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Dissolved and Particulate Organic Carbon Characteristics in Summer and Winter Waters of the Lena Delta

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ABSTRACT

Rapid Arctic warming accelerates permafrost thaw, altering water flow and organic matter transport to aquatic ecosystems. To identify sources and seasonality of OC at the mouth of the Lena River, we measured summer and winter concentrations and C isotopes ($\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$) of DOC and POC along a 140-km transect of the Lena Delta. Despite low water flow during winter, DOC concentrations in the Lena Delta were higher than those measured at the end of the summer (6.31 ± 0.60 and $5.54 \pm 0.17 \text{ mg L}^{-1}$, respectively). We found pronounced differences in the DOC isotopic composition of waters between seasons (winter: mean $= -16\% \pm 16\%$ ranging between -14% and 46% and summer: mean $= 41\% \pm 26\%$ in the range between -47% and 79%). $\Delta^{14}\text{C}$ of winter DOC suggested higher relative contributions of older carbon compared to summer DOC, which is enriched in ^{14}C . POC in winter was lower (0.13 ± 0.06 and $0.40 \pm 0.10 \text{ mg C L}^{-1}$, respectively) and enriched in $\delta^{13}\text{C}$ (-29.7 ± 2.2 and $-32.4 \pm 0.8\%$, respectively) compared to summer, while no difference was found for $\Delta^{14}\text{C}$. This study with its unique dataset on the largest Arctic delta will help to assess the ongoing changes with climate warming at this frontier between the land and the ocean realm. Explicitly, the inclusion of winter sampling and isotopic analysis makes this study very valuable for assessing the biogeochemical response of the Arctic's biggest delta, as well as beyond.

1 | Introduction

The Arctic is experiencing unprecedented environmental change in response to global climate change. High-latitude landscapes are susceptible to ongoing surface permafrost thaw due to warming Arctic air and soil temperatures [1, 2]. Terrestrial landscape changes accelerate the release of organic matter (OM), containing organic carbon (OC), and nitrogen from peat, soil, and permafrost to inland aquatic systems [3–8]. Terrigenous OM and nutrients can be further transported to the Arctic coastal waters [9–11] with subsequent impacts on biogeochemical cycles, primary production, and dissolved inorganic carbon in the Arctic Ocean [12–14]. Contributions of permafrost-derived OC

in aquatic and Arctic coastal waters are affected by changing river discharge regimes [15–17] and supply from rapidly eroding river banks [18–20] and coastlines [21]. Once mobilized into inland or coastal waters, permafrost and peat-derived OC from deepening active layers may be rapidly utilized by aquatic microorganisms and emitted to the atmosphere as carbon dioxide or methane [22, 23–25] enhancing riverine C emissions from river basins and nearshore waters [26]. Permafrost OC inputs to Arctic rivers and streams, particularly dissolved organic carbon (DOC) from ice- and organic-rich Yedoma permafrost, are highly labile and preferentially utilized by aquatic microorganisms, leading to patterns of decreasing permafrost contributions in OC pools with increasing water residence times [27–29].

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Nearshore regions across the Arctic (including deltas, estuaries, and coasts) are biogeochemically active areas and hotspots for environmental change [28] where major transformation processes of terrestrial material are expected to take place [10, 21, 30, 31, 32]. Estuarine and deltaic regions can be regarded as filters for riverine inputs and solute fluxes to the coastal waters and the open ocean [33–36]. Despite the importance of Arctic estuarine and deltaic environments in OM biogeochemistry [17], their functioning is still poorly understood, and the number of studies and therewith datasets that consider the supply, composition, and transformation of terrestrial OM during transport from river-to-shelf is limited. Moreover, studies comparing the different seasons are lacking for OC or scarce for other nutrients [37], limiting the ability to predict the impact of shifting seasonality and of intensification of the Arctic freshwater cycle. In the specific context of the Lena River, one of the Arctic's largest rivers with increasing winter water discharge, total carbon entering the sea with the Lena discharge is estimated to be almost 10 Tg C year⁻¹. Lena River discharge rates have increased over the last decade by around 10% (relative to the 1971–2000 baseline) and are expected to continue to increase (+25% by 2100; [28] and references herein). River ice break-up is generally occurring earlier, and freeze-up later [38], shortening the period of winter base flow. Moreover, the delayed active layer freeze-up increases winter river runoff [39–43]. Changing connectivity between terrestrial landscapes and coastal waters has potentially significant ramifications for coastal carbon budgets and ecosystem structure but depends upon how OM is transformed with estuarine and deltaic environments.

In this study, we examine how the supply and composition of DOC and POC (using $\Delta^{14}\text{C}$, $\delta^{13}\text{C}$) in the Lena River delta varied between winter and summer seasons. Further, we examine how dissolved and particulate OC loads vary during transit using seasonal transects across the Lena Delta. Our aim was to identify the dominant sources of DOC and POC to deltaic waters and to consider how OC modification differs between dominant seasons.

2 | Materials and Methods

2.1 | Study Area

The Lena Delta is the largest delta in the Arctic and represents one of the largest in the world. It occupies an area of $29.17 \times 10^3 \text{ km}^2$ and combines more than 800 transverse channels and branches with a total length of 6500 km [44]. The delta receives approximately $543 \text{ km}^3 \text{ year}^{-1}$ of freshwater (mean discharge for 1936–2019 at the upstream Kyusyur discharge station; Wang et al. [43]). The Lena River catchment drains an area covering $\sim 2.61 \times 10^6 \text{ km}^2$, of which more than 94% contains permafrost and $\sim 70.5\%$ of the catchment area is underlain by continuous permafrost [45, 46]. Pleistocene-aged permafrost deposits (Yedoma) cover approximately 3.5% of the Lena watershed area [4, 47]. Yedoma cliffs are actively eroding along the Lena River and delta banks [19, 48].

In this study, we sampled the Sardakhskaya channel (Figure 1), which, together with the Trofimovskaya channel, forms a system through which about 60%–75% of Lena freshwater export

[44] and up to 70% of sediments are routed to the Arctic Ocean coastline [52, 53].

2.1.1 | Sample Campaigns and Water Collection

Two sampling campaigns to the Lena Delta were conducted during the winter and summer months of 2019. The winter campaign took place between March 26 to April 8, during which river, delta, and coastal waters were fully ice-covered. Sampling was conducted using an on-ice camp comprising a cabin, a freight sled, a tractor, and a caterpillar all-terrain vehicle, enabling unprecedented sampling across the difficult-to-access land-to-ocean region during the highly data-lean winter period (Figure 2). Sampling took place approximately every 5 km, starting at the easternmost location upon sea-ice (Figure 1; CAC19-03/-04). Sampling continued westward along a transect toward the delta and then up ~ 40 km into the Sardakhskaya channel (the last sampling site LEN19-S-01, Table S1). At LEN19-S-06, we passed a rapidly eroding Yedoma Cliff (Figure 2c) previously studied by Fuchs et al. [19] and Wetterich et al. [54, 55].

At each site, a borehole was drilled through the ice, and 20 L of water was collected using multiple deployments of a 5 L water sampler (UWITEC, Austria). Water was collected from between one and three depths per station depending on the bottom depth at the sampling location. Water was collected directly under the ice (around 2 m) at each sampling site. When river depths exceeded 4–7 m at a site, an additional water sample was collected from just above the bottom (4–7 m). At water depths > 7 m, an additional water sample was collected from the mid-depth between the under-ice and bottom samples (Table S1). Winter water samples ($n=21$) were stored in 20 L plastic canisters (pre-cleaned with 10% HCl for 3 days) and kept cool until returning to the mobile laboratory in the camp, where they were further processed.

The summer sampling campaign was conducted between the August 7–28, 2019. Sampling by ship started in the West at Stolb Island, near the transition point between the Lena River and Delta (Figure 1). Sampling subsequently took place at locations along the Sardakhskaya branch out into the Eastern Laptev Sea, and stations previously sampled during the winter campaign were re-occupied. Distances between sample sites varied between ~ 5 km along the channel, increasing to ~ 20 km within the Laptev Sea coast. Summer water sampling and collection ($n=23$) was conducted identically to that of winter samples, except surface samples were taken from between 0- and 1-m depth.

2.1.2 | Archive Samples and Datasets

In addition to samples from the two sampling campaigns described above, we also include previously unpublished datasets from samples collected during similar expeditions from 2016 to 2017. These include water samples from near the island of Stolb at the apex of the delta ($n=32$), as well as water samples from small ponds on Samoylov Island ($n=6$), which were collected in August 2016 and July 2017 (see also [29]). The collection of these

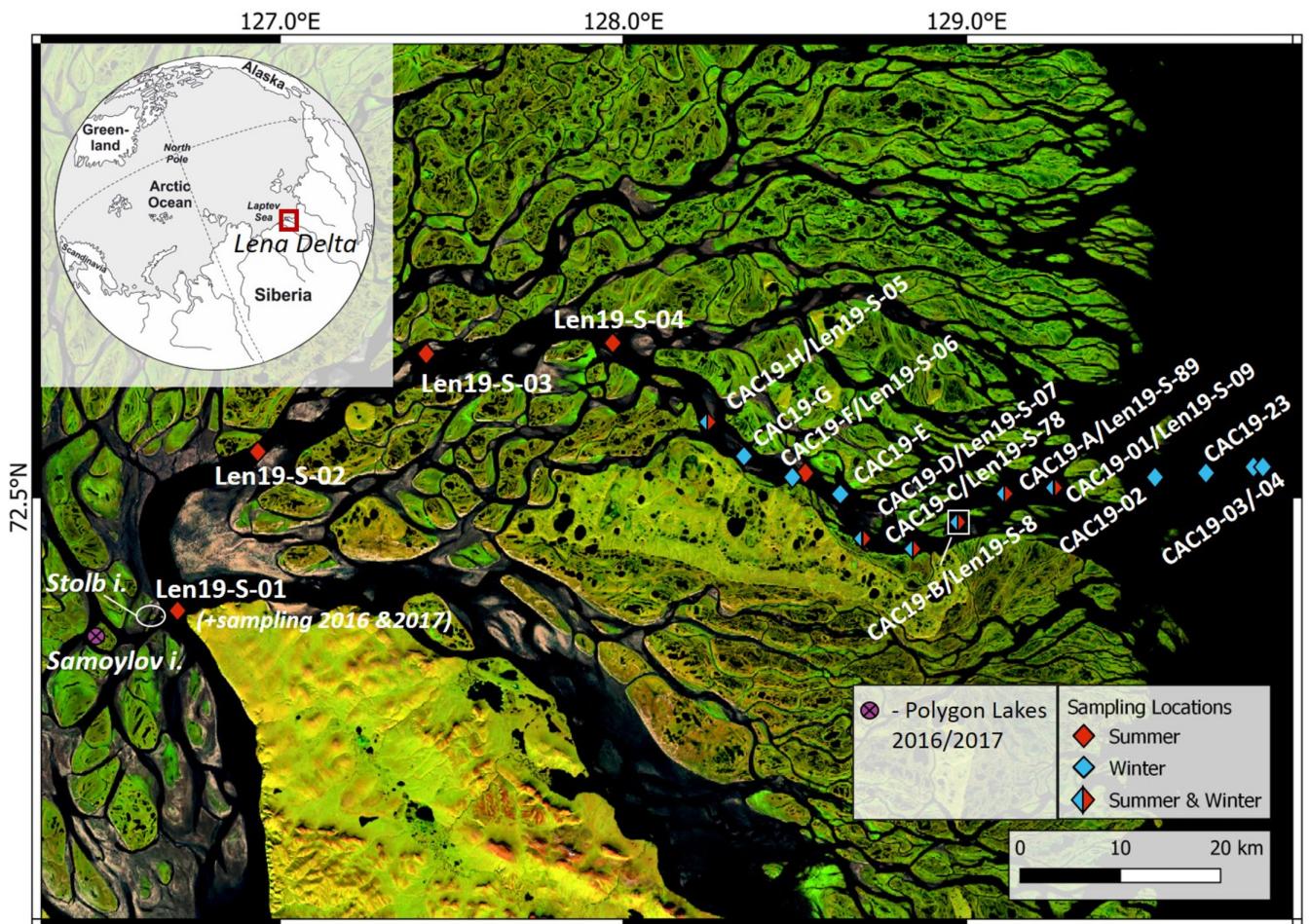


FIGURE 1 | Sampling transect in the Lena Delta along the Sardakhskaya branch in 2019 and sampling locations in the Lena Delta in previous years near Stolb Island. “CAC” samples represent on-ice winter sampling during the CACOON project, while “Len” denotes the *Lena* summer (S) ship-based sampling. A total of 19 includes the year of sampling (2019). Adapted from M. Fuchs et al. [49, 50] and Strauss et al. [51]. [Colour figure can be viewed at wileyonlinelibrary.com]

water samples and subsequent DOC concentration and isotope analyses (described below) was identical to our summer sampling campaign approach.

2.2 | Laboratory Analyses

Water samples were immediately filtered on-site during the winter sampling campaign, or in summer, after less than 10 days’ storage, at the laboratory at the Samoylov Island Research Station. Waters were passed through a precombusted (4.5 h, 450°C) and pre-weighed glass fiber filter (GF/F Whatman, 0.7 µm nominal pore-size, Ø 2.5 cm) to separate POC (remaining on the filter) and DOC (dissolved in water which passed through the filter). Filters were packed into precombusted glass petri dishes, and waters were collected into 150 mL of HDPE bottles (precleaned with 10% HCl) and both kept frozen at -18°C until further analyses.

2.2.1 | DOC Concentration and Carbon Isotope Analyses ($\Delta^{14}\text{C}$, $\delta^{13}\text{C}$)

DOC concentrations (mgC L^{-1}) were measured using a high temperature catalytic oxidation analyzer (TOC-VPH Shimadzu) at

Alfred Wegener Institute (AWI) Potsdam. After every tenth sample, one blank (Milli-Q water) and one standard were measured. Five different certified standards covering a range between 1.26 mgC L^{-1} (SUPER_05_1) and 27.31 mgC L^{-1} (Std. US-QC) were used to calibrate the instrument. The results of standards provided a precision of $\pm 10\%$.

Radiocarbon analyses by accelerator mass spectrometry (AMS) were carried out on a Mini Carbon Dating System (MICADAS) at AWI Bremerhaven, following the standard operation procedures described in detail by Mollenhauer et al. [56]. Filtered water samples (GF/F Whatman, 0.7 µm membrane) were acidified to $\text{pH} \sim 1-2$ with HCl. The aliquot for analyses (a volume containing approximately 100 µgC) was dried using a rotary evaporator. Isolated DOC was transferred in an aqueous solution into 50 µL of liquid tin capsules. After complete drying at 40°C in a desiccator under vacuum (500–600 bar), capsules with DOC were folded, combusted in an elemental analyzer (Elementar Analysensysteme) connected via the Gas Interface System [57] to the MICADAS, and analyzed using the option to obtain ^{14}C results on CO_2 gas. The radio-carbon data were normalized against CO_2 gas produced from oxalic acid II (OxAlII, NIST SRM4990C) and blank corrected against ^{14}C -free CO_2 gas. Additionally, the processing blank



FIGURE 2 | Sampling vehicles in winter and summer. (a) all terrain caterpillar vehicle in the Lena Delta on the Lena Delta river ice for reaching field sites further from the camp. (b) Working process on ice in the Lena Delta in winter: drilling and water sampling. (c) The research vessel “Merzlotoved” with the Sobo-Sise Yedoma cliff in the background. (d) Water sampling at the back of the research vessel in the Lena Delta in summer 2019. [Colour figure can be viewed at wileyonlinelibrary.com]

was determined following Sun et al. [58], and raw radiocarbon data were subsequently blank corrected following Wacker et al. [59]. We report radiocarbon data as $\Delta^{14}\text{C}$ values in ‰, which express the relative difference in ^{14}C activity between the absolute international standard (base year 1950) corrected for sampling time and normalized to $\delta^{13}\text{C} = 25\text{‰}$ [60]. To facilitate the discussion of the results, we refer to the most ^{14}C -depleted samples (less than -900‰ or older than $\sim 18,500$ ^{14}C years) as “ancient”, samples with $\Delta^{14}\text{C}$ ranging between -50‰ and -900‰ (corresponding to ~ 400 – $18,500$ ^{14}C years) are referred to as “old”, and samples with $\Delta^{14}\text{C}$ above -50‰ to 0‰ are identified as “young” and “modern” (above 0‰).

The preparation for the stable C isotope analyses was the same as for $\Delta^{14}\text{C}$, but samples were measured on a Sercon-20-20 isotope ratio mass spectrometer (IRMS) coupled to an Automated Nitrogen Carbon Analyzer (ANCA) at AWI Bremerhaven. Stable C isotope values were expressed as $\delta^{13}\text{C}$ in per mil (‰) and normalized against the Pee Dee Belemnite (PDB) standard. The analyzer was calibrated using Isoleucine (NIST, RM 8573,

USGS40) with a known isotopic composition. The precision of $\delta^{13}\text{C}$ measurements was better than $\pm 0.2\text{‰}$.

2.2.2 | Particulate Organic Carbon Concentration and Carbon Isotope Analyses ($\delta^{13}\text{C}$, $\Delta^{14}\text{C}$)

To determine total POC concentration (mgC L^{-1}) together with stable C isotopes and to determine $\Delta^{14}\text{C}$ of POC, filters were dried for 24 h at 40°C , then acidified with drops of 10% HCl (sufficient to wet the filter surface, including its sediment) to remove inorganic C. If the amount of C exceeded $100\text{ }\mu\text{g}$, only a subsample of the filter was used. After carbonate removal, the filters were oven-dried under the same conditions and packed/rolled into tin boats ($8 \times 8 \times 15\text{ mm}$; IVA Analysentechnik Part no.: IVA2213742001).

POC content on the filter and its $\delta^{13}\text{C}$ signature were measured on a Sercon-20-20 IRMS. The mean uncertainty for POC was $\pm 3.34\text{ }\mu\text{gC}$, and concentrations were determined by dividing the

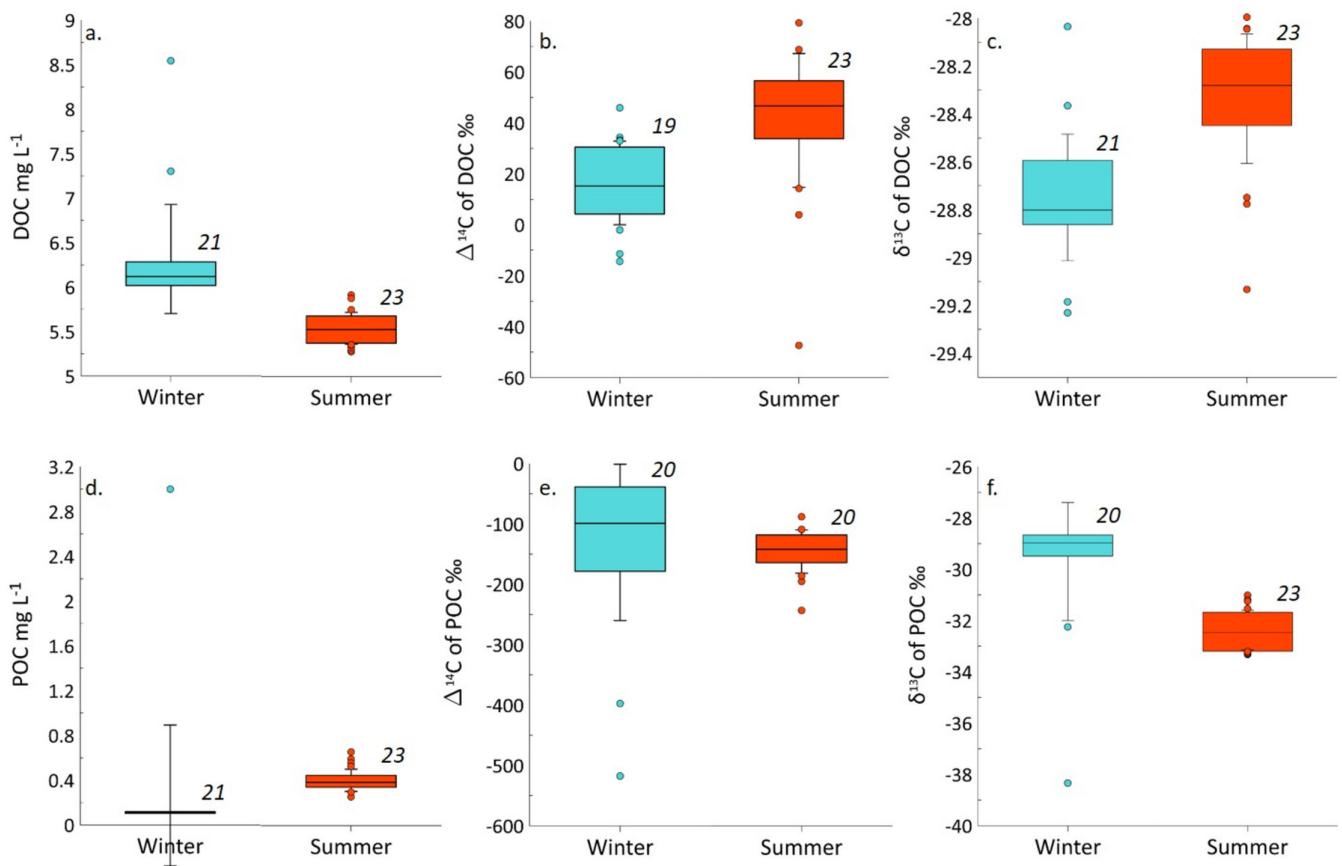


FIGURE 3 | Box-whisker plots displaying the variability in dissolved (DOC, top row: a-c) and particulate (POC, bottom row: d-f) organic carbon concentration and isotopic composition ($\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$) (for all sampling depths) between winter and summer in the Lena Delta in 2019. Central lines in plots represent the median, boxes are defined by the lower and upper quartiles, and whiskers indicate the standard deviation, numbers denote sample count. [Colour figure can be viewed at wileyonlinelibrary.com]

POC content per filter by the volume of water filtered through that filter. The isotope ratios and C masses were evaluated for precision and accuracy by conducting multiple analyses of in-house standards (isoleucine, NIST, RM 8573, USGS40) with known isotopic composition ($-26.39\% \pm 0.09\%$). The $\delta^{13}\text{C}$ measurements exhibited a precision better than $\pm 0.2\%$, while the average uncertainty for POC was $\pm 3.34\mu\text{g}$. The concentrations were determined by dividing the POC content per filter by the volume of water filtered through that filter.

Radiocarbon analyses of POC like for DOC were carried out on a Mini Carbon Dating System (MICADAS) following the procedure described in detail in Mollenhauer et al. [56]. The processing blank was determined by analyzing five empty combusted blank filters (GF/F, 2.5 cm Ø) treated identically to the samples [56]. POC data for the summer transect only has previously been described in Ogneva et al. [17, 61].

2.3 | Statistics

We used the Welch *t*-test to examine whether differences between parameters measured in 2019 in the Lena Delta in winter and in summer were statistically significant. The significance level (alpha) was set at 0.01.

3 | Results

3.1 | OM Transformation Within the Delta: Winter vs. Summer

3.1.1 | Concentrations of DOC and POC

Concentrations of DOC and POC were not significantly different by sample depth, so here we only present results separated by sampling period and study site. Deltaic POC and DOC concentrations differed between summer and winter campaigns (Figures 3 and 4). Winter DOC concentrations displayed a greater range (mean $6.31 \pm 0.61 \text{ mgC L}^{-1}$, $n=21$, ranging between 5.82 and 8.54 mgC L^{-1}) than summer month samples (average: $5.54 \pm 0.18 \text{ mgC L}^{-1}$, range of 5.28 to 5.92 mgC L^{-1}). Two specific winter locations had markedly higher DOC concentrations than the winter mean concentration. One of these samples from the middle of the sampling transect (LEN19-S-07/CAC19-F) was collected from a station 1–2 km downstream from an eroding Yedoma bank (Sobo-Sise cliff; Figures 3 and 4) and contained a DOC concentration of 8.54 mgC L^{-1} . A second higher DOC concentration of 7.30 mgC L^{-1} was also found at the Delta mouth (CAC19-02). DOC concentrations in the Lena Delta were significantly higher during winter months relative to summer, even when the influence of CAC19-F (erosion-influenced)

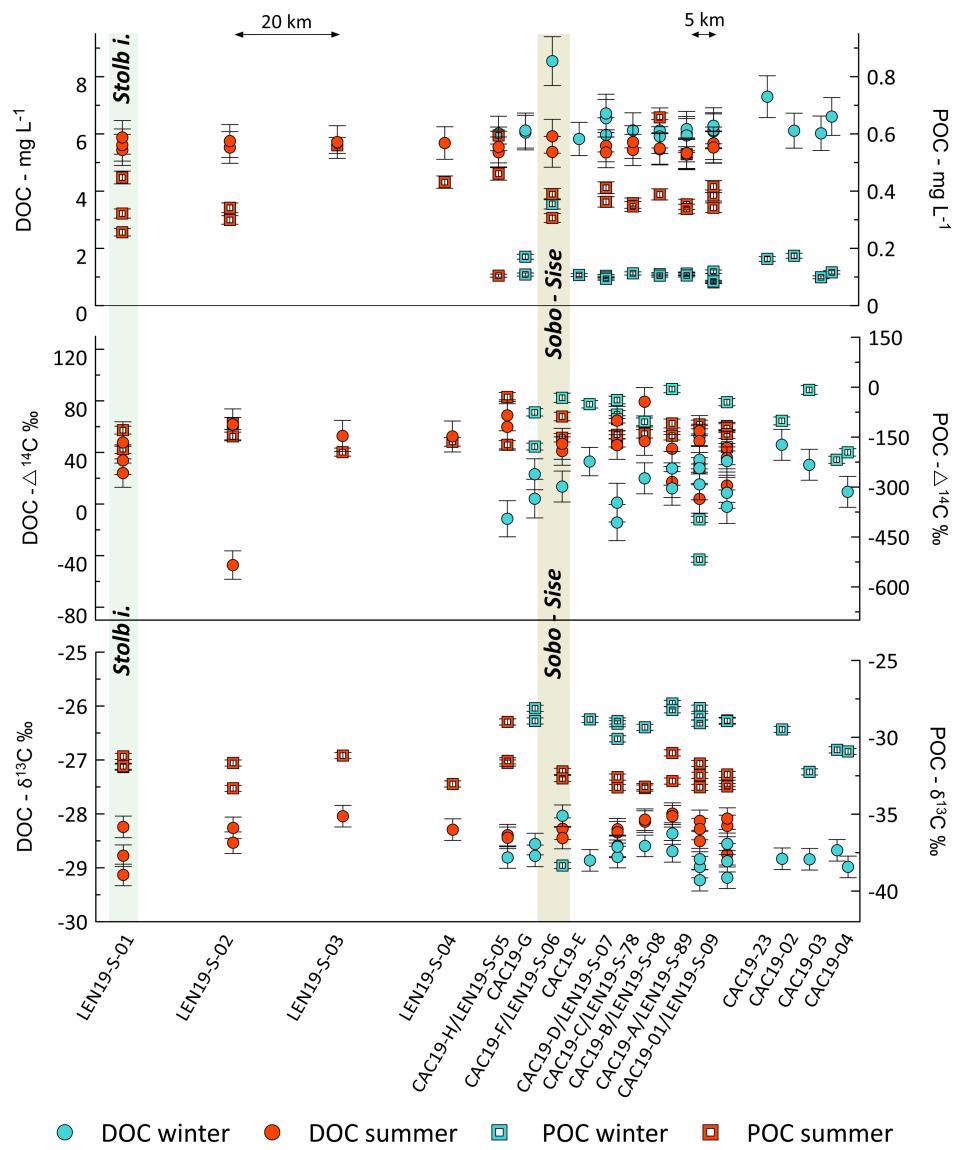


FIGURE 4 | Variability of DOC and POC concentration (a) and isotope composition: $\Delta^{14}\text{C}$ (b) and $\delta^{13}\text{C}$ (c) (for all sampling depths) along the transects across the Sardakhskaya branch in the Lena Delta in summer and winter 2019. The relative distance on the x-axis represents the actual distance between sampling sites, except for sites CAC19-03 and CAC19-04, where the actual distance is 1 km, but the larger distance (5 km) on the x-axis was kept on the figure to avoid overlapping. [Colour figure can be viewed at wileyonlinelibrary.com]

was removed (Welch t -test; $t(42)=5.8$, $p<0.001$ or $t(41)=8.0$, $p<0.001$). In general, no clear trends in DOC concentrations were observed with distance downstream and offshore, across either season.

POC concentrations differed significantly between seasons ($t(42)=-11.2$, $p<0.001$ or $t(41)=-12.7$, $p<0.001$, when the very high value measured at site CAC19-F was excluded from the calculation), displaying an inverse seasonal pattern relative to the DOC pool (Figure 3a,d). Mean summer POC concentrations are ~ 3 times higher ($0.40 \pm 0.10 \text{ mgC L}^{-1}$) than those measured during winter ($0.13 \pm 0.06 \text{ mgC L}^{-1}$). The highest winter POC concentration was recorded at the same station as the highest DOC concentration, namely, at station LEN19-S-07/CAC19-F (Figures 3 and 4—Sobo-Sise cliff), where the winter POC value is as high as in summer and reaches 0.36 mgC L^{-1} . In summer, this location is not characterized by high OM concentrations either in POC or in DOC.

We find winter POC concentration to be distributed relatively homogeneously along the Lena Delta (except LEN19-S-07/ CAC19-F site), while in summer, POC concentration values are more variable along the channel.

3.1.2 | Carbon Isotopic Composition of DOC and POC

The mean $\Delta^{14}\text{C}$ of DOC was significantly lower in winter samples than summer ($t(40)=-3.5$, $p=0.001$), yet summer values varied over a larger range (summer: mean = $41\text{\textperthousand} \pm 26\text{\textperthousand}$, range: $47\text{\textperthousand}$ – $79\text{\textperthousand}$; winter: mean = $-16\text{\textperthousand} \pm 16\text{\textperthousand}$ ranging between $-14\text{\textperthousand}$ and $46\text{\textperthousand}$; Figures 3 and 4). One sample (LEN19-S-02) drove the large variance in summer DOC values $\Delta^{14}\text{C}$ ($-47\text{\textperthousand}$), representing a highly depleted DOC sample within deeper Lena River mainstem waters. $\Delta^{14}\text{C}$ of DOC decreased at the three most seaward sampling locations (LEN19-S-08, -09 and -09, respectively). By contrast, winter $\Delta^{14}\text{C}$ DOC

values were found to be homogeneously distributed along the transect. No distinct patterns of difference in $\Delta^{14}\text{C}$ of POC ($t(38)=0.5, p=0.61$) or any particular trends in distribution are obvious, neither for the winter season nor for the summer (Figure 4).

Stable C isotopic composition varied between seasons for both DOC and POC (Figures 3 and 4). DOC concentrations in summer were slightly more enriched in ^{13}C compared to winter DOC (summer mean: $-28.3\% \pm 0.3\%$ in the range of -29.1% and -28.0% ; winter mean: $-28.8\% \pm 0.3\%$ in the range between -29.2% and -28.0% ; $t(42)=-5.1, p<0.001$ or $t(41)=-6.0, p<0.001$, when the very high value measured at site CAC19-F is removed from calculation). The highest $\delta^{13}\text{C}$ of DOC (-28.0%) is measured in summer at station LEN19-S-08/CAC19-B and in winter (-28.0%) at station LEN19-S-07/CAC19-F displaying the highest DOC and POC concentrations, and the lowest value (-29.1%) is measured in summer at the Stolb island (LEN19-S-01).

$\delta^{13}\text{C}$ of POC is lower in summer ($-32.4\% \pm 0.8\%$) and higher in winter ($-29.7\% \pm 2.2\%$) ($t(41)=5.3, p<0.001$ or $t(40)=10.8, p<0.001$, when the very low value measured at site CAC19-F is removed from calculation). There are no discernible spatial patterns observed in $\delta^{13}\text{C}$ of POC along the transect in both seasons.

3.2 | DOC Concentration and Isotope Composition in Polygonal Tundra Lakes and at Stolb in 2016–2017

The DOC concentration in the polygon lakes at Samoylov Island varied between 3.71 and 7.63 mgC L^{-1} , with a mean of $5.01 \pm 1.24 \text{ mgC L}^{-1}$, which is slightly lower than in winter and in summer 2019 within the Lena Delta (6.31 ± 0.60 and $5.54 \pm 0.17 \text{ mgC L}^{-1}$, respectively). Average $\Delta^{14}\text{C}$ is $45\% \pm 19\%$, similar to the $\Delta^{14}\text{C}$ for DOC in deltaic waters in summer 2019 and higher than in winter ($41\% \pm 26\%$ mean in summer and $-16\% \pm 16\%$ in winter). No ^{13}C stable isotope data are available for this sample set.

DOC concentrations at Stolb Island in the Lena Delta in 2016–2017 varied within a broader range than in 2019, between 3.17 and 8.45 mgC L^{-1} , with the mean of $7.16 \pm 1.33 \text{ mgC L}^{-1}$ in 2016 and $6.29 \pm 1.07 \text{ mgC L}^{-1}$ in 2017. DOC levels in the summers of 2017 and 2016 are higher than in the summer of 2019 ($5.54 \pm 0.17 \text{ mgC L}^{-1}$). Additionally, DOC levels in 2016 are higher, while in 2017, they are comparable to the levels in winter 2019 ($6.31 \pm 0.60 \text{ mgC L}^{-1}$).

The $\Delta^{14}\text{C}$ of DOC in 2016–2017 at Stolb varied between 35‰ and 69‰. There is no difference between the 2 years, with an average of $54\% \pm 8\%$ in both years. This is statistically similar to $\Delta^{14}\text{C}$ of DOC in the delta in summer 2019 ($t(52)=-2.7, p=0.01$) and higher than in winter 2019 ($t(48)=-10.9, p<0.001$), thus supporting the trend of winter DOC being more depleted in ^{14}C as we observed in 2019.

The $\delta^{13}\text{C}$ values of DOC at Stolb in 2016–2017 vary between -28.4% and -27.9% and in 2016 do not differ statistically from values in 2017 (with a mean for both years of $-28.1\% \pm 0.1\%$;

Figure 5). Thus, $\delta^{13}\text{C}$ values of DOC in 2016–2017 are slightly higher than in 2019 ($t(33)=-2.9, p=0.007$) and higher than in winter 2019 ($t(31)=-7.9, p<0.001$).

4 | Discussion

4.1 | Seasonal and Spatial Differences of Organic Carbon Released From the Lena Delta to the Arctic Ocean

In our data set for the Lena Delta, we find DOC concentrations in winter to be higher than in summer, in contrast with previous findings. It has been reported at Zhigansk (ArcticGRO) and Samoylov Island in the Lena Delta from 2014–2019 that DOC concentrations in April (at the end of the winter season) were the lowest (Juhls et al. [38, 45] or were the same as at the end of the summer [62, 63]. We propose several reasons causing this difference between our observations and the published data.

On the one hand, the hydrology of the Lena delta differs from the Lena River main stem [64]. In the Lena Delta, the extensive branching of channels results in a decrease in velocity, leading to the settling and sedimentation of total suspended matter, predominantly occurring on floodplains during the high-water flooding season [10]. In the main stem of the Lena River, active enrichment of suspended matter with subsequent sedimentation occurs on the way to the delta but before reaching it [44, 65]. It is conceivable that DOC concentrations also vary spatially from the river main stem (Zhigansk ~ 800 km south of the delta) to the delta, like spatial patterns of POC [17], making a comparison between our transect and the published data from a single site difficult. In fact, DOC concentrations Delta measured by us in 2019 in the Lena River were lower than average winter (November–April) DOC reported by ArcticGRO ($7.2 \pm 1.9 \text{ mgL}^{-1}$; [66]), supporting a systematic difference between DOC concentrations at Zhigansk and Delta. In addition, the carbon isotopic differences we observe between winter and summer imply that DOC in the delta derives from different sources during the two seasons. Lastly, the year 2019 was extremely warm and dry in summer; August 2019 was the record low on an almost 90 years timeseries [67], resulting in unusually low water discharge ($\sim 19,000 \text{ m}^3 \text{ s}^{-1}$) (Roshydromet, published by Shiklomanov et al. [68]), and subsequently, lower DOC concentrations may have been additionally reduced by higher rates of photo-oxidation in the summer compared to winter.

Our POC results demonstrated opposing patterns relative to the DOC pool. We confirmed an established pattern, where POC flux, as well as water yield and total suspended matter in Arctic rivers, was seen to be lower in winter than in summer [69]. Thus, winter POC concentrations were approximately three times lower than in summer (0.13 ± 0.06 and $0.40 \pm 0.10 \text{ mgL}^{-1}$, respectively). Compared to samples measured upstream in the Lena River (ArcticGRO data, McClelland et al. [66]), deltaic POC concentrations during summer months are generally lower (<50%) and contain a greater contribution of autochthonous-derived (phytoplankton) OM (more enriched in ^{13}C) [17]. As discussed by these authors, more efficient settling of particles in slower flowing waters of the delta compared to the Lena River can be a simple explanation. Nevertheless, POC in the

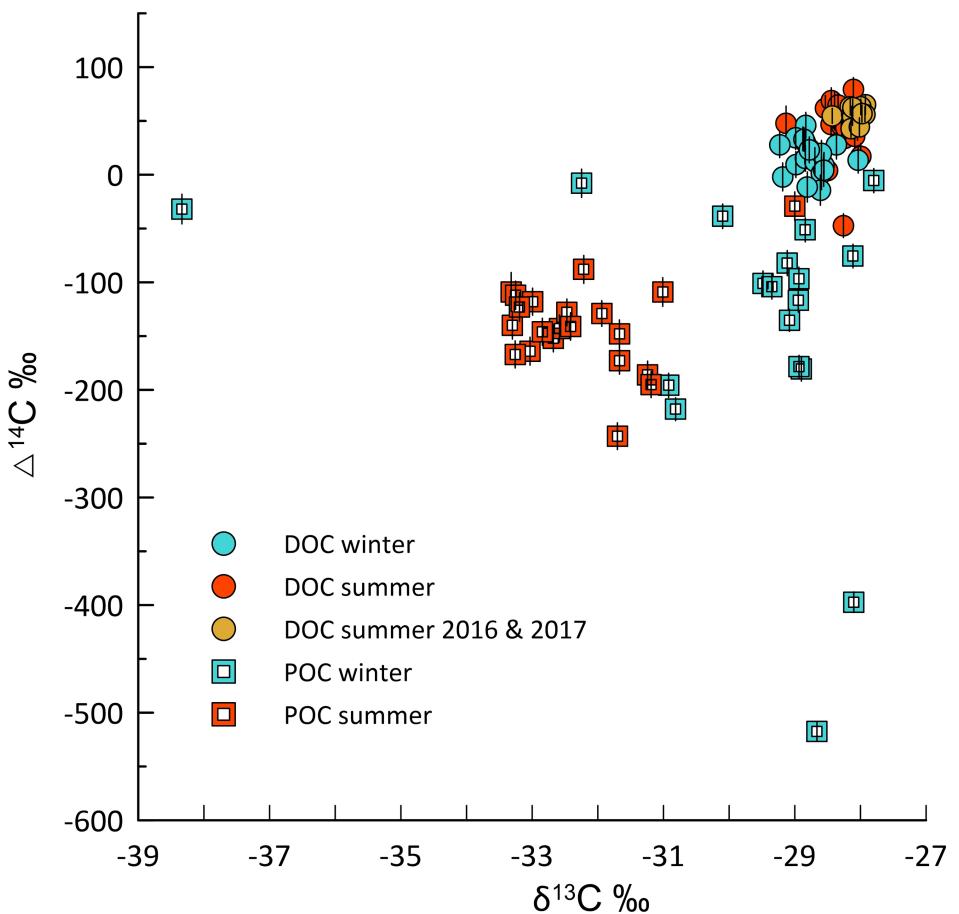


FIGURE 5 | Isotopic composition ($\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$) of dissolved (DOC) and particulate (POC) organic C in the Lena Delta in summer (August 2019) and in winter (April 2019). Additional data are shown for water samples taken at Stolb Island in the summers of 2016 and 2017. [Colour figure can be viewed at wileyonlinelibrary.com]

Lena Delta in winter 2019 did not differ from the average winter (November–April) POC concentration reported by ArcticGRO ($0.14 \pm 0.4 \text{ mg L}^{-1}$) [66].

The highest winter concentrations of DOC (8.54 mg L^{-1}) and POC (0.36 mg L^{-1} , almost three times higher than the average value of $0.13 \pm 0.06 \text{ mg L}^{-1}$) were observed downstream from the Sobo-Sise Yedoma cliff (station CAC19-F). Sanders et al. [10] showed that at this station, water temperature, ammonium, phosphate, and silicon concentrations were likewise higher than at any other sampling station. These combined data indicate strong exchange between water and sediment and suggest a disturbance of river bottom sediment. Meanwhile, all the sediments taken in summer in the delta were very sandy (97.8%–99.7%) with very low content of total organic C (0.1%–0.2%) [10] which makes direct mixing with a sediment an unlikely explanation for the unusual values of the CAC19-F sample, but it could have been an erosional source from the cliff instead of sediment from upwelling.

4.2 | Main Sources and Transformation Patterns of Organic Carbon in the Lena Delta

Generally, sources of DOC can be categorized into three groups based on the seasonal hydrological shifts. During the spring

freshet, DOC is reported to primarily originate from surface runoff, including leached components from fresh litter of angiosperm and gymnosperm plants, which have young ^{14}C ages [62, 70]. In late summer and winter, when the dissolved OM concentrations are the lowest, dissolved OM sources in Arctic rivers are associated with peat of different origin and the near-surface thawed soil layers, which exhibit older radiocarbon signatures indicative of drainage from deeper soil horizons [71, 72].

Using the isotopic composition of DOC measured in our samples, we investigated whether these general patterns can be confirmed also for the Lena Delta. The stable C isotopic composition of DOC was statistically indistinguishable between the sampled seasons ($-28.33\% \pm 0.26\%$ in summer, $-28.75\% \pm 0.26\%$ in winter) and remained within the range of contemporary OC pools [27], including vegetation, surface soils, and surface active layer signals. Kutscher et al. [73] similarly demonstrated that vegetation was the dominant source of DOC in the Lena River and its main tributaries, with its contribution to DOC increasing downstream. Thus, $\delta^{13}\text{C}$ of DOC cannot be used as a parameter to describe the difference between summer and winter DOC. However, combined $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ evidence reveals distinct patterns.

In summer, the DOC in the Lena River and its Delta is characterized by modern carbon, and its combined $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$

values suggest that it originates from vascular plants and from autochthonous primary production, such as algal-derived OM and does not contain any contributions from fossil materials, excluding Yedoma as a major source. The $\Delta^{14}\text{C}$ signal of modern plants is assumed to reflect the radiocarbon composition of the atmosphere during their growth, which was enriched in ^{14}C due to nuclear bomb testing in the 1950s, and $\Delta^{14}\text{C}$ values have remained above 0 until around 2020. Atmospheric $\Delta^{14}\text{C}$ in 2016–2019 ranged between 0‰ and 13‰ [74]. The measured $\Delta^{14}\text{C}$ of $41\text{‰} \pm 26\text{‰}$ for DOC in summer 2019 was above atmospheric levels of that year. Instead, these higher $\Delta^{14}\text{C}$ values for DOC from the Lena Delta correspond to the atmospheric values observed in the previous decade (2001–2009) [75].

Water percolating through the active layer in the polygonal tundra often forms small ponds in the central depressions of the low-centered polygons. Our observations on DOC from three small polygon lakes on Samoylov Island, with $\Delta^{14}\text{C}$ values of $45\text{‰} \pm 19\text{‰}$ (Figure 6), support our suggestion that DOC in the river is derived from leaching of the active layer, which may contain elevated ^{14}C levels inherited from the growing seasons of previous decades. The sampling of these ponds took place in the time of the year when the active layer in the Lena Delta reaches its maximum thickness [76]. This might suggest that during late summer, the oldest DOC from the thawing permafrost would be expected. However, due to nuclear weapons testing, atmospheric ^{14}C levels in recent decades were higher than they are now, which explains why “young” or even “above modern” values are obtained for DOC that might be leached from soil layers that have accumulated during previous decades. This suggests that as the active layer deepens and permafrost thaws, the ^{14}C signatures in DOC leached from these layers may initially increase and only begin to decrease when older permafrost OM accumulated before 1950 is released. This pattern

should be considered when studying the age and composition of DOC. We acknowledge that using three lakes on one island in the delta to interpret the deltaic signal is not sufficient but may provide some insight into the terrestrial background.

Our data from Stolb in 2016–2017 and the deltaic transect in 2019 do not show clear differences in $\Delta^{14}\text{C}$ values of DOC. Because the active layer on Samoylov Island in 2019 was only 16 and 14 cm thicker than in 2016 and 2017 [77], respectively, differences in the mean ^{14}C content of the active layers of the respective years are thus likely too small to be resolved within our measurement uncertainty.

A distinct contrast between the seasons was found for the $\Delta^{14}\text{C}$ of DOC in the Lena Delta, with winter DOC exhibiting older ages (lower $\Delta^{14}\text{C}$) compared to summer ($\Delta^{14}\text{C}$: $-16\text{‰} \pm 16\text{‰}$ and $41\text{‰} \pm 26\text{‰}$, respectively). This finding aligns with some previous studies [8,63] but differs from others such as Behnke et al. [62], who reported minimal differences between winter and summer $\Delta^{14}\text{C}$ signals for DOC in the Lena River. During the low discharge period in winter, DOC is reported to be mainly transported by groundwater [45, 70, 78], suggesting an input of aged carbon from terrestrial sources. The majority of the Lena River catchment is underlain by continuous permafrost (70.5%) [45,46] making intra-permafrost groundwater [79] draining taliks [62], the more likely type of groundwater for the Lena Delta. If this permafrost contributed to the Lena deltaic DOC in winter, an older radiocarbon signal would be expected. However, the DOC $\Delta^{14}\text{C}$ values in winter resemble those of the atmosphere during the time of sampling, suggesting that a considerable proportion of this DOC derives from OM that was photosynthetically produced during the respective years. The potential influence of interannually and seasonally pooled waters from the Vilui reservoir [45], which accounts for a significant percentage of the water that is discharged via the Lena River in winter [80], further complicates the interpretation.

POC in summer was depleted in ^{13}C ($\delta^{13}\text{C}$: $-32.4\text{‰} \pm 0.8\text{‰}$), likely due to contributions from aquatic organisms and the potential effect of an algal bloom taking place at the end of the summer in August [17]. Algal blooms in rivers can result in low $\delta^{13}\text{C}$ values of POC as the DIC pool is drawn down by photosynthesis and then replenished by respiration and transport of DIC from upstream (Finlay and Kendall, 2008). In winter, different sources of POM would be expected. Winter $\delta^{13}\text{C}$ of POC in the Lena Delta is higher than in summer (mean $-29.7\text{‰} \pm 2.2\text{‰}$) and corresponds to the typical terrestrial $\delta^{13}\text{C}$ values according to Vonk et al. [81]. However, it was lower than the $\delta^{13}\text{C}$ values directly measured for Holocene soils ($-26.6\text{‰} \pm 1.0\text{‰}$) and Yedoma ($-26.3\text{‰} \pm 0.7\text{‰}$) in the region [8, 82]. In contrast to our findings, analyses of POC annual fluxes provided by the ArcticGRO initiative [66] revealed that unlike in the Lena Delta winter, POC in Arctic rivers is more depleted in ^{13}C than summer POC. These authors reported winter and summer mean $\delta^{13}\text{C}$ values of POC at Zhitansk in the Lena River as $-32.8\text{‰} \pm 1\text{‰}$ and $-29.0\text{‰} \pm 0.4\text{‰}$, respectively, showing the opposite contrast between seasons. As discussed in Ogneva et al. [17] for the summer, this discrepancy could be attributed to the combined factors of low flow velocity in the shallow delta channels, where sunlight penetrates much of the water column containing smaller amounts of suspended particles, and the extremely warm summer conditions during our sampling campaign in 2019, leading to high primary

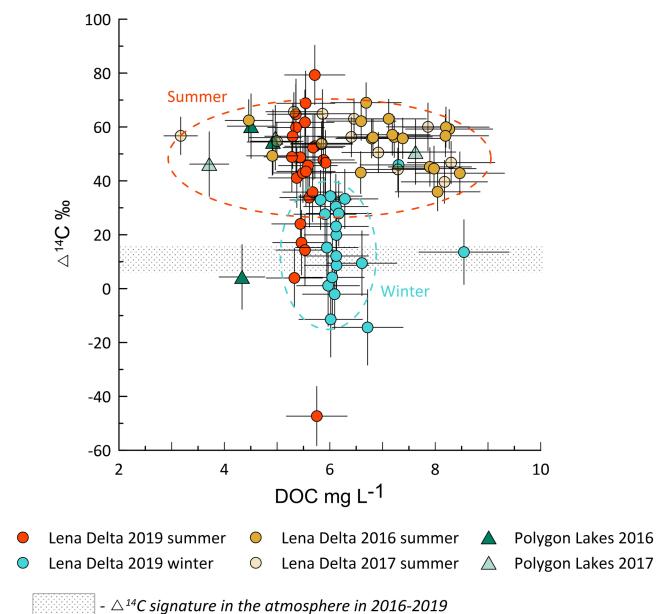


FIGURE 6 | DOC concentration and its radiocarbon content in the Lena Delta (2016, 2017, 2019) and in the Polygon Lakes (2016, 2017) at Samoylov Island. The atmospheric $\Delta^{14}\text{C}$ signature in 2016–2019 is based on Emmenegger et al. [74]. [Colour figure can be viewed at wileyonlinelibrary.com]

production and low $\delta^{13}\text{C}$ values of POC in the Lena Delta. By contrast, McClelland et al. [69] reported lower winter $\delta^{13}\text{C}$ than ours, which was explained by contributions from bacterial communities, particularly methanotrophs, a process that appears to be less active in the shallow delta channels.

We found no significant difference between the summer and winter $\Delta^{14}\text{C}$ signals of POC, contrary to our expectation of a more ^{14}C -depleted signal in winter indicative of less phytoplankton contribution in winter. The lack of a radiocarbon age difference might also suggest that aquatic production uses the same carbon source as is contained in POC. This suggests an additional modern source of POC in winter, potentially aquatic bacterial communities. Since the concentration of POC in winter is very low ($0.13 \pm 0.06 \text{ mg L}^{-1}$) contributions from methanogenic bacteria may have a significant impact on the composition of winter POC.

5 | Conclusion

We found that the amount and composition of DOC and particulate organic carbon (POC) in the Lena Delta vary between the summer and winter seasons. Surprisingly, despite lower water flow in winter, the concentration of DOC in the Lena Delta was higher during this season as compared to the end of summer. The sources of DOC also differ between the two seasons. In summer, the DOC is enriched in $\Delta^{14}\text{C}$, likely due to plant debris from the post-1950s period. In contrast, winter DOC contains slightly older carbon. This study provides a unique dataset on the largest Arctic delta, which will be key to assess the ongoing changes with climate warming at this frontier between the land and the ocean realm. Winter $\delta^{13}\text{C}$ of POC in the Lena Delta is higher than in summer, when it was depleted due to the August 2019 algal bloom. However, no significant difference in summer and winter $\Delta^{14}\text{C}$ signals of POC was observed, suggesting an additional modern winter source, potentially aquatic communities such as methanogenic bacteria.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data presented in this study are freely available in the PANGAEA data repository, [61] (10.1594/PANGAEA.962633). POC data are available from [83] (10.1594/PANGAEA.950668).

Supporting Information

Supporting information (Table S1, including the sample location descriptions) accompanies this manuscript.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. **Table S1:** Sample station locations in the Lena Delta along the Sardakhskaya branch from 2019 and sampling locations in the Lena Delta in previous years. Sampling depth equal to 0.5 m in summer season means that samples were taken from the surface.