOC from the Viliui River was low (Figure 2), although DOC concentrations were high (Figure 2a). The area specific export of TOC was significantly lower compared to other parts of the Lena basin (Table 1 and Figure 2). A large dam in the upstream area of the Viliui catchment reduces discharge at the mouth of the Viliui River during summer peak flow by 20–25% [Ye *et al.*, 2003], which partly could explain the low export of TOC from that area.

Previous studies of seasonal OC dynamics in the Lena main channel have shown that discharge and OC concentrations peak during spring flood and reach annual lows during winter [Holmes *et al.*, 2012; Le Fouest *et al.*, 2013; Raymond *et al.*, 2007]. This behavior is typical for both DOC and POC dynamics in Arctic and boreal rivers [Eckhardt and Moore, 1990; Holmes *et al.*, 2012; Le Fouest *et al.*, 2013; Mulholland, 2003; Winterdahl *et al.*, 2014]. Water samples collected earlier in the summer could thus have higher concentrations compared to samples collected later in the summer. Sampling during two different months and in two different years, the fact that most sample stations were only sampled once and that the POC likely was underestimated were uncertainties that likely have influenced our export estimates. Since spatial patterns in OC transport within the Lena River basin currently are not well understood, our export calculations could be considered as a first preliminary estimate of spatial OC export in the area. To reduce the uncertainties in the carbon export calculations, due to the incomplete mixing in the Lena River, the average concentration of the two most downstream sample stations in the Lena main channel were used. One sample was collected at the eastern side of the main channel whereas the other was collected on the western side.

4.3. $\delta^{13}\text{C}$ Values and C/N Atomic Ratios

The $\delta^{13}\text{C}$ values and the C/N ratios of freshwater OC vary depending on sources and biogeochemical pathways [Cleveland and Liptzin, 2007; Finlay and Kendall, 2008; McGroddy *et al.*, 2004; Sterner and Elser, 2002]. These parameters could therefore be used to identify sources or processes controlling OC. The results in this study showed different patterns in $\delta^{13}\text{C}$ and C/N between DOC and POC (Figure 6). In general, there was a narrow range in $\delta^{13}\text{C}_{\text{DOC}}$, but a wide range in C/N_{DOM} . In contrast, there was high variation in $\delta^{13}\text{C}_{\text{POC}}$ and a low variation in C/N_{POM} . These patterns indicated different dominant sources and water flow paths influencing the OC composition. The DOC composition agreed with the range in both $\delta^{13}\text{C}$ values and C/N ratios for the plant material collected in this study and SOM in the Tamara river catchment (central part of Lena basin) reported by Zech *et al.* [2007] (Figure 6). The majority of the dissolved samples in this study had C/N_{DOM} higher than what is typical for autochthonous sources, thus excluding these as main sources of DOC. This indicated that riverine DOC was primarily a mixture of relatively fresh plant material and SOM. Previous studies in the main channel of the Lena River and other Arctic Rivers based on organic biomarkers and ^{14}C data suggest that the major source of DOC in the spring freshet is primarily relatively fresh vegetation material [Amon *et al.*, 2012; Lara *et al.*, 1998; Lobbes *et al.*, 2000].

The sources of DOC could be treated as a mixture between different distinctive sources (Figure 5a). Since fresh plant material primarily is found at the soil surface and in surficial soil layers while SOM is derived from deeper soil layers, these results are consistent with water flow paths through fresh organic material in top soils in catchments with high C/N_{DOM} and low $\delta^{13}\text{C}_{\text{DOC}}$, while the OC in catchments with low C/N_{DOM} and high $\delta^{13}\text{C}_{\text{DOC}}$ values was influenced more by SOM and deeper water flow paths (Figures 5a and 5b). Studies of Siberian soil profiles have shown higher C/N in surficial soil layers compared to deeper soil layers [Gittel *et al.*, 2014; Hugelius *et al.*, 2012; Menyailo and Hungate, 2006; Palmtag *et al.*, 2015; Zech *et al.*, 2007] and lower $\delta^{13}\text{C}$ values (–28 to –25‰) in surficial soil layers [Kaiser *et al.*, 2007; Menyailo and Hungate, 2006; Zech *et al.*, 2007]. The processes causing isotope fractionation of SOM are still not fully understood but are believed to be associated with decomposition or diagenesis. For instance, during decomposition of plant litter, microbial organisms favor the light isotope (^{12}C) and specific components of SOM resulting in increasing $\delta^{13}\text{C}$ values [Boström *et al.*, 2007; Ehleringer *et al.*, 2000]. In addition, the release of CO_2 during aerobic consumption of SOM can result in lower SOM C/N ratios. In general, the C/N is lower in SOM than in terrestrial plant material [Cleveland and Liptzin, 2007; McGroddy *et al.*, 2004], suggesting the importance of organic matter microbial recycling in soils.

There were differences in C/N_{DOM} and $\delta^{13}C_{DOC}$ values (Figure 5a) among sampling months and years. Water samples collected early in the summer (June 2013) in the northern parts of the Lena basin, in for example the Viliui River and the northernmost tributaries of CSP and VER, showed higher C/N_{DOM} and lower $\delta^{13}C_{DOC}$ (Figures 3b and 3c). This indicated a stronger influence of fresh plant material, which could be due to surficial water flow paths caused by the shallow active layer in the beginning of summer and sampling at higher latitudes in the 2013 samples (Figures 5a and 5b). In contrast, samples collected at lower latitudes and later in the summer when temperatures were higher (July 2012), and at lower latitudes in the tributaries of Aldan and Upper Lena, showed opposite patterns (high $\delta^{13}C_{DOC}$ and low C/N_{DOM}) indicating more influence of SOM and thus probably deeper dominant water flow paths due to deeper active layers (Figures 3b, 3c, and 5). The samples from the Upper Lena were collected late in the summer (July 2012), when air temperatures were high and thus the active layers probably were thicker. This in turn allowed for deeper water flow paths and an increased leakage of more degraded OC, probably primarily derived from SOM to the rivers. The Upper Lena region has the mildest climate and more widespread discontinuous permafrost distribution than in other parts of the Lena basin [Chevychev and Bosikov, 2010] that might impact water flow paths and OC fluxes [Frey and McClelland, 2009]. Similarly, the samples collected from the Aldan outflow in early summer (June 2013) had higher OC concentrations and C/N_{DOM} and lower $\delta^{13}C_{DOC}$, indicating a larger influence of fresh plant material as a source of DOC, whereas the influence of autochthonous sources as a source of POC was lower than in July 2012. These results showed that the OC was strongly dependent on sampling period and year.

The results presented here indicate that POC was derived from partly different sources than DOC or was influenced by other processes either in the river water or in the soil during transport to the river. The $\delta^{13}C_{POC}$ and C/N_{POM} varied between typical values of terrestrial SOM and values found in freshwater autochthonous sources (Figure 6) [Farquhar *et al.*, 1989; Finlay and Kendall, 2008; Osmond *et al.*, 1981; Rundel *et al.*, 1979; Smith and Epstein, 1971]. In general, the C/N_{POM} was lower than C/N_{plant} , excluding fresh terrestrial vegetation as a main source of riverine POC. The sources of POC were thus probably mixtures of SOM and a substantial fraction of autochthonous sources (Figure 6). In some of the rivers in the LAIRA and CSP regions, where results indicated autochthonous sources of POC, there were visible algae blooms during field sampling.

The results of this study suggest that POC in the Lena River is influenced by aquatic primary production. Previous studies in the Lena River found that aquatic primary production is low [Sorokin and Sorokin, 1996] and that the POC mainly is derived from old and highly degraded terrestrial plant material [Kuptsov and Lisitsin, 1996; Lobbes *et al.*, 2000]. In contrast, recent studies report $\delta^{13}C_{POC}$ and C/N_{POM} that suggests an influence of autochthonous sources, whereas ^{14}C data indicate that the POC is old and thus of SOM origin [Karlsson, 2015; McClelland *et al.*, 2016]. These seemingly contradictory results could be due to recycling of respired old POC within the aquatic food web.

$\delta^{13}C_{POC}$ was correlated to both mean catchment slope and water temperatures (Figures 4c and S3). These results indicate that tributaries draining lowland areas, like the Viliui River and CSP tributaries, in general had a larger influence from autochthonous sources and higher water temperatures than tributaries originating in mountainous areas. High water temperatures induce higher metabolic rates and thus aquatic primary production as well as heterotrophic activity [Allen *et al.*, 2005; Gillooly *et al.*, 2001], which could partly explain the correlation between high water temperatures and low $\delta^{13}C_{POC}$. Also, the low gradient in the CSP region could have led to longer watershed water residence times and in turn higher water temperatures. Boike *et al.* [2016] show that surface land temperatures and lake area in this region have increased consistently during the period 2002 to 2009. Larger lake coverage generally favors growth of aquatic organisms, including primary producers. The large influence of SOM in both POC and DOC isotope compositions and concentrations, as well as the lower water temperatures, further indicates deeper water flow paths in the rivers mainly draining mountainous areas.

5. Conclusions

This study has provided insights into river OC dynamics in the Lena River basin. Spatially variable concentrations, export, and major sources of OC were found within the basin. These were probably driven by differences in water flow pathways, topography, climate, and land cover. The slope in elevation across each subcatchment was a significant predictor of OC concentrations. Water derived from mountainous

areas had in general low concentrations of OC and cold water temperatures, and the primary source of DOC and POC was dominated by SOM. In contrast, lowland areas had high concentrations of OC, higher water temperatures and the DOC sources were dominated by relatively fresh plant litter, while the primary POC sources were autochthonous produced material. We also observed temporal differences in DOC sources with tributaries sampled early in the summer having a larger influence of fresh plant material, indicating that the dominant water flow paths were concentrated to surficial soil horizons. DOC in tributaries sampled later in the summer had a larger influence of SOM, indicating deeper water flow paths. In addition, the results showed that the spatial variations in export and area specific export of OC were mainly due to variation in discharge but with additional impacts by local climate, variations in vegetation, and differences in permafrost extent.

The results suggested that the Lena River basin could experience shifts in OC fluxes and concentrations with continued permafrost degradation. A warming climate will likely lead to changes in water flow paths that in turn would affect the export of OC from the Lena River basin. Our results indicate that continued permafrost degradation leading to deeper water flow pathways may induce lower concentrations of OC. Exports and fluxes of OC are, however, driven by variations in discharge, and as this study indicates fluxes and concentration are spatially variable, and so future shifts will ultimately be dependent on changes in hydrology and precipitation patterns in the Lena River basin.

Acknowledgments

We thank the staff at the laboratories at department of Geosciences at the Museum of Natural History, department of Environmental Science and Analytical Chemistry at Stockholm University and the Stable Isotope Laboratory at Stockholm University for assistance. We also thank Peter Raymond, Sebastian Sobek, and Mattias Winterdahl for insightful comments on earlier version of the manuscript. Finally, we want to thank all our Russian colleagues at the International Center for BioGeoScience Education and Scientific Training (BEST) of the North-Eastern Federal University in Yakutsk for all assistance in the field as well as the crew members at R/V Geokryolog, R/V Akademik, and R/V Merelotoved. Biogeochemical data used in this study are included as one table in an SI file. The discharge data used were measured by the Russian Hydrometeorological Services and are freely available at R-ArcticNet (<http://www.r-arcticnet.sr.unh.edu/v4.0/index.html>) and ArcticRIMS (<http://rims.unh.edu/data.shtml>). GIS data were downloaded from (<https://lta.cr.usgs.gov/GTOPO30>). This project was supported by the Swedish Research Council (VR 621-2010-3917) and the Swedish Polar Research Secretariat (SIMO 2011-165 and 2012-213).

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