



Origin and Pathways of Dissolved Organic Carbon in a Small Catchment in the Lena River Delta

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The Arctic is rich in aquatic systems and experiences rapid warming due to climate change. The accelerated warming causes permafrost thaw and the mobilization of organic carbon. When dissolved organic carbon is mobilized, this DOC can be transported to aquatic systems and degraded in the water bodies and further downstream. Here, we analyze the influence of different landscape components on DOC concentrations and export in a small (6.45 km²) stream catchment in the Lena River Delta. The catchment includes lakes and ponds, with the flow path from Pleistocene yedoma deposits across Holocene non-yedoma deposits to the river outlet. In addition to DOC concentrations, we use radiocarbon dating of DOC as well as stable oxygen and hydrogen isotopes ($\delta^{18}\text{O}$ and δD) to assess the origin of DOC. We find significantly higher DOC concentrations in the Pleistocene yedoma area of the catchment compared to the Holocene non-yedoma area with medians of 5 and 4.5 mg L⁻¹ ($p < 0.05$), respectively. When yedoma thaw streams with high DOC concentration reach a large yedoma thermokarst lake, we observe an abrupt decrease in DOC concentration, which we attribute to dilution and lake processes such as mineralization. The DOC ages in the large thermokarst lake (between 3,428 and 3,637 ¹⁴C y BP) can be attributed to a mixing of mobilized old yedoma and Holocene carbon. Further downstream after the large thermokarst lake, we find progressively younger DOC ages in the stream water to its mouth, paired with decreasing DOC concentrations. This process could result from dilution with leaching water from Holocene deposits and/or emission of ancient yedoma carbon to the atmosphere. Our study shows that thermokarst lakes and ponds may act as DOC filters, predominantly by diluting incoming waters of higher DOC concentrations or by re-mineralizing DOC to CO₂ and CH₄. Nevertheless, our results also confirm that the small catchment still contributes DOC on the order of 1.2 kg km⁻² per day from a permafrost landscape with ice-rich yedoma deposits to the Lena River.

Keywords: Arctic lakes, ice complex, yedoma, thermokarst lakes, DOC, aquatic carbon cycle, permafrost, radiocarbon dating

INTRODUCTION

The Arctic experienced an increase of averaged near-surface air temperatures by 3.1°C since the 1970s (AMAP, 2021), which is accompanied by warming of Arctic permafrost (Biskaborn et al., 2019). The Arctic is rich in aquatic systems, which interact with the thawing permafrost, and Arctic warming has a strong influence on biogeochemical processes in these aquatic systems (Frey and McClelland, 2009; Wik et al., 2016). In particular, ice-rich permafrost, such as yedoma, is prone to rapid thaw causing ground-ice melt and associated volume loss. As a result, the land surface subsides and landforms like thermo-erosional valleys, thaw slumps, and thermokarst lakes develop. Thermokarst lakes and drained lake basins may regionally occupy three quarters of the yedoma landscape surface (Jones et al., 2011; Grosse et al., 2013). The presence of thermokarst lakes accelerates the thaw of permafrost (Langer et al., 2016), which delivers freshly thawed sediments and organic matter into water bodies (Turetsky et al., 2020). Here, soil carbon (C) can be mobilized and enters aquatic systems as dissolved organic C (DOC) and particulate organic C (POC), whereas DOC, once mobilized, can be degraded microbially or photo-chemically and emitted to the atmosphere as C dioxide (CO₂) and methane (CH₄) (Frey & Smith, 2005; Battin et al., 2008; Tranvik et al., 2009; Vonk et al., 2013a,b). Lakes and especially thermokarst lakes release a significant amount of CO₂ and CH₄ and are biogeochemical hotspots since they act as turnover of organic C (Abnizova et al., 2012; Walter Anthony et al., 2016; Evans et al., 2017; Hughes-Allen et al., 2021). Since these processes may amplify climate change (Schuur et al., 2015), investigations of DOC contributions to C cycling in Arctic freshwater systems on different scales are necessary to understand and predict climate change impacts and permafrost region C cycle feedbacks.

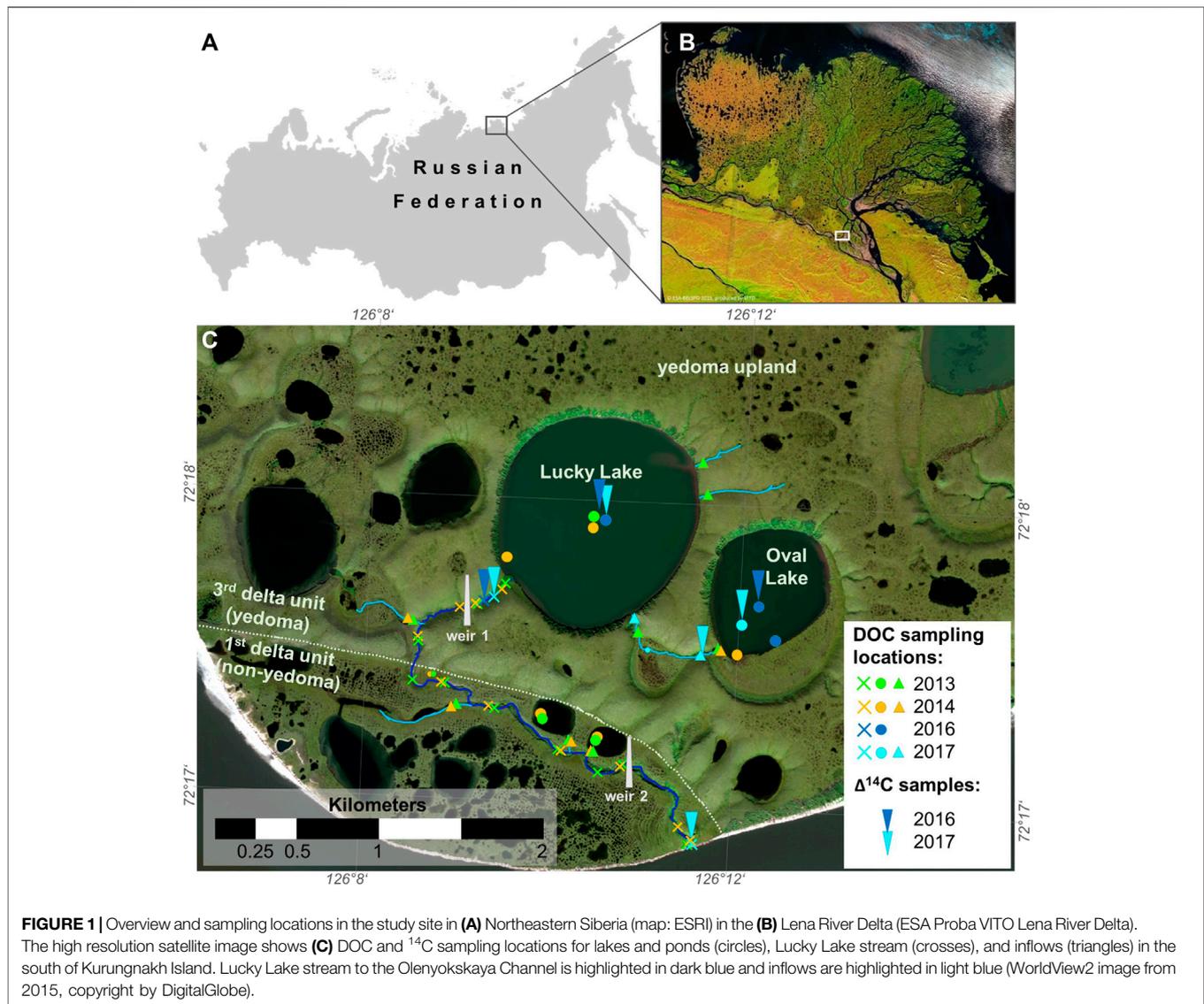
From non-yedoma permafrost landscapes in western Siberia, we know that DOC concentration decreases during the formation of thermokarst lakes from thermokarst depressions to ponds and thermokarst lakes, and increases after lake drainage due to autochthonous DOC production (Manasypov et al., 2014). The DOC concentrations in surface waters in thermokarst dominated permafrost landscapes vary with size (e.g. lake surface area), vegetation density, hydrological connectivity, and water residence time (Evans et al., 2017). Hydrological connectivity and water residence time influence the re-mineralization of DOC by lake processes, such as photo-degradation, microbial activity, and/or flocculation. Therefore, lakes with short water residence time typically have higher DOC concentrations (Manasypov et al., 2015). A recent pan-Arctic synthesis study for DOC in permafrost lakes suggests that there are differences between yedoma and non-yedoma lakes and that lake DOC concentrations can be highly variable, likely based on local environmental factors (Stolpmann et al., 2021a). For Arctic rivers and streams, thawing permafrost may lead to a shift of DOC fluxes. Previous studies found increasing DOC mobilization due to permafrost thaw (Frey and Smith, 2005; Guo et al., 2007; Drake et al., 2015; Mann et al., 2015; Spencer et al., 2015), and Littlefair et al. (2017) highlight the high variation of DOC mobilization in permafrost thaw-affected landscapes on a

regional scale. (Kokelj et al., 2005). found that thermokarst, such as thaw slumping, regionally results in DOC mobilization decrease. Contrastingly, Spencer et al. (2015) found increasing DOC mobilization due to thermokarst processes, which suggests that the amount of mobilized DOC depends on the composition of the eroding material in different permafrost landscapes. In regions underlain by yedoma deposits, a rapid loss of ancient DOC was observed in connection with DOC age, bioavailability, and metabolism (Mann et al., 2015; Spencer et al., 2015). For the Siberian Kolyma River basin, incubation experiments showed microbial utilization of ancient organic C (OC) resulting in decreasing DOC concentrations and younger $\Delta^{14}\text{C}$ signatures downstream (Mann et al., 2015). The latter was confirmed by Rogers et al. (2021), highlighting the export of predominantly modern DOC to the Kolyma River. In contrast, Mann et al. (2015) found that small yedoma thaw streams featured higher DOC concentrations compared to larger main river channels. Kling et al. (2000) investigated the influence of lakes and rivers on a landscape mass balance and found that lake and river processes are critical for surface water chemistry. For delta systems, it was found that river deltas, especially with lake-rich floodplains, affect nutrient fluxes to the Arctic Ocean (Emmerton et al., 2008). Their model showed enhanced DOC in stream water flowing through the Mackenzie Delta. However, the contribution and influence of lakes and ponds, which are interposed in a stream catchment, on the DOC concentration and how the concentration changes due to these different landscape units are unexplored in yedoma permafrost regions.

Since we assume that DOC modification processes such as photo-oxidation are more prominent in standing waters, we hypothesize a measurable influence of lakes and ponds that are part of a runoff network on DOC processing within the stream catchment. Our goal in this study is to assess the influence of interposed lakes and ponds on DOC in a small stream catchment with and without the influence of yedoma deposits before the DOC reaches the main channels of the Lena River Delta. The specific objectives of this paper are to (1) characterize differences in DOC concentrations in different surface waters, (2) determine DOC ages, and (3) analyze changes in DOC concentrations and the DOC contribution along a flow path in a small yedoma influenced catchment. With this, our study provides insights on the origin of DOC in surface waters in yedoma landscapes and how DOC may be transformed on a flow path through different landscape units before reaching the river outlet.

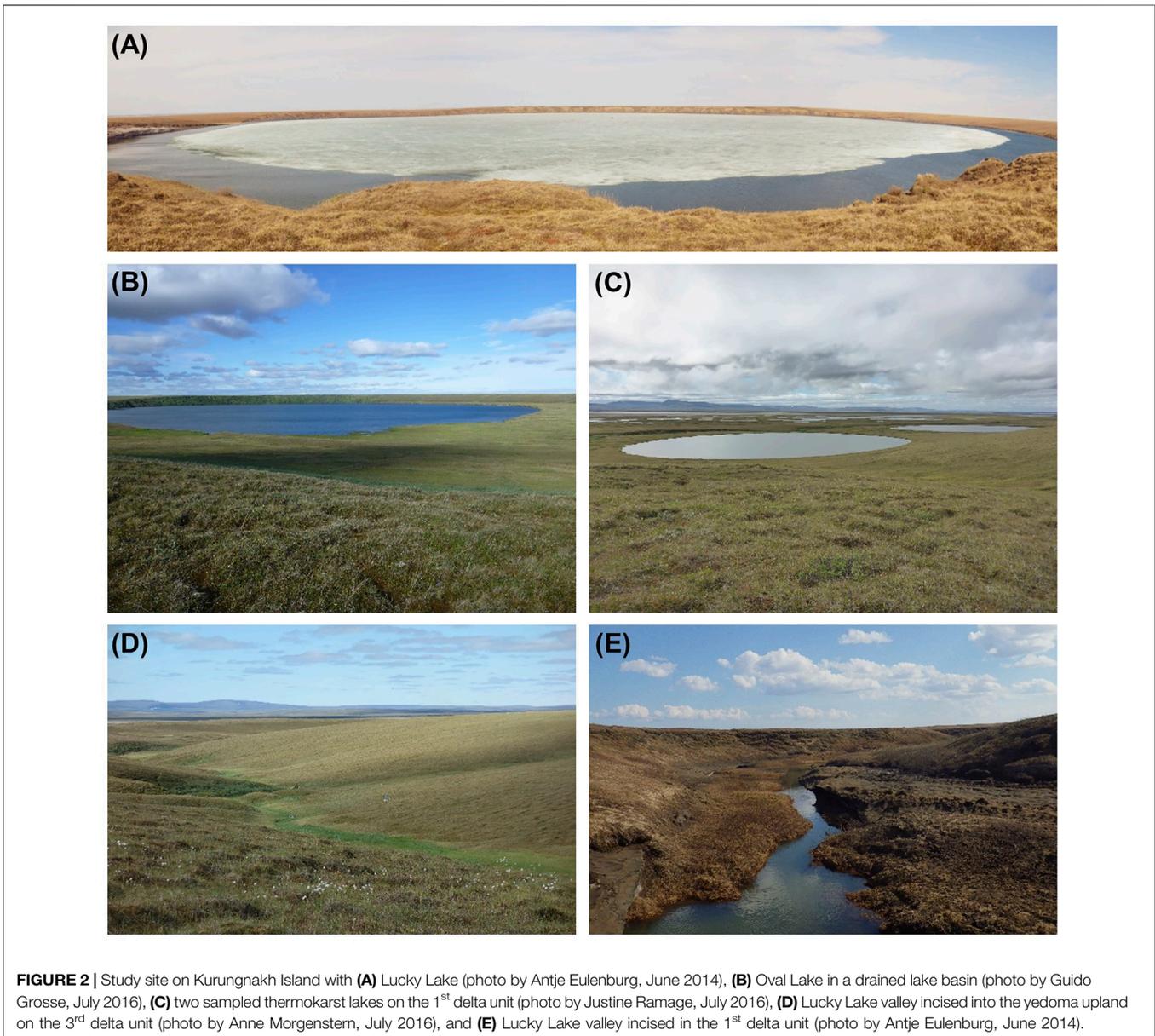
STUDY SITE

Our study site on Kurungnakh Island (72° 23'N; 126° 03'E) is located in the central Lena River Delta in the continuous permafrost and subarctic tundra zone. The Lena Delta is divided into three main geomorphological units (Schwamborn et al., 2002). Our study site is situated partially on the 1st and on the 3rd Lena Delta unit. Whereas both units consist of ice-rich permafrost, they differ in depositional age, soil C stock characteristics, and watershed hydrology (Figure 1C). The 1st



is the youngest geomorphological unit representing the modern active delta. It was formed in the middle Holocene and is characterized by active floodplains and polygonal tundra with small and shallow lakes and ponds, and ice-wedges. The 3rd delta unit is the oldest unit of the Lena River Delta and was formed during the Late Pleistocene. It consists of remnants of yedoma deposits, which accumulated between 43 and 14 ka BP, overlies fluvial sands and is covered by Holocene deposits (Schwamborn et al., 2002). Kurungnakh Island belongs mainly to the 3rd Lena Delta unit (Grigoriev, 1993) and is characterized by typical yedoma landforms such as yedoma uplands with surface elevations of up to 55 m above river level (Morgenstern et al., 2011) and deeply incised thermo-erosional valleys, thermokarst depressions, and large thermokarst lakes, what formed since about 13 to 12 ka BP (Morgenstern et al., 2013). Kurungnakh Island is located close to Samoylov Island, which hosts the Research Station Samoylov Island that facilitated organization and logistics of our local field work.

Our study site is located in the south of Kurungnakh Island and comprises the catchment of Lucky Lake (unofficial name). On both units, supra-permafrost aquifers are restricted to the shallow active layer (Helbig et al., 2013). On the 3rd delta unit of Kurungnakh Island the active layer depth ranges from 38 to 82 cm (Ulrich et al., 2010). For active layer on the 1st delta unit we refer to data from Samoylov Island, where the mean depth ranges from 41 to 57 cm (Boike et al., 2019). The catchment includes several thermokarst lakes and ponds as well as inflowing and outflowing streams, both on the 3rd (yedoma) and on the 1st (non-yedoma) delta units. The outflowing stream and its associated valley are referred to as Lucky Lake stream and Lucky Lake valley, respectively. This study site offers the opportunity to analyze DOC released on the yedoma and non-yedoma delta units and how DOC is transformed along this flow path and reaches the Olenyokskaya Channel (the Lena River branch into which the Lucky Lake stream drains) (Figure 1). Lucky Lake and its neighbor Oval Lake (also unofficial name), which is located in



a partially drained lake basin, are both large thermokarst lakes that are deeply subsided into the local yedoma upland (**Figure 2**). Lucky Lake covers a surface area of 1.22 km², with a mean depth of 3.1 m and a maximum depth of 6.5 m, and is bordered by steep slopes with active erosion. Two inflows from the yedoma upland drain into Lucky Lake in the northeast. In the southwest, Lucky Lake drains into Lucky Lake stream via the yedoma upland and the 1st delta unit into the Olenyokskaya Channel. The Oval Lake has a surface area of 0.45 km² and a maximum depth of 9 m (Morgenstern et al., 2011).

Dry Arctic-continental climate with a mean annual air temperature of -13.6°C and a mean annual precipitation of about 140 mm characterize the climate in the Lena Delta (Boike et al., 2008). Due to these extreme climate conditions the vegetation period is limited to 3 months from mid-June to

mid-September. According to the two different geomorphological delta units in our study site, vegetation differs between 3rd and 1st delta unit. Whereas the 1st delta unit is covered with wet, sedge and moss as well as moist grass dominated tundra vegetation, the 3rd is dominated by dry tussock and moist grass and moss tundra (Schneider et al., 2009).

MATERIALS AND METHODS

For our analysis, we collected 113 surface water samples in the Lucky Lake catchment on the 3rd delta unit with yedoma deposits (61 samples) and the 1st delta unit (52 samples). The samples were collected during summer expeditions in July and August 2013, June to September 2014, July and August 2016 and July 2017

(**Figure 1**). A total of 30 samples were from Lucky Lake and the surrounding lakes on both delta units, 27 samples of inflows, such as from neighboring ponds and yedoma uplands, and 56 samples from the Lucky Lake stream and crossing both delta units. We analyzed concentration and radiocarbon ages of DOC, stable isotopes of oxygen ($\delta^{18}\text{O}$) and hydrogen (δD), and obtained discharge measurements to examine the origin of DOC and to understand transformations of DOC on the flow path in our study site. We measured the DOC concentration of all samples, and $\delta^{18}\text{O}$ and δD of 99 samples. During the two sampling campaigns in 2016 and 2017, we were able to obtain 9 additional samples large enough to perform radiocarbon measurements of DOC.

Laboratory Analysis

DOC Measurements

For DOC measurements, we filtered the water samples in the field with a 0.7 μm pore size fiberglass (GF/F) syringe filter. Sampling bottles and filters were pre-rinsed with the sample. All samples were preserved with hydrochloric acid and kept cool and dark until analysis, which sometimes occurred months after sampling. In the lab, we used the non-purgeable OC method (NPOC) with the Shimadzu TOC-VCPH high-temperature catalytic combustion (Manual Shimadzu/TOC-V, 2008), recording DOC concentration in milligrams per liter (mg L^{-1}).

Radiocarbon Dating

We carried out Accelerator Mass Spectrometry (AMS) radiocarbon (^{14}C) dating with a Mini Carbon Dating System (MICADAS), which is described by Synal et al. (2007), and followed the methods outlined in Mollenhauer et al. (2021). In short, we dried our samples with a Heidolph LABORATA roto-evaporation apparatus to extract dissolved organic matter (DOM). Dried DOM samples were subsequently transferred into 50 μL liquid tin capsules using Milli-Q water, and radiocarbon analyses were conducted after combustion in an Elementar element analyzer using helium at 950°C. The gas mixture is transmitted to a gas interface system, described by Ruff et al. (2010), and further to the CO_2 accepting ion source of the AMS instruments. We report results as $\Delta^{14}\text{C}$ and as conventional radiocarbon ages (^{14}C years before present).

Stable Isotopes of $\delta^{18}\text{O}$ and δD

We measured stable isotopes of $\delta^{18}\text{O}$ and δD with a Finnigan MAT Delta-S mass spectrometer in the isotopic laboratory of Alfred Wegener Institute in Potsdam, applying the equilibration technique described by Meyer et al. (2000).

Discharge and DOC Flux

We conducted discharge measurements with two weirs with combined radar height sensors installed in the Lucky Lake stream on the 3rd and on the 1st delta unit during field work in 2013, when also DOC samples from the Lucky Lake stream were collected (**Figure 1**). The sill referenced water level in millimeter (mm) was measured in 10 min intervals and was subtracted from the sensor height. The discharge (Q) in liter per seconds (L s^{-1}) was calculated as follows:

$$Q = 0.0000004 \cdot (WL)^3 + 0.0011 \cdot (WL)^2 + 0.1358 \cdot WL - \sqrt{WL} + 3.488 \quad (1)$$

where WL is the sill referenced water level (Eijkelkamp, 2010). We converted the discharge into cubic meters per day ($\text{m}^3 \text{d}^{-1}$). The discharge data cover the time period from July 27, 2013 to August 26, 2013. Additionally, we estimated a watershed area of 6.45 km^2 for the entire study site above weir 2. Using the weir data, we calculated the DOC flux (kg km^{-2}) as a product of summed discharge and averaged concentration of DOC of Lucky Lake stream water samples at each weir for this time period, and calculated a DOC flux per day by dividing the DOC flux with 29, which is the number of discharge measurement days.

Statistical Analysis

For our statistical analysis we used RStudio (version 4.0.5). To test the distribution of values of our parameter (DOC concentration, $\delta^{18}\text{O}$, δD , and $\Delta^{14}\text{C}$) we used the Shapiro-Wilk normality test. Because our data do not follow a normal distribution we used the Spearman rank correlation coefficient (ρ) to measure the relationship between two parameters. We tested differences between mean values of a parameter by delta terrace or water source (lake water, stream water, and inflow water) using the Wilcoxon-Mann-Whitney test.

RESULTS

DOC Concentration

The DOC concentration in our study site ranged from 2.9 to 15.6 mg L^{-1} . The median was 5 mg L^{-1} . We found the highest DOC concentration of the entire catchment of 15.6 mg L^{-1} in a small polygonal pond on Holocene non-yedoma. Generally, we found significantly higher DOC concentrations in surface waters on Pleistocene yedoma (median DOC of 5 mg L^{-1} ; exception see above) compared to surface waters on Holocene non-yedoma (median DOC of 4.5 mg L^{-1}) (**Figure 3A**; $\rho = 0.4$; $p < 0.05$). Regarding surface waters on Pleistocene yedoma unit, we found the highest DOC concentrations of 9–12.7 mg L^{-1} in inflows from the yedoma upland in the northeast of Lucky Lake. For inflows, we found significantly higher DOC concentrations and a larger range in DOC concentration on Pleistocene yedoma compared to Holocene non-yedoma, with median DOC concentrations of 4.1 and 3.3 mg L^{-1} , respectively ($p < 0.05$; **Figure 3D**; **Table 1**). For lakes, the median DOC concentration was higher on Pleistocene yedoma with 5.2 mg L^{-1} compared to Holocene non-yedoma with 3.6 mg L^{-1} (**Table 1**). However, this difference was not statistically significant. Additionally, we found a larger range of lake DOC concentration on Holocene non-yedoma (**Figure 3B**). For Lucky Lake stream, we found generally higher DOC concentrations on Pleistocene yedoma compared to Holocene non-yedoma, with median DOC concentrations of 5.2 mg L^{-1} and 4.8 mg L^{-1} , respectively (**Figure 3C**; **Table 1**). We analyzed changes in DOC concentration downstream in the catchment. Therefore, we defined Lucky Lake inflows, including yedoma thaw streams from the yedoma upland and Oval Lake, as the beginning of the flow path, draining into Lucky

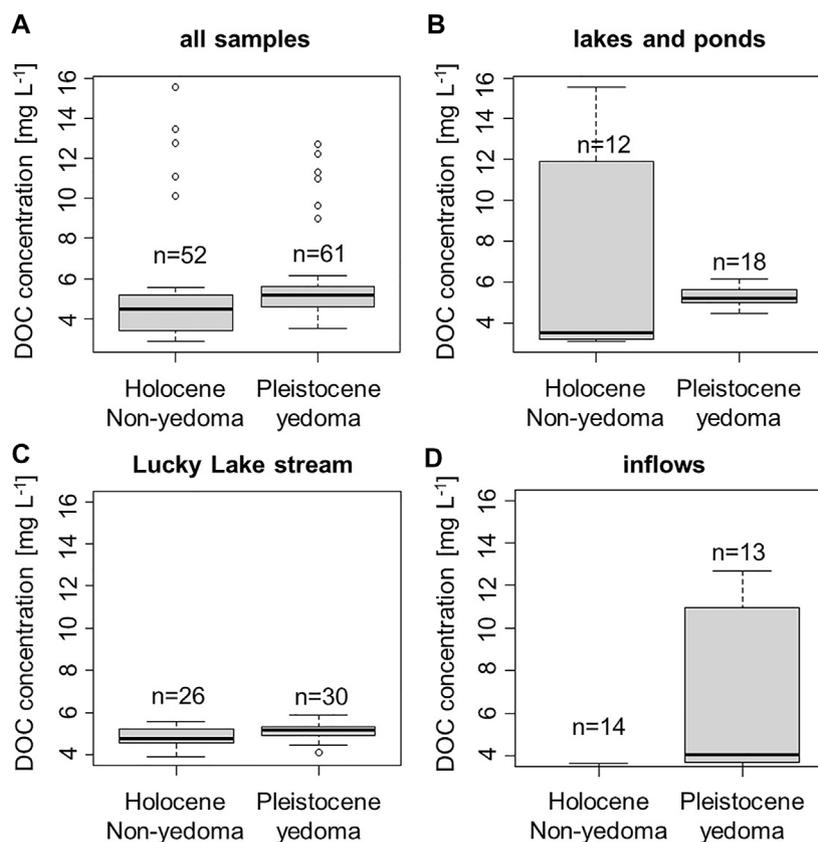


FIGURE 3 | Comparison of DOC concentrations of (A) all samples in the Lucky Lake catchment, (B) samples from lakes and ponds, (C) samples from Lucky Lake stream, and (D) from inflows; by Holocene non-yedoma and Pleistocene yedoma. The greyish shaded boxes represent the standard deviation from the mean (solid line).

TABLE 1 | Overview of DOC concentration and isotopic composition for lakes, Lucky Lake stream, and inflows on Pleistocene yedoma and Holocene non-yedoma from the years 2013, 2014, 2016, and 2017.

		Pleistocene yedoma			Holocene non-yedoma		
		Lakes	Stream	Inflows	Lakes	Stream	Inflows
DOC concentration [mg L ⁻¹]	range	4.5–6.2	4.1–5.9	3.5–12.7	3.1–15.6	3.9–5.6	2.9–3.6
	median	5.2	5.2	4.1	3.5	4.8	3.3
$\delta^{18}\text{O}$ (‰) vs. SMOW	range	-17.8–7.1	-18.8–16.8	-19.6–16.2	-17.9–15.3	-20.7–16.9	-16.4–14.7
	median	-17.2	-17.4	-16.8	-16.1	-17.1	-15.6
δD (‰) vs. SMOW	range	-138.3–135.4	-146.4–133	-144.9–125.5	-138.9–123.2	-160.8–132.7	-131.1–119.4
	median	-137.3	-138.1	-131.5	-128.9	-135.3	-125.4

Lake, which then drains into the Lucky Lake stream, and subsequently discharges into the Olenyokskaya Channel (Figure 4). Along this flow path, we found decreasing DOC concentrations downstream to the Olenyokskaya Channel.

Radiocarbon Content of DOC

To define the origin of DOC in our study site, we conducted DOC radiocarbon analyses for a total of 9 samples. The $\Delta^{14}\text{C}$ values ranged from -373 ‰ (3,637 ¹⁴C y BP) to -300 ‰ (2,782 ¹⁴C y BP) (Figure 4). We found lowest $\Delta^{14}\text{C}$ values in the Lucky Lake samples and highest $\Delta^{14}\text{C}$ values in Oval Lake samples and in

the Lucky Lake stream at the mouth to Olenyokskaya Channel (Table 2). We found decreasing $\Delta^{14}\text{C}$ values from Oval Lake to Lucky Lake and increasing $\Delta^{14}\text{C}$ values from Lucky Lake to the mouth at the Olenyokskaya Channel.

Stable Isotopes of $\delta^{18}\text{O}$ and δD

To discuss the origin of the sampled surface waters in our study site we used the stable isotopic composition of the water samples. We found a negative correlation between DOC concentration and water isotopic composition with significantly lower values in $\delta^{18}\text{O}$ and δD in samples of higher DOC concentration ($\rho = -0.57$ and -0.45 ,

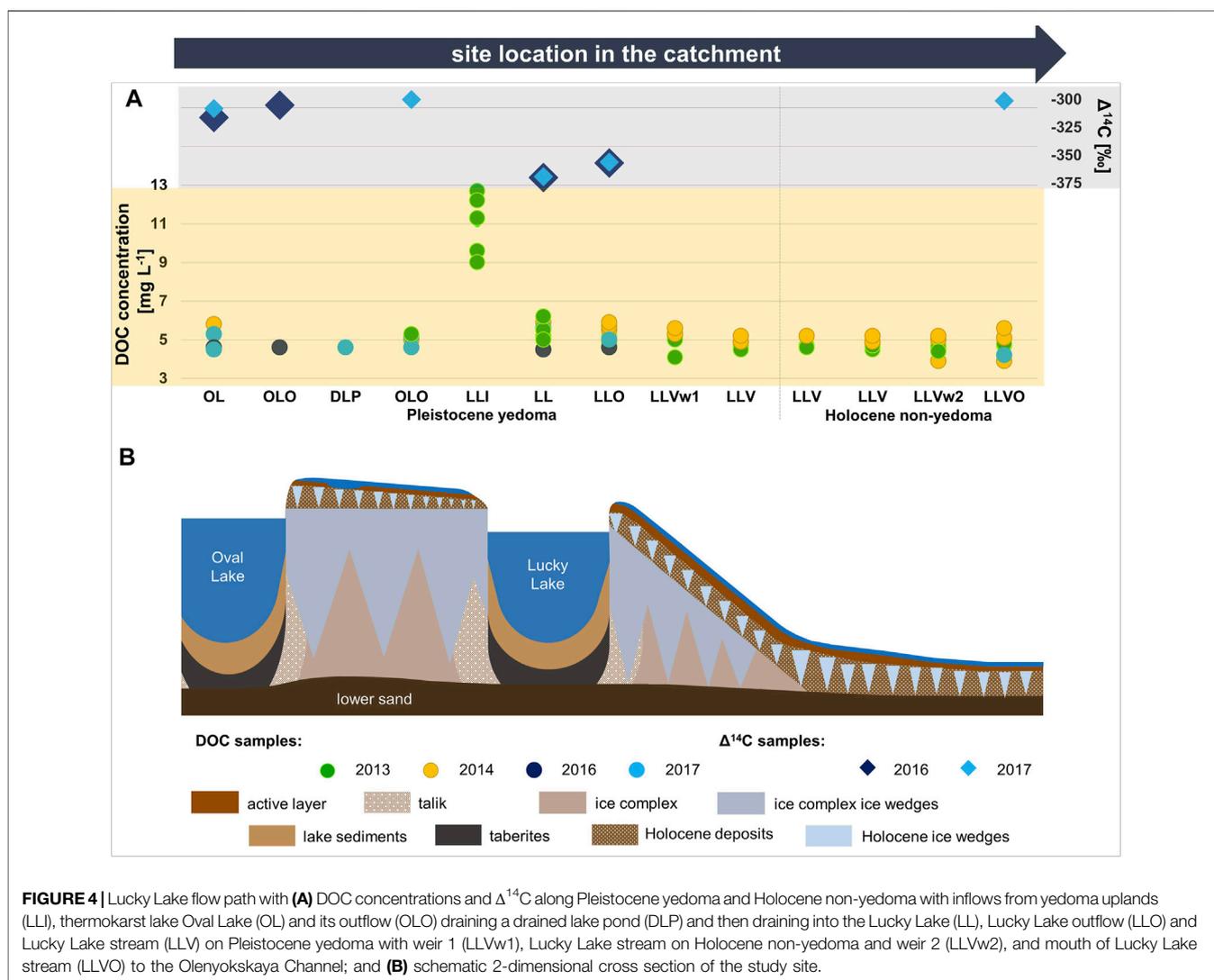


TABLE 2 | Results of the radiocarbon analyses of DOC samples collected in July and August 2016 and July 2017.

Sample location	2016		2017	
	$\Delta^{14}\text{C}$ [‰]	Age [^{14}C y BP]	$\Delta^{14}\text{C}$ [‰]	Age [^{14}C y BP]
OL - Oval Lake, lake center	-322	3,021	-308	2,871
OLO—Oval Lake, outflow	-305	2,840	-300	2,782
LL—Lucky Lake, lake center	-370	3,597	-371	3,613
LL—Lucky Lake, before outflow	-373	3,637	-356	3,428
LLO—Lucky Lake, outflow	-357	3,442		
LLVO—Lucky Lake stream, mouth to Olenyokskaya Channel			-301	2,783

respectively; $p < 0.05$). The stable isotopic composition ranged from -21.3 to -14.7 ‰ VSMOW in $\delta^{18}\text{O}$ and from -162.6 to -119.4 ‰ VSMOW in δD . Most of the samples fell below the Global Meteoric Water Line (GMWL) (Figure 5). For lakes and ponds, we found lower values for $\delta^{18}\text{O}$ and δD on Pleistocene yedoma compared to Holocene non-yedoma, and increasing values for $\delta^{18}\text{O}$ from Lucky Lake via the Lucky Lake stream from Pleistocene yedoma to

Holocene non-yedoma. Additionally, two samples with lowest $\delta^{18}\text{O}$ and δD were outliers from the Lucky Lake stream on Holocene non-yedoma.

Water isotopic values of one sample lay close (within δD 10 ‰ and $\delta^{18}\text{O}$ 1 ‰) to the GMWL. This sample was collected in early September from a small polygonal pond on Holocene non-yedoma, draining into the Lucky Lake stream. Isotopic values

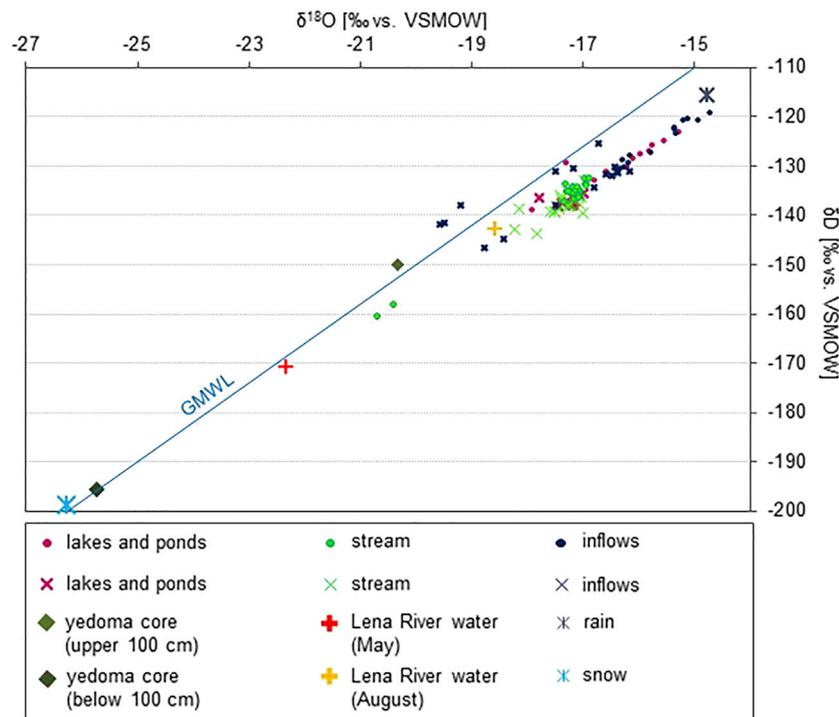


FIGURE 5 | δD and $\delta^{18}O$ values of lakes and ponds, Lucky Lake stream, and inflows in the Lucky Lake catchment by Holocene non-yedoma (dots) and Pleistocene yedoma (crosses). As references we added δD and $\delta^{18}O$ values of Lena River water samples from May and August 2014, rain and snow samples from Bykovsky Peninsula in the Lena River Delta (Meyer et al., 2002), a yedoma core collected north of the Lucky Lake catchment, and the GMWL.

of three samples of the yedoma upland inflows lay above the GMWL. We found the highest $\delta^{18}O$ and δD in samples of inflows and lakes on Holocene non-yedoma.

Discharge and DOC Flux

For the observation period from July 27, 2013 to August 26, 2013 we found decreasing discharge from 1,700 to 200 m³ d⁻¹ at weir 1, corresponding to decreasing DOC concentrations from 5 to 4.1 mg L⁻¹ at weir 1. For weir 2, the discharge decreased from 3,000–500 m³ d⁻¹, corresponding to decreasing DOC concentrations from 4.6 to 4.4 mg L⁻¹ at weir 2. Based on these values, we estimated a DOC flux of 92.5 kg at weir 1 and 220.5 kg at weir 2 for 29 days in 2013. With an estimated watershed area of 6.45 km², we calculated a DOC flux of 34.2 kg km⁻² for 29 days of discharge measurements and 1.2 kg km⁻² per day.

DISCUSSION

Differing DOC Concentrations in Inflows, Lakes, and Outflows on Holocene Non-yedoma and Pleistocene Yedoma

Yedoma deposits are especially prone to degradation processes (Strauss et al., 2013) such as erosion of thermokarst lake shores (Larsen et al., 2017), as for Lucky Lake and Oval Lake, and thermo-erosional gullies, as for yedoma thaw streams in our

study area. During these processes organic-rich materials from degrading yedoma deposits are thawed and transported into lakes and streams, resulting in higher median DOC concentration in surface waters on Pleistocene yedoma. Whereas findings from Siberia and Alaska show that yedoma lakes have significantly higher DOC concentrations compared to non-yedoma lakes (Sepulveda-Jauregui et al., 2015), the differences in lake DOC concentration at our study site between Pleistocene yedoma and Holocene non-yedoma are statistically not significant (Figure 3B). We found high DOC concentration of 15.6 mg L⁻¹ in the smallest pond in the catchment on Holocene non-yedoma, which is characterized by low water depth and a high amount of submerged vegetation, whereas Lucky Lake and the neighboring Oval Lake on Pleistocene yedoma are lakes of lowest DOC concentrations and largest lake surface area. This negative correlation of lake DOC concentration and lake surface area was already shown for lakes in western Siberia with discontinuous and sporadic permafrost (Shirokova et al., 2013). As possible causes, they suggest low water depth and the fast release of DOC from vegetation in small ponds. Furthermore, small ponds are known to have a high input of allochthonous DOC due to a higher ratio of lake surface area and lake volume. Here, short water residence time results in reduced lake processes, such as mineralization or photo-degradation, causing higher DOC concentrations (Shirokova et al., 2013; Manasypov et al., 2014, 2015). In larger lakes an increasing consumption of dissolved

organic matter by bacterio-plankton and photo-degradation takes place, resulting in decreasing DOC concentrations from small ponds to large thermokarst lakes. As a result of an increase in lake surface area due to thermokarst lake shore erosion, solar irradiance becomes increasingly important for stimulating photochemical processes such as photo-oxidation of DOC, causing a decrease in DOC concentrations in such larger lakes (Surdu et al., 2014; Williamson et al., 2014).

Coch et al. (2019) analyzed two catchments in the high and low Canadian Arctic, finding generally higher DOC concentrations in samples of standing waters compared to stream water samples. For our study site, we contrastingly found high DOC concentrations in samples of thaw streams from the yedoma uplands on Pleistocene yedoma, which are two to three times higher than in lakes of our study site. Additionally, DOC concentrations of Lucky Lake stream are partially higher than DOC concentrations of some of the lakes in our study site. This difference between their study and our results can be attributed to the presence of C-rich yedoma deposits on Pleistocene yedoma in our study site, whereas the study sites of Coch et al. (2019) are characterized by marine and glacial sediments underlain by bedrock deposits. A study in the Kolyma River basin also found the highest concentrations of highly labile DOC in small yedoma thaw streams compared to larger rivers and streams in the basin (Mann et al., 2015). Another study reports DOC concentrations from 155 to 196 mg L⁻¹ in actively eroding streams of a yedoma exposure on the Kolyma River bank (Vonk et al., 2013b), which is more than ten times higher than in thaw streams of our study site. In contrast, inflows on Holocene non-yedoma have DOC concentrations between 2.9 and 3.6 mg L⁻¹. By mixing with water from the yedoma upland these inflows might cause a dilution effect in the Lucky Lake stream, resulting in decreasing DOC concentrations along the flow path to the Olenyokskaya Channel.

DOC Flux From Pleistocene Yedoma to the Olenyokskaya Channel

When discussing fluxes in permafrost affected landscapes we need to clarify the role of groundwater. Walvoord and Kurylyk (2016) summarized supra-permafrost aquifers including unfrozen taliks below lakes and streams and the active layer on top of the permafrost table, intra-permafrost groundwater, and sub-permafrost aquifers as groundwater zones in discontinuous permafrost. In our study area in the continuous permafrost zone, groundwater exchange predominantly occurs in the supra-permafrost aquifer (Helbig et al., 2013). The amount of DOC released from permafrost to Arctic freshwaters changes in response to climate change (Wickland et al., 2018). These changes differ e.g. by permafrost composition and catchment characteristics, such as permafrost distribution. In regions of warmer discontinuous permafrost with connected intra- and supra-permafrost ground water aquifers, the DOC was shown to decline over the past 40 years (Striegl et al., 2005). In contrast, in regions of cold continuous permafrost with limited ground water flow, DOC mobilization increases (Wickland et al., 2018). Vonk et al. (2019) defined permafrost continuity, ice content, soil

composition and morphology, as well as topography, as critical factors for C mobilization and transport in waters in thaw affected permafrost landscapes. In yedoma regions, DOC mobilization and transport is presumably accelerated by permafrost thaw and associated processes (Vonk et al., 2013a; Drake et al., 2015; Mann et al., 2015; Spencer et al., 2015). Moreover, it is important to mention climate change driven influencing processes such as abrupt thaw of permafrost, drainage of thaw lake basins, and active layer deepening (Opfergelt, 2020). Changes and especially increasing of active layer depth reveal subsurface fluxes (Helbig et al., 2013), which may affect the transport and flux of DOC. The stream from Lucky Lake to its mouth at the Lena River branch is dominated by rapid fluvial processes, transporting sediments and dissolved material. For our small lake catchment, we estimate a DOC flux of 1.2 kg km⁻² per day. Lewis et al. (2012) found comparable DOC flux of 350 kg km⁻² per year and a downscaled value of approximately 1 kg km⁻² per day for a small watershed of 8 km² in the high Canadian Arctic. For the entire Lena River a DOC flux of 6.79 Tg C per year was calculated (Juhls et al., 2020). This corresponds to a DOC flux of approximately 2,602 kg km⁻² and to 7 kg km⁻² per day, which is almost 6 times higher than DOC flux in our small catchment. With an estimation of approximately 3 kg km⁻² per day the Mackenzie River has a lower summer DOC flux (Coch et al., 2018). This lower flux might be caused by the absence of ice-rich yedoma deposits, the presence of large lakes, which might decrease DOC fluxes, and extensive wetlands in the Mackenzie Delta, as well as the glacial history (Raymond et al., 2007; Burn and Kokelj, 2009). Moreover, in contrast to previously mentioned catchments and watersheds the Mackenzie River catchment covers large areas with only sporadic permafrost distribution and also without permafrost deposits. On the other hand, peatlands, which occur in the Mackenzie Delta, are known to increase DOC mobilization (Frey and Smith, 2005; Olefeldt and Roulet, 2012).

Our estimates do not account for the impacts of rain or storm events, and DOC flux at our study site was calculated for only 29 days of a year. Furthermore, we do not account for seasonal variability in discharge, especially during spring snowmelt, since we are focusing on the summer season. In a recent study of Juhls et al. (2020) it was possible to calculate the seasonal variability of DOC flux for the Lena River. They found that approximately half of the annual DOC flux of 6.79 Tg C is transported in summer and 41% in spring, when spring snowmelt, the biggest hydrological event in this region, occurs. Furthermore, Ahmed et al. (2020) predicted a shift of seasonal discharge towards earlier spring floods resulting in decreasing summer discharge, which may lead to a decrease in summer DOC flux. Our estimations suggest that if we merge all catchments in the Lena River Delta or even in the Lena River watershed that are similar to the small catchment on Kurungnakh Island we present here, comparatively less DOC is released to the Lena River than further upstream in the catchment, and that this DOC is relatively young. Coch et al. (2018) compared their calculations of a 17 days summer DOC flux (mean flux: 82.7 ± 30.7 kg km⁻²) of a small watershed on Herschel Island in the Canadian Arctic with different Arctic locations and found higher DOC fluxes in their low Arctic site

compared to high Arctic locations. They also highlight the contribution of small watersheds and consider the snowmelt and related discharge to be a major influencing process of DOC flux. Based on expected changes in discharge due to climate change (Bintanja and Andry, 2017), such as shifting seasonal discharge, we assume changing DOC fluxes from small Arctic catchments as well as for major Arctic river catchments, and especially increasing DOC fluxes from yedoma catchments (Tank et al., 2020) like our Lucky Lake stream catchment in the coming decades.

DOC Sources in the Lucky Lake Catchment

Generally, lake DOC can be produced in the lake itself (autochthonous) and in the lake catchment (allochthonous), and previous studies identified several parameters influencing lake DOC concentration, such as lake perimeter and lake elevation (Xenopoulos et al., 2003), lake area (Tranvik et al., 2009; Shirokova et al., 2013; Zabelina et al., 2021), and hydrological connectivity of a lake (Bogard et al., 2019; Johnston et al., 2020). Lucky Lake is a thermokarst lake where lake shore erosion is active and causes transport of old organic material from the surrounding late Pleistocene permafrost deposits into the lake. For streams, erosion of stream banks leads to a higher amount of ^{14}C depleted DOC (Mann et al., 2015). Pleistocene yedoma is rich in fossil OC. But also Holocene deposits are rich in OC (Schirrmeister et al., 2011) and a Holocene surface layer overlies yedoma deposits on the 3rd delta unit. However, yedoma C is more bioavailable (Vonk et al., 2013a; Strauss et al., 2015) and decomposable than C in other thawed mineral soils (Walter Anthony et al., 2014). We found the lowest $\Delta^{14}\text{C}$ values in samples of Lucky Lake. Here, the DOC in our water samples is approximately 3,600 years old. $\Delta^{14}\text{C}$ values in an aquatic system such as Lucky Lake may result from fresh aquatic production in the water body, from leached material from the active layer, and/or from leached organic matter from eroded yedoma deposits.

Our results show DOC concentrations of 9–12.7 mg L⁻¹ in inflows in the north-east of Lucky Lake, where yedoma upland is drained by thaw streams. After thaw stream water reaches Lucky Lake, DOC concentrations decrease to less than half, suggesting that yedoma-derived DOC likely is rapidly utilized or that low Lucky Lake DOC concentration causes a dilution. Regarding C input from the two delta units in our study area, we assume that C in the Lucky Lake also originates from both eroding Holocene materials and from the older yedoma deposits. With a DOC age of approximately 3,600 ^{14}C y BP we found the oldest DOC of the entire dataset in samples from the Lucky Lake with $\Delta^{14}\text{C}$ values between -370 and -373 ‰.

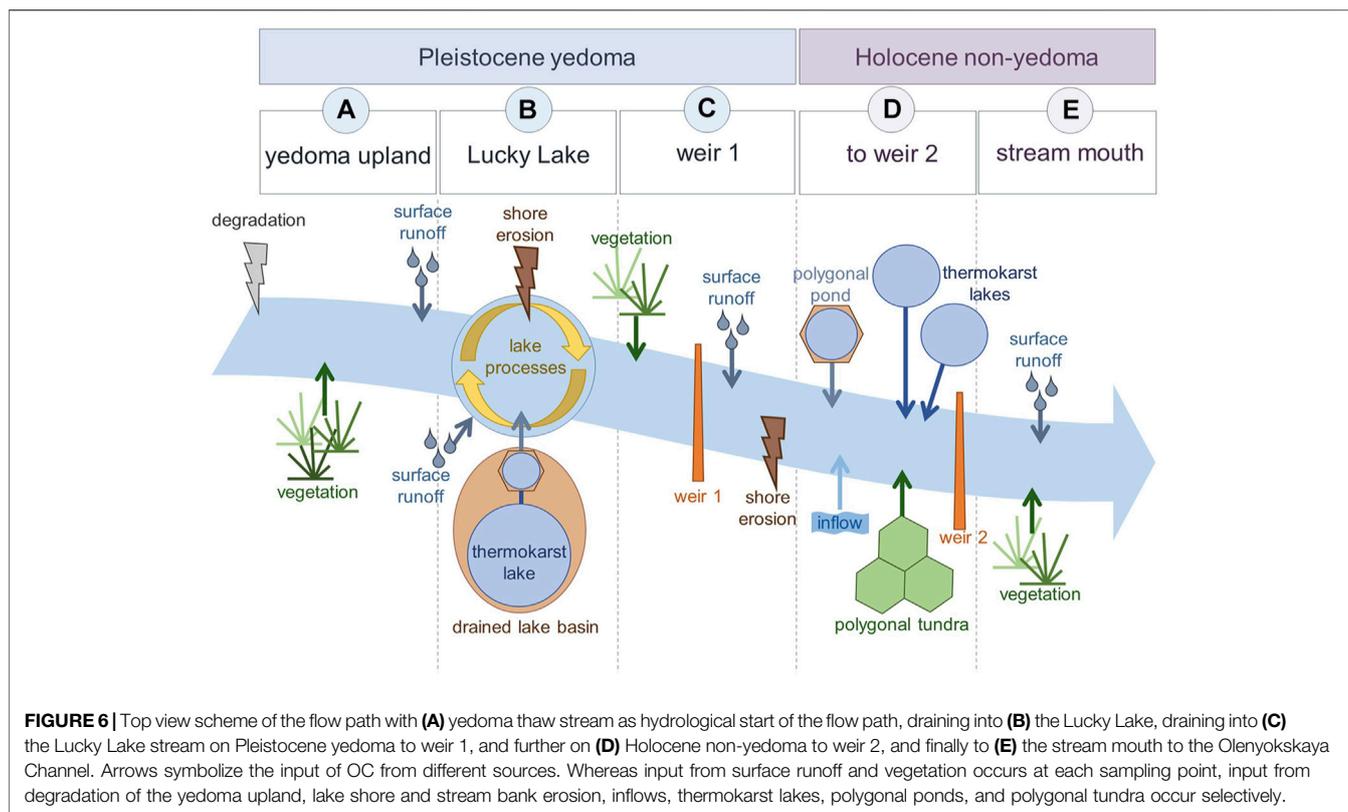
DOC in the Flow Path From Yedoma Uplands to the Olenyokskaya Channel

According to **Figure 6**, yedoma thaw streams, draining Pleistocene yedoma uplands, represent the hydrological start of the flow path in our study area. These inflows (**Figure 6A**) show DOC concentrations more than twice as high as the dataset's median. The inflows drain thaw-affected yedoma

uplands and mobilize OC, as well as active layer and tundra vegetation. Unfortunately, we did not perform radiocarbon dating for samples of these inflows in our study site but for small yedoma thaw streams in the Kolyma River basin, Mann et al. (2015) found $\Delta^{14}\text{C}$ signatures of 883 ± 41 ‰, which corresponds to an age of more than 20,000 years. In contrast, DOC in large rivers and streams of the Kolyma River basin is modern (Neff et al., 2006; Vonk et al., 2013b; Mann et al., 2015; Spencer et al., 2015). The water isotopic composition ($\delta^{18}\text{O}$ and δD) of three yedoma inflow samples plot above the GMWL (**Figure 5**) caused by disequilibrium processes like thawing and freezing of the active layer in spring and fall.

After water from the yedoma thaw streams reached Lucky Lake (**Figure 6B**), we observed an abrupt decrease of DOC concentrations. The question of what happens to DOC in the lake arises. In contrast to yedoma thaw streams, which are small trickles with low discharge, Lucky Lake has a greater water volume of low DOC concentration and may dilute the inflowing yedoma thaw stream water of higher DOC concentrations. Moreover, the neighboring thermokarst lake Oval Lake, situated in a drained lake basin, drains into Lucky Lake via a small stream and a drained pond. Whereas DOC concentrations of Oval Lake are similar to those of Lucky Lake, $\Delta^{14}\text{C}$ signatures of Oval Lake (between -322 and -300 ‰) are much higher comparing to Lucky Lake $\Delta^{14}\text{C}$ signatures (between -373 and -370 ‰). These samples contain the oldest DOC in our entire dataset and might be a result of eroding yedoma lake shore and Holocene deposits. There are a number of lake processes that may lead to decreasing lake DOC concentration. Photo-oxidation or microbial activity lead to mineralization of DOC to CO_2 and CH_4 , which can be emitted to the atmosphere (Tranvik et al., 2009; Vonk et al., 2013a, b). Studies of Mann et al. (2015) and Spencer et al. (2015) found that the lability of old yedoma DOC causes rapid mineralization and further lower DOC concentrations when yedoma thaw stream waters reach main stream water. Additionally, the process of flocculation may lead to deposition of DOC in lake sediments and hence to decreasing DOC concentration in the lake water (Tranvik et al., 2009). If we assume a yedoma age from the Pleistocene of approximately 30,000 years, DOC from this material should have a $\Delta^{14}\text{C}$ value of -980 ‰. Using isotope mass balance calculation and assuming that one part of this old DOC mixes with two parts of young DOC from surface leaching (Holocene, $\Delta^{14}\text{C} = -80$ ‰) a theoretical lake DOC $\Delta^{14}\text{C}$ value of -373 ‰ is estimated. This suggests that dilution, rather than preferential utilization of DOC supplied to the lake by inflows, caused the decrease in DOC concentration from yedoma upland thaw streams to the Lucky Lake. We also assume a smaller amount of DOC released along the lake shore and from the talik than DOC that is mobilized at the surface and transported via inflows.

Directly after the Lucky Lake outflow, where Lucky Lake drained into the Lucky Lake valley and formed the Lucky Lake stream on Pleistocene yedoma (**Figure 6C**), we observed an increase in $\Delta^{14}\text{C}$ signature to -356 ‰. Samples from the Lucky Lake stream on Pleistocene yedoma show similar values of $\delta^{18}\text{O}$ and δD compared to those of Lucky Lake, and let us assume that the flowing water at this point is mostly influenced by



Lucky Lake water. We found slightly decreasing DOC concentrations downstream to weir 1, where the concentration of DOC is mainly influenced by hydrological connectivity and erosion along river stream banks (Raymond et al., 2007; Mann et al., 2015). The main DOC sources here are Lucky Lake waters, as well as fresh DOC leached from vegetation, which also leads to younger DOC ages.

Where Lucky Lake stream reaches Holocene non-yedoma (Figure 6D), the setting changes rapidly to wetter polygonal tundra vegetation and the eroding Lucky Lake valley with its incised stream. Two small thermokarst lakes in the north of Lucky Lake stream and a stream from a neighboring side valley have low DOC concentrations between 3.1 and 3.6 mg L⁻¹ and are connected with the Lucky Lake stream. Up to weir 2 we observed slightly decreasing DOC concentrations and similar $\delta^{18}\text{O}$ and δD values compared to $\delta^{18}\text{O}$ and δD values of thermokarst lakes of Pleistocene yedoma as well as Lucky Lake stream on Pleistocene yedoma.

After weir 2, the DOC concentration further decreases towards the mouth of Lucky Lake stream at the Olenyokskaya Channel (Figure 6E). In contrast, we observed increasing $\delta^{18}\text{O}$ values attributed to contributions from melted ice frozen during conditions of higher air temperatures during the Holocene. Two Lucky Lake stream samples close to the mouth at the Olenyokskaya Channel show especially low values for $\delta^{18}\text{O}$ and δD (Figure 5). These samples were collected in mid-June. Here, expedition members observed high flow on Olenyokskaya Channel during spring flood, causing tailback from river channel

water into the Lucky Lake stream. At the mouth of Lucky Lake stream to the Olenyokskaya Channel we found an increased $\Delta^{14}\text{C}$ signature of -301 ‰, which corresponds to a DOC age of 2,783 ¹⁴C y BP. For comparison, major streams and the main stream of the Kolyma River basin have $\Delta^{14}\text{C}$ signatures of 22–51 ‰ (Mann et al., 2015), and the averaged $\Delta^{14}\text{C}$ of DOC in the Lena River was determined to be 78 ‰ (Raymond et al., 2007). The younger DOC ages combined with decreasing DOC concentrations after the water of Lucky Lake exit the lake and drains into Lucky Lake stream and further to the mouth at the Olenyokskaya Channel could be caused by several processes: 1) Supply to the Lucky Lake stream fresh and young DOC leached from vegetation (e.g. Schuur et al., 2008; Coch et al., 2020). The downstream changes in $\Delta^{14}\text{C}$ along the flow path might thus result from mixing of water of different DOC concentrations and radiocarbon levels when the stream drains different geomorphological units. A mixing with younger DOC contributed from water of Holocene non-yedoma and deposits likely causes an overall dilution of older DOC signatures with younger DOC with increasing distance from the old DOC sources. 2) Old DOC might be re-mineralized. Old yedoma DOC has been shown to decompose and outgas faster than younger DOC because of higher bioavailability of the materials leached from yedoma permafrost (Vonk et al., 2013a; Drake et al., 2015). Drake et al. (2015) highlighted an especially rapid DOC turnover to CO₂ due to yedoma permafrost thaw and directly initiated DOC mineralization. Spencer et al. (2015) analyzed water samples of the Kolyma River basin and found the

youngest C signature downstream at the river mouth. They suggest that a faster utilization of ancient C and thus outgassing is a possible cause for this observed downstream gradient. Our results show increasing $\Delta^{14}\text{C}$ from Pleistocene yedoma to the Holocene non-yedoma, which would be consistent with both scenarios, preferential re-mineralization of old DOC and a dilution by inflows. Because DOC radiocarbon values are higher (younger) at the mouth at the Olenyokskaya Channel, photochemical or microbial processes from Lucky Lake to the Olenyokskaya Channel might be rapidly degrading old DOC. Our observation of progressively younger signature of DOC downstream is consistent with trends also described for major Arctic rivers (Benner et al., 2004; Raymond et al., 2007; Striegl et al., 2007; Aiken et al., 2014). Changes in the isotopic composition with increasing values for $\delta^{18}\text{O}$ and δD from Pleistocene yedoma suggest the mixing of different waters.

In order to assess whether preferential re-mineralization of old DOC or mixing of waters with different DOC concentrations and higher $\Delta^{14}\text{C}$ values is the more likely process, we can again use isotope mass balance calculations. As radiocarbon measurements were only carried out for samples from a limited number of sites and no streams from Holocene non-yedoma were sampled for this purpose, we need to estimate which radiocarbon signature DOC in those streams would need to carry in order to explain the observed trend through mixing. We used as input parameters the DOC concentrations and the discharge measured at both weirs (minimum and maximum values to determine the possible range), and the $\Delta^{14}\text{C}$ values measured for DOC at the Lucky Lake outflow, and at the outflow of the small drainage system to the Olenyokskaya Channel. Based on this mass balance calculation, we estimate the DOC $\Delta^{14}\text{C}$ values of inflows between weir 1 and 2 to range between -292 and -287 ‰, respectively. Note that for this estimate, DOC concentrations and radiocarbon ages from different sampling years and months were used. We consider this an acceptable assumption, since we found no major inter-annual changes in DOC concentration and no major ranges in the sampled years. The $\Delta^{14}\text{C}$ signature of -292 to -287 ‰ (corresponding to ^{14}C ages of approximately 2,800 ^{14}C years) estimated for Holocene non-yedoma tributaries to the drainage system appears a plausible value considering the geological setting. While not excluded, preferential re-mineralization of old DOC likewise resulting in a progressive increase in $\Delta^{14}\text{C}$ and decrease in DOC concentration along the flow path is at least not the only plausible scenario for our study site.

CONCLUSION

In our study we analyze the influence of lakes and ponds, which are interposed in a stream catchment, on DOC concentrations and export; we discuss the origin of DOC in a small lake catchment on Kurungnakh Island in the central Lena River Delta, and how it may transform on its flow path to the river outlet. We show decreasing DOC concentrations from the beginning of the flow path at the yedoma upland thaw streams to the biggest lake in our study site, the Lucky Lake.

Additionally, we found decreasing DOC concentration from Lucky Lake via the Lucky Lake stream to the Olenyokskaya Channel. Further, our data indicate that old yedoma and Holocene C might be mobilized into thermokarst lake systems. We found progressively younger DOC signatures in the downstream segments of the catchment. We discussed degrading yedoma and non-yedoma permafrost, active layer, and vegetation to be the major DOC sources in the catchment. Changes and especially the decrease of DOC concentration on the flow path might be ascribed to 1) dilution/mixing with water of lower concentrations and/or younger DOC; 2) lake processes such as photo-oxidation, microbial activity, and flocculation; and 3) a possible outgassing of DOC on the flow path downstream; or a combination of these processes. Finally, the input of DOC from our catchment to the Lena River is dominated by younger DOC compared to Lucky Lake DOC age. In our study, we discuss how OC from yedoma deposits enters aquatic systems as DOC and how DOC is transformed during transport in fluvial networks. Under future climate, degrading yedoma permafrost will cause changes in groundwater and subsurface flow in Arctic watersheds and may increase DOC export. We demonstrate that lakes and ponds may act as DOC filters by diluting incoming waters of higher DOC concentrations and modifying DOC to CO_2 and CH_4 . Small thermokarst lake catchments may therefore critically determine C emissions of yedoma landscapes in a rapidly warming Arctic.

DATA AVAILABILITY STATEMENT

The ^{14}C , discharge, DOC and stable water isotopes data presented in this study are deposited in the PANGAEA repository (Hammes and Mollenhauer, 2020; Stolpmann et al., 2021b; Stolpmann et al., 2021c).

AUTHOR CONTRIBUTIONS

LS, GM, and AM developed the study design. JB designed the hydrological set up for lake level and discharge measurements in the field. Field work was conducted by AM in 2013, 2014. LS and JH ran laboratory analyses for DOC concentration and radiocarbon ages. LS performed statistical analyses, interpreted the data with input from JH, GM, AM, JB, PO, and GG and prepared the paper with contributions from all co-authors.

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