

Response to the editors comments
Dear Dr. Bussmann

This is an interesting study, particularly considering the difficulty for sampling

Both reviewers provide critical comments which cannot be corrected by minor changes. Substantial changes appear to be needed to address adequately the questions raised by both reviewers.

In addition, the following are for the authors' reference by a quick editorial review again
(1) Please justify the rationale why two sampling could be defined as "seasonal dynamics"
ok, the title has been changed to "Methane dynamics in three different Siberian water bodies under winter and summer conditions"

(2) Please describe the most rapid climate warming on Earth in a quantitative manner in the introduction section

The increase of atmospheric methane has been specified with more recent data.
"Worldwide, the mixing ratio of methane has been increasing rapidly since 2000, from 2.1 ppm/y for the time span 2000 – 2009, to 6.6 ppb/ y for the time span 2008 – 2007 and to 6.1 ppb/y in 2017 (Saunois et al., 2020). Understanding and quantifying the global methane budget is important for assessing realistic pathways to mitigate climate change. For the 2008 – 2017 decade, global methane emissions are estimated by a top-down approach to be 576 TgCH₄ /y (range 550–594, corresponding to the minimum and maximum estimates of the model ensemble) (Saunois et al., 2020)"

(3) Line 36. Is it positive influence when compared to temperate environments

The comparison to temperate environments has been removed from the abstract. In the text however, it can be seen that the Q10 is for polar environments was higher than for temperate environments.

(4) In the abstract, please conclude with a more general finding if possible. Or maybe the authors could add some important implications of this study

The following sentence has now been added to the end of the abstract
..."In a changing, warming Arctic, a shortened time of ice coverage on the water bodies is predicted. In respect to our study this would imply a shortened time for methane to accumulate below the ice and a shorter time for the less efficient winter-MOX. Especially for lakes, an extended time of ice-free conditions could reduce the methane flux from the Arctic water bodies.

(5) L53. Pls specify the current concentration of methane in the atmosphere.

The atmospheric methane concentration as measured in the polar region of Svalbard is given with 1939 ppb for 2017 (Platt et al 2018) and with 1932 ppb for Tiksi (Hydrometeorological Observatory of Tiksi, Russia (TIK)).

(6) L55-58. Why a better quantification of the emissions of different contributors is needed. What is the current estimate, and how uncertain it is, and the range ?

We added the following sentence:

Understanding and quantifying the global methane budget is important for assessing realistic pathways to mitigate climate change. For the 2008–2017 decade, global methane emissions are estimated by a top-down approach to be 576 TgCH₄ /y (range 550–594, corresponding to the minimum and maximum estimates of the model ensemble) [Saunio, 2020 #3085].

And

Especially in northern latitudes natural wetlands contribute with 59% to the northern methane emissions (Saunio et al., 2020)

I would like to have your attention that simple changes will not be able to correct all the comments raised by the reviewers

Thank you for your submission to BG and once again I apologize for some delay of managing the ms.

Kind regards

Interactive comment on “Seasonal methane dynamics in three different Siberian water bodies”

by Ingeborg Bussmann et al.

Anonymous Referee #1

Received and published: 9 July 2020

The manuscript "Seasonal methane dynamics in three different Siberian water bodies" by Bussmann et al. presents measurements of methane concentrations and oxidation rates (MOX) from different samplings in an Arctic river delta, in the adjacent coastal waters and in a nearby small Arctic lake from sampling campaigns carried out during ice-free summer conditions and under ice cover in late winter. The presented data close knowledge gaps on winter time methane dynamics in Arctic waters. The authors find that CH₄ accumulation under ice is governed by the exchange of waters under the ice, with insignificant accumulation in the river and strong accumulation in the stagnant lake waters. Additional measurements from ice cores did not show large CH₄ accumulation and no CH₄ gradients, and confirmed that the ice cover is an effective barrier for the CH₄ flux to the atmosphere.

The manuscript is generally well written, and the presentation of the CH₄ concentration and ice core data is clear and comprehensive.

I have some issues with the part on the MOX rates, however, which need to be resolved by the authors: in addition to the methane concentrations from the different water bodies, MOX rates were determined in winter time in the Lena river. The authors use these MOX rates to determine the potential of MOX for reducing the CH₄ accumulation under ice. Based on their assumptions, they conclude that the measured low MOX would lead to a rather small reduction of the CH₄ inventory, and strong CH₄ accumulation under ice is likely. However, this estimate is based on a number of assumptions the authors make, and I am missing a detailed chain of arguments that justifies these assumptions:

- the authors transfer the fractional MOX rate determined from the Lena river sampling to the coastal and lake waters (lines 318-320), but they miss a convincing argumentation if this is justified. In lines 460 to 470, they discuss the influence of different environmental factors such as oxygen and phosphate concentration and temperature (lines 460-470), but these factors are not taken into account or even discussed when the MOX rates from the coastal and lake waters are calculated. Instead, the authors present the MOX rates from these two water bodies as if these were independently determined (lines 320-325, Fig.5), and not calculated from the CH₄ concentrations.

Yes, the referee is right here, we determined k' for river water and for the ice cores from the Lake and Tiksi Bay, but k' was not determined for each single sample. This is a major drawback, but the best

we could do at these difficult locations. We also removed the term “in situ MOX” and name it only MOX. This fact is stressed more now in the abstract, the result section, as well as in the discussion: “The fractional turnover rate k' was determined in ice cores from the lake and Tiksi Bay, and in river water. Within these locations k' was evenly distributed. However, k' may vary between different environments (river, lake and brackish water) as well as between ice cores and underlying water. The fractional turnover rate is influenced by temperature, methane and oxygen concentrations [Steinle, 2017 #2755]. Thus, in our calculations an error is embedded. However, as temperature was low at all locations, and the methanotrophic population of the ice cores is similar to the ones in the water below, we assume that the application of one k' to all samples is a good assumption.”

- the authors conclude that the accumulation of CH₄ under ice will lead to a pulse release of CH₄ when the ice melts, and the low MOX in winter compared to summer conditions induce larger overall emissions from winter than from summer conditions. I think this argumentation is a bit too superficial
We agree that the line of discussion was too simplified. We added now a more detailed discussion on the sources and sinks of methane in an aquatic system. As can be seen in the last part of the new discussion

– summer and winter conditions not only differ in the effectiveness of CH₄ oxidation, but also in the fact that during summer, gas exchange is an additional sink for CH₄, and that CH₄ emissions are determined by the effectiveness of CH₄ oxidation vs. gas exchange. To fully assess the potential of CH₄ emissions from pulse release during ice-off or from continuous emissions under ice-free conditions, all sink terms would need to be carefully taken into account.

We agree that the line of discussion was too simplified. We added now a more detailed discussion on the sources and sinks of methane in an aquatic system. As can be seen in the last part of the new discussion.

Specific comments:

Line 20: "Arctic regions and their water bodies are affected..."

Changed accordingly

Line 134: "the Bykovskaya Bay"

No, Buor-Kaya Bay is correct

Line 467: "But temperature is also: : :": remove "but"

Changed accordingly

Lines 504-505: A 60% reduction of the methane inventory by MOX does not seem like an insignificant reduction of the CH₄ inventory to me.

This part has been removed and rewritten.

*Lines 508-509: "we assume, that after ice-on, both parameters increase/decrease in a linear way"
Could you explain this assumption? Why would this be justified?*

This part has been removed and rewritten.

Lines 546-550: I think this statement is somewhat speculative- I doubt that the increased MOX rates in summer than in winter is the only factor that determines CH₄ emissions from the Arctic water bodies. The authors should at least mention which additional factors could influence CH₄ emissions.

This part has been removed and rewritten.

Line 554: The data should be submitted to PANGAEA and the data availability statement updated.

The data are submitted in Pangaea and details are given in the revised version.

Anonymous Referee #2

Received and published: 7 September 2020

This is a very interesting study and it is also practically difficult to conduct field research in waterbodies of Arctic regions. It is thus a timely contribution of methane cycles.

The manuscript is well organized, and the writing appears to be somehow redundant.

The major concerns are the following.

*(1) Title. Is the term seasonal dynamic appropriate? There are only two sampling period for some rivers. A better title might be formulated such methane dynamics under contrasting ***?*

As the editor also suggest to remove the “seasonal” , we changed the title to “Methane dynamics in three different Siberian water bodies under winter and summer conditions”

(2) Conclusion. The rationale behind the higher methane concentrations in winter than that in summer is not very clear for Tiksi bay and Lake Golzovoye. Please make a brief and focused discussion about the possible mechanisms.

A new part of the discussion has been added, to explain in more detail the different sinks and sources of methane in a water body. (in the last part of the discussion)

(3) Methane production potential. If these data are not available, the authors may discuss methanogenesis a little bit more. Or methane simply stored in waterbodies due to physiochemical mechanisms?

Colleagues have determined the methane production rate in Lake G. and a marine setting, with 0.5 - 0.2 nmolg/g/d at surface sediment. However, active AOM and SRR reduce methane concentrations at the sediment surface to about 2 mM in freshwater sediment and to < 0.1 mM in marine sediments (unpublished data from S. Liebner and C. Knoblauch). Added in Line 445ff

(4) Oxygen concentrations. Please provide these data as much as possible if available.

Unfortunately, there are no data on the oxygen concentrations at any of the locations.

Minor concerns

(1) L20. How to define “the most rapid climate warming on Earth”?

Changed to “affected by a rapidly warming climate”

(2) L22. Maybe the authors can briefly introduce the proportion of these poorly unexplored water bodies.

More details on the different water bodies are given in the introduction, . L 87 ff and L 111 ff. To elaborate this in the abstract would be too long

(2) L35. It is somehow abrupt to compare with temperate environments. This is more appropriate in the review paper

The relation to temperate environments has been removed.

(4) L45. Please give concluding remarks as a summary of the key findings.

We have now added our concluding remarks to the abstract.

“The winter situation seemed to favor a methane accumulation under ice, especially in the lake with a stagnant water body. While on the other hand, in the Lena River with its flowing water no methane accumulation under ice was observed. In a changing, warming Arctic, a shorter ice cover period is predicted. In respect to our study this would imply a shortened time for methane to accumulate below the ice and a shorter time for the less efficient winter-MOX. Especially for lakes, an extended time of ice-free conditions could reduce the methane flux from the Arctic water bodies.”

(5) L55. Pls describe the range of variability

This variability is mentioned in the abstract of the reference, but not further elucidated in the text. Therefore we changed our statement to “...and the sea-air flux of methane is mainly affected by increasing water temperatures (Wahlström et al., 2016)“

(6) L65-67. This sentence seems irrelevant to the previous one. The ebullition mode and transportation from Arctic rivers to the shelf seems to be different.

In the sentences above we describe the role of lakes for the methane flux. In the lines 111 ff we wanted to elucidate the role of rivers as methane source to the atmosphere, not the shelf.....

(7) L108. Pls write the conclusions in the abstract in line with the hypothesis.

We have now added our concluding remarks to the abstract.

(8) L111. Why not measure the potential of methanogenesis, and how to integrate these potentials in situ sink with the budget estimate of methane emission?

We were not able to measure methanogenesis in the field, however there are unpublished data on methane production and anaerobic oxidation in the sediment of Lake Golzovoye. We mention now these unpublished data in L 445ff.

(9) L125. The freeze-up and ice-off days can be specified for each waterbody

We have now added additional information, for lakes in L 105ff and L151, and for Tiksi Bay in L 158.

(10) L168. How low it is below the ice?

Just at the interface between ice and water, changed to “In winter, water samples at the ice – water interface were taken....”

(11) L174. Is the sampling procedure the same for different rivers?

No, the river water has been processed in the same way.

(12) L195. Please describe the procedures for methane concentration measurement. For example, is there any vigorous shaking?

Yes, samples were shaken for 2 min, to assure equilibrium between water and gas phase. This has been added to the Result section (3.4 Methane analysis).

(13) L305. Figure 3 and Figure 4 can be merged.

Ok, the figures have been merged

(14) L340. Figure 5 and Figure 6 can be merged

No, Figure 5 shows the methane oxidation rate at the different locations, while figure 6 shows the influence of temperature on the methane oxidation rate.

Methane dynamics in three different Siberian water bodies under winter and summer conditions

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Abstract

Arctic regions and their water bodies are affected by a rapidly warming climate. Arctic lakes and small ponds are known to act as an important source of atmospheric methane.

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However, not much is known about other types of water bodies in permafrost regions, which include major rivers and coastal bays as a transition type between freshwater and marine

environments. We monitored dissolved methane concentrations in three different water bodies (Lena River, Tiksi Bay and Lake Golzovoye, Siberia, Russia) over a period of two years.

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Sampling was carried out under ice cover (April) and in open water (July / August). The

methane oxidation (MOX) rate in water and melted ice samples from the late winter of 2017

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was determined with radiotracer method and fractional turnover rates (k') from river water and melted ice cores.

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In the Lena River winter methane concentrations were a quarter of the summer concentrations (8 vs 31 nmol L⁻¹) and mean winter MOX rate was low (0.023 nmol L⁻¹ d⁻¹). In contrast, Tiksi

45 Bay winter methane concentrations were 10-times higher than in summer (103 vs 13 nmol L⁻¹). Winter MOX rates showed a median of 0.305 nmol L⁻¹ d⁻¹. In Lake Golzovoye, median methane concentrations in winter were 40-times higher than in summer (1957 vs 49 nmol L⁻¹). However, MOX was much higher in the lake (2.95 nmol L⁻¹ d⁻¹) than in either the river or bay. The temperature had a strong influence on the MOX, (Q10 = 2.72 ± 0.69). In the ice cores a median methane concentration of 9 nM was observed, with no gradient between the ice surface and the bottom layer at the ice-water-interface. MOX in the (melted) ice cores was mostly below the detection limit. Comparing methane concentrations in the ice with the underlaying water column revealed 100 - 1000-times higher methane concentration in the water column.

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55 The winter situation seemed to favor a methane accumulation under ice, especially in the lake with a stagnant water body. While on the other hand, in the Lena River with its flowing water no methane accumulation under ice was observed. In a changing, warming Arctic, a shorter ice cover period is predicted. In respect to our study this would imply a shortened time for methane to accumulate below the ice and a shorter time for the less efficient winter-MOX. Especially for lakes, an extended time of ice-free conditions could reduce the methane flux from the Arctic water bodies.

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1 Introduction

Worldwide, the mixing ratio of methane has been increasing rapidly since 2000, from 2.1 ppm/y for the time span 2000 – 2009, to 6.6 ppb/y for the time span 2008 – 2007 and to 6.1 ppb/y in 2017 (Saunois et al., 2020). Understanding and quantifying the global methane budget is important for assessing realistic pathways to mitigate climate change. For the 2008 – 2017 decade, global methane emissions are estimated by a top-down approach to be 576 TgCH₄/y (range 550–594, corresponding to the minimum and maximum estimates of the model ensemble) (Saunois et al., 2020). The reasons for the observed increases in atmospheric methane are unclear. A probable explanation is increased methane emissions from wetlands, both in the tropics (Nisbet et al., 2016) as well as in the Arctic (Fisher et al., 2011), and from other Arctic water bodies (Walter Anthony et al., 2016; Kohnert et al., 2018) or geological methane emissions (Kohnert et al., 2017). Especially in northern latitudes natural wetlands contribute with 59% to the northern methane emissions (Saunois et al., 2020). In the Arctic, mean atmospheric methane mixing ratio increased by 6 ppb per year from 2001 to 2017 ; resulting in an atmospheric concentration of 1939 ppb in 2017 at Svalbard (Platt et al., 2018)

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85 and with a median of 1932 ppb in 2017 for Tiksi (Hydrometeorological Observatory of Tiksi, Russia). Especially the Laptev Sea, in eastern Siberia, is generally a source of methane to the atmosphere, and the sea-air flux of methane is mainly affected by increasing water temperatures (Wählström et al., 2016). Also, Saunois and co-authors estimated increased methane emission for freshwater systems and wetlands, but a better quantification of the emissions of different contributor (streams, rivers, lakes and ponds) is needed (Saunois et al., 90 2016).

Lakes are important sources of atmospheric methane on a regional to global scale (Bastviken et al., 2004; Cole et al., 2007), and their contribution is predicted to increase in response to climate change and rapidly warming waters (O'Reilly et al., 2015; Tan and Zhuang, 2015; Wik et al., 2018). Most of the methane produced in lake sediments enters the 95 atmosphere via ebullition (Bastviken et al., 2004; Walter et al., 2007), a temperature-sensitive transport mode with high spatial and temporal heterogeneity (DeSontro et al., 2015). The role of Arctic rivers as a methane source to the shelf seas is poorly described. Some studies present rivers as strong methane sources (Morozumi et al., 2019), while other studies revealed a complex pattern of riverine methane input (Bussmann et al., 2017).

100 One major drawback from most of these studies is that sampling was conducted in the ice-free season, although most of the year Arctic water bodies are ice covered. Thus, the seasonal variation could not be captured within those studies. The ice cover on lakes decouples the water body from the atmosphere and the circulation changes from wind driven to thermohaline. After ice formation a stable winter stratification is set up. As there is no more 105 external oxygen supply, enhanced anaerobic degradation leads to accumulation of methane, H₂S and NH₃ (Leppäranta, 2015). In several lakes in Alaska and Canada, dissolved methane was highest under the ice-cover, indicating that the spring ice off period is a large source of atmospheric methane (Townsend-Small et al., 2017; Cunada et al., 2018; Serikova et al., 2019). In lakes at > 65°N the ice duration is 9 months, typically from around mid-September 110 to mid-June (Cortés and MacIntyre, 2020). However, the hydrography of a lake is also an important factor to consider since stratification of the water column counteracts an intense gas exchange. Thus, examples are known for lakes with incomplete spring mixing and consequently a maximum gas exchange in autumn during complete mixing (Deshpande et al., 2015).

115 Rivers of permafrost regions are characterized by an ice season of >100 days duration between autumn freeze-up and spring ice-off; for the Lena River it is >160 days (Shiklomanov and Lammers, 2014). Ice effects and the demobilization of liquid water result

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120 in very low discharge during winter freeze-up and runoff is lowest during late winter
(Lininger and Wohl, 2019). A decrease in ice thickness of the largest Siberian Rivers during
the last 10 to 15 years (Shiklomanov and Lammers, 2014) could enhance the channel
connectivity to subchannel and groundwater flow, causing an increase in winter base flow, as
suggested by (Gurevich, 2009). An increased winter flow as well as increasing temperatures
125 in the Lena River are also supported by (Tananaev et al., 2016; Yang et al., 2002; Peterson et
al., 2002).

An important filter, counter-acting the methane flux into the atmosphere, is microbial
methane oxidation. Methane can be oxidized under anoxic conditions close to sediment
horizons where it is produced (Martinez-Cruz et al., 2017; Winkel et al., 2018) or during
130 migration through the oxic water column to the atmosphere (Mau et al., 2017a; Bussmann et
al., 2017). Under ice cover, it is important to consider methane oxidation below ice as it may
reduce the total amount of methane emitted to the atmosphere during ice-off. Active methane
oxidation and a methanotrophic community has been shown for permafrost thaw ponds and
lakes (Kallistova et al., 2019). Yet, the methane oxidation capacity in such lakes during ice
135 cover with low temperatures and low oxygen concentrations is unknown. In a study covering
several boreal lakes, methane oxidation was restricted to three lakes, where the phosphate
concentrations were highest (Denfeld et al., 2016). Rates of methane oxidation during the
winter have been found to be much lower than summer rates, yet, there is no clear consensus
on the factors limiting methane oxidation in winter (Ricão Canelhas et al., 2016). Beside
140 oxygen concentration, the geological background (i.e. yedoma type permafrost lakes versus
non-yedoma type lakes) also had a significant impact on the methane oxidation rate
(Martinez-Cruz et al., 2015).

Our study tests the hypothesis that winter ice blocks methane emissions, leading to the
accumulation of methane in the underlying water bodies. By studying hydrographically
145 different water bodies (lake, river and sea), we expect insights into the influence of water
column dynamics on methane accumulation to result. In addition, we measure methane
oxidation rates in the water column and in melted ice to assess oxidation as a potential sink.

150 2 Study area

This study was conducted on the southern coast of Bykovsky Peninsula in northeast Siberia,
Russia (Figure 1). Thermokarst lakes in that area commonly originated in the early Holocene
when surficial permafrost started to thaw, leading to accumulation of lake sediments with
organic contents of about 5-30 % (Biskaborn et al., 2016; Schelusner et al., 2015).

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Thermokarst lakes in the Lena River Delta seem to be ice free a little later after the coastal ice break up, depending strongly on the air temperature in the individual year (B. Juhls unpublished data).

160 Offshore of the Bykovsky Peninsula, part of the Yedoma Ice Complex is submerged and subsea permafrost is currently degrading. The coastline erodes at mean rates of between 0.5 and 2 m per year and can intersect inland water bodies, draining them or leading to the formation of thermokarst lagoons (Lantuit et al., 2011). The sea ice season in Tiksi Bay and the Buor-Khaya Bay typically starts in late September / early October and ends in beginning /
165 mid July, (Angelopoulos et al., 2019; Janout et al., 2020). Due to its isolation behind Muostakh Island and Cape Muostakh, sea ice tends to be preserved longer than in the Central Laptev Sea resulting in approx. 96 days of open water (Günther et al., 2015).

The Lena River has a mean annual discharge of 581 km³/year. It is the 2nd largest Arctic river by annual discharge and the 6th largest globally. There are no dams on the main-
170 stem of the Lena, but there is a dam on the Viluy River, one of the Lena's main tributaries (Holmes et al., 2013). The Lena's watershed is 2.6 million km², of which 70.5% is underlain by continuous permafrost (Juhls et al., 2020). Most of the water is discharged during end of May and beginning of June when the ice in the rivers breaks up, but the Laptev Sea is still covered by sea ice (Holmes et al., 2012). The main Lena River branches enter the Buor-
175 Khaya Bay through the northern and eastern part of the delta and through the Bykovskaya Channel with 20–25% of the Lena River water discharge (Charkin et al., 2011). The further distribution of the river water in Buor-Khaya Bay is mainly driven by the atmospheric systems of the cyclonic or anticyclonic Arctic circulation (Thibodeau et al., 2014; Wegner et al., 2013).

3 Material and Methods

3.1 Study sites

185 In the course of several expeditions to the Lena Delta (Siberia, Russia), we were able to repeatedly sample the same locations in winter and summer over the years (Table 1).

In September 2016, water samples were taken in the Bykovsky Channel and mouth of the Lena River (Overduin et al., 2017). In April 2017 with ice cover on the water bodies, ice cores were taken at Lake Golzovoye, the Lena River and Tiksi Bay, as well as the water below the ice sampled. Lake Golzovoye is an oval-shaped thermokarst lake about 0.5 km in
190 diameter with a maximal depth of 10 m, surrounded by Yedoma uplands at various stages of

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degradation and with no ice grounding in its center (Spangenberg, 2018; Strauss et al., 2018).
 195 Tiksi Bay is a shallow brackish bay at the southern end of Buor Khaya Bay, but still strongly
 influenced by the Lena River outflow. The water column is usually stratified, with a colder,
 more saline water underlying the brackish surface layer (Overduin et al., 2015). The water of
 the Lena River was sampled near Samoylov island. In July and August 2017, we sampled the
 same locations (Lena River near Samoylov, Tiksi Bay and Lake Golzovoye) with open water
 200 (Strauss et al., 2018). The transect to the “outer” Tiksi Bay has been investigated repeatedly
 over the previous years (Bussmann, 2013; Bussmann et al., 2017). In April 2018, again under
 ice samples were taken from the Lena River (Kruse et al., 2019).

Table 1: Locations and sampling dates of water samples and ice cores

Location	Sampling date	season	Number of samples
Lena River	7. – 8.9.2016	Summer	22
Lena River	19. – 22.4.2017	Winter	6
Lena River	29.7.2017	Summer	13
Lena River	21. – 24.4.2018	Winter	10
Lake Golzovoye	7. – 8.4.2017	Winter	8
Lake Golzovoye	7. – 8.4.2017	Ice cores	5
Lake Golzovoye	5.8.2017	Summer	5
Tiksi Bay	10.4.2017	Winter	2
Tiksi Bay	10.4.2017	Ice cores	4
Tiksi Bay	15.7 – 5.8.2017	Summer	6

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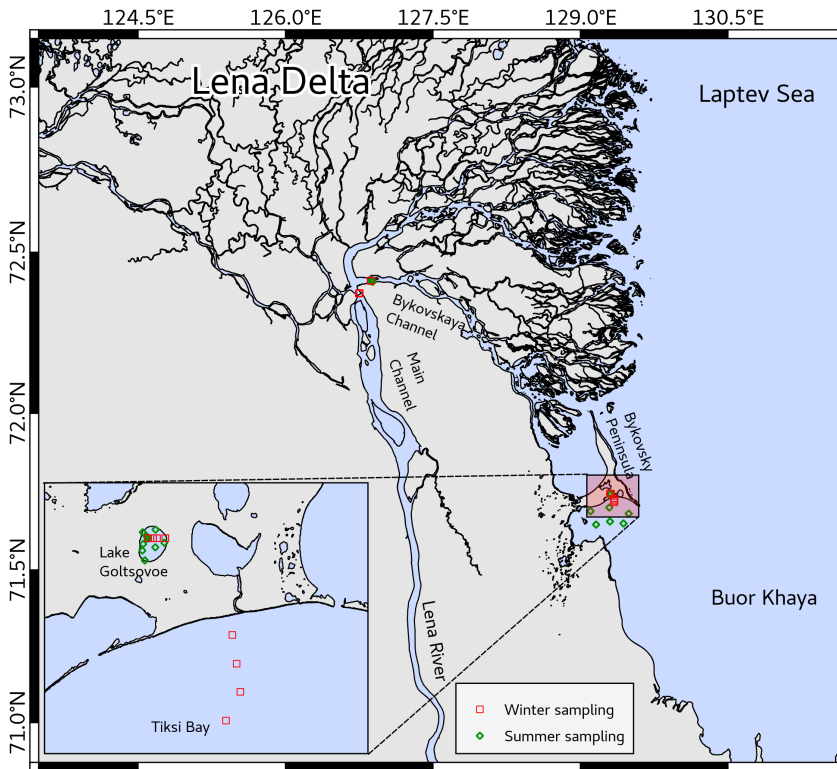


Figure 1: Map of the study area and sampling locations (winter sampling locations in red, summer sampling locations in green).

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3.2 Water sampling for methane analysis

In winter, water samples **at the ice – water interface**, were taken with a 1 L water sampler (Uwitec Austria), transferred to 0.5 L Nalgene bottles. In the field camp, the water was transferred with a 60 ml syringe into glass bottles, closed with butyl stoppers and crimps, poisoned with 0.2 ml 25% H₂SO₄ and stored upside down at 4 °C. In summer the water samples were directly transferred from the water sampler into the glass bottles, and further processed as described above.

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In winter 2017, due to problems of freezing, 40 ml of sample water was shaken for 2 min with 20 ml nitrogen in a 60 ml syringe. This headspace was then transferred into glass bottles filled with a saturated NaCl solution. Comparative measurements showed no significant difference between these two sampling strategies (Triputra, 2018). This dataset has been published already <https://doi.pangaea.de/10.1594/PANGAEA.905776> .

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225 3.3 Ice cores

In winter 2017, in addition to water sampling we also investigated ice cores for their methane content and methane oxidation rates. For each station three ice cores were drilled with a Kovacs Mark II ice coring system (9 cm diameter). One core was used to measure the in-situ temperature and back-up, one core was used for methane analysis, the third core was drilled for later molecular analyses. For determining the methane oxidation rates and later methane concentrations the ice cores were processed at the Research Station Samoylov Island. The top 10 cm, a 10 cm mid-section and three 10 cm-sections at the bottom of the core were cut off and transferred to special PVDF gas sampling bags (Keikaventures). The remaining parts of the cores were kept frozen. The bags were evacuated and the cores melted within approx. 5 h in a water bath at 8 °C. The sample was distributed with a 60 ml syringe to 3 glass bottles for determination of MOX and 1 bottle (which was poisoned immediately with H₂SO₄) for analysis of methane concentration (Strauss et al., 2018).

240 3.4 Methane analysis

Methane concentrations were determined via head-space method by adding 20 ml of N₂ as headspace and vigorous shaking for 2 min. Headspace methane concentrations were analyzed in the home laboratory with a gas chromatograph (GC 2014, Shimadzu) equipped with a flame ionization detector and a Shinyarbon column (Restek, USA). The temperatures of the oven, the injector, and detector were 100, 120, and 160 °C, respectively. The carrier gas (N₂) flow was 20 mL min⁻¹, with 40 mL min⁻¹ and 400 mL min⁻¹ synthetic air. Gas standards (Air Liquide) with methane concentrations of 10 and 100 ppm were used for calibration. The calculation of the methane concentration was performed according to (Magen et al., 2014). The precision of the calibration line was $r^2 = 0.99$ and the reproducibility of the samples was < 5%.

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3.5 Methane oxidation rate (MOX)

The MOX rate was determined by adding radioactive, tritiated methane to triplicate samples (Bussmann et al., 2015). The principle of the MOX rate determination is based on the ratio of produced tritiated hydrogen from the added tritiated methane. This ratio corrected for the incubation time gives the fractional turnover rate (k' [d⁻¹]). To obtain the MOX rate, k' is multiplied with the in-situ methane concentration. Radioactive, tritiated methane (0.1 ml of ³H-methane, 2 kBq ml⁻¹, American Radiolabeled Chemicals) was added to triplicate samples. Samples were incubated for 60 h at 1 °C, in the dark. Incubation was stopped by adding 0.2

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260 ml of 25% H₂SO₄. Abiotic controls were poisoned before adding the tracer. Radioactivity was determined with a liquid scintillation counter (Triathler, Finland) and Ultima Gold (Perkin Elmer) as a scintillation cocktail. For MOX, the limit of detection was calculated as described in Bussmann et al. (2015) and was determined to be 0.009 nmol L⁻¹ d⁻¹ for this data set.

In a set of experiments, we also assessed the influence of temperature on the MOX rate. Water samples (Main Channel, April 2017) that were incubated at temperatures from 1; 265 4; 7 and 10 °C show the temperature curve of the MOX reaction. At a previous cruise off Svalbard identical experiments were performed (Mau et al., 2017a) with water from stations HE449-CTD-2, 10, 33 and 37 incubated at temperatures from 0; 4; 8; 13 and 20 °C. We determined the Q10-factor, which indicates the temperature dependence of a biological 270 process according to (Raven and Geider, 1988):

$$Q_{10} = \exp\left(-10 * \frac{m}{T_{is}^2}\right)$$

where T_{is} is the in-situ temperature and “m” the slope of the regression line of the Arrhenius 275 plot (the inverse of the absolute temperature vs. the natural logarithm of the MOX rate).

3.6 Hydrochemistry and hydrology

Profiles of water temperature and conductivity were measured with a Cast-away CTD (SonTek) in summer 2016, and in winter and summer 2017. Water depth measurements were 280 made with an echo-sounding device (Garmin), every 10 m along the profile. Water velocity was measured in three horizons at each vertical profile: 0.2H; 0.6H; 0.8H, with H = total water depth) using a hydrological speed recorder (GR-21). Water discharges were calculated according to the recommendations for Russian hydrometeorological stations (Fedorova et al., 2015).

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4 Results

4.1 Methane concentrations and hydrochemistry

In the Lena River in summer 2016, median methane concentrations were 37 nM (n = 21) in 290 surface water, and significantly higher than the winter concentrations of 2017 (8 nM, Wilcoxon test, p = 0.0004, Figure 2). Comparing the summer and winter concentrations for 2017/18 also showed significantly higher values in summer with a median of 25 nM versus 10

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nM in winter (Wilcoxon test, $p = 0.0009$, Figure 2). Pooling the complete data set into winter and summer data for the Lena River showed that summer concentrations were significantly higher than winter concentrations (median of 31 nM versus 8 nM, $p < 0.001$). In summer 2016, water temperature ranged from 8.5 – 10.3 °C, electric conductivity from 135 – 185 $\mu\text{S cm}^{-1}$ and a pH-value around 6 that showed almost no variation in the water columns of 8.3 to 10 m (M. Winkler, unpublished data).

In winter 2017, the water column under the ice of the Lena River was about 1.3 °C cold with a conductivity of 275 $\mu\text{S cm}^{-1}$; no stratification was evident. The water was flowing at a speed of 0.043 m sec^{-1} under the ice and with 0.1 m sec^{-1} above the bottom (0.85 H) (Strauss et al., 2018) chapter 2.5. In the Main Channel of the Lena River the maximal water depth in winter 2017 was 25.3 m, versus 28 m in summer at the same point. In winter the water velocity in the channels is much lower than in summer: in April 2017 it was 0.18 m/sec in the Bykovskaya channel but reached 1.53 m/sec in summer 2016.

In summer 2017, the water discharge in Bykovskaya channel was 5,313.4 m^3/sec . Water temperature ranged from 13 – 14 °C with no changes with water depth, indicating a full mixing (Strauss et al., 2018) chapter 2.8. In Winter 2018, the water column under the ice of the Lena River was about -0.7 °C cold with a median conductivity of 465 $\mu\text{S cm}^{-1}$ and a median pH of 7.3. In Bykowski Channel oxygen saturation had a median value of 51%. No stratification of the water column was evident; based on water temperature and conductivity (Kruse et al., 2019). These findings agree to the Lena River monitoring observations by (Juhls et al., 2020).

In Lake Golzovoye, in summer 2017, the median methane concentration was 49 nM ($n = 5$), while in winter concentrations were about 40-times higher with a median of 1957 nM ($n = 8$, Wilcoxon test, $p = 0.002$, Figure 2). In winter, the water temperature of the lake was cold at the surface (median 0.3 °C in top 1 m) and warmer (2 °C) at the bottom with a thermocline at 2 meters. In summer, the surface water was heated to 7 – 10 °C, while the bottom water only warmed to 4 – 5 °C.

In Tiksi Bay and in summer 2017, the median methane concentration was 14 nM ($n = 6$). In contrast, in winter concentrations were about 7-times higher with a median of 103 nM ($n = 2$). Due to low sample numbers, no statistical test was possible. In summer 2017, water temperature ranged from 4.3 – 10.9 °C, electric conductivity from 20 – 6200 $\mu\text{S cm}^{-1}$. In winter, bottom water had a temperature of 3.3 °C and 0.3 °C at the top 1 m, with a thermocline at 1 – 2 meters.

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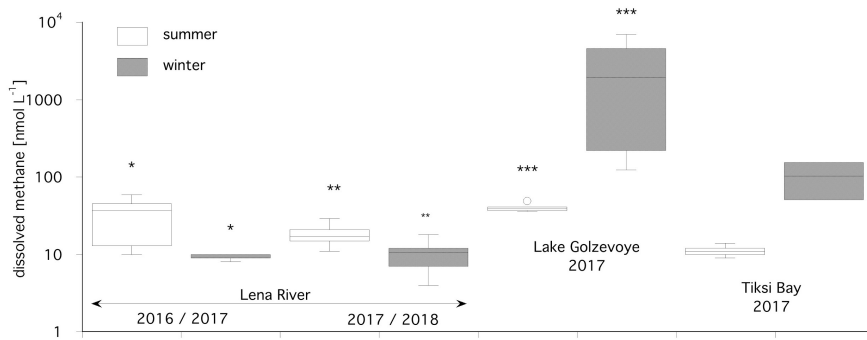


Figure 2: Median summer and winter methane concentrations at the Lena River, Lake Golzevoye and Tiksi Bay in the years 2016 – 2018. Bright columns indicate summer values, dark columns indicate winter values. The Asterisks indicate significantly different data sets.

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4.2 Ice cores

Ice cores were taken at Lake Golzevoye and in Tiksi Bay, whereas no ice core data are available for the Lena River itself (Strauss et al., 2018). Methane concentrations in the ice cores of Lake Golzevoye and Tiksi Bay were rather low (both with a median of 9 nM). No depth gradients from the ice surface, middle section and the three lower most sections were evident. On a close up for the bottom layers, there was a slight increase of methane towards the ice-water interface for the ice cores from Lake Golzevoye, but not from Tiksi Bay. In Figure 3 the median methane concentrations at the ice-water interface are related to the concentration in the ice cores. Water column concentrations were 11 times and 109 times higher than in the ice cores, for Tiksi Bay and Lake Golzevoye, respectively, with a median of 102 nM and 985 nM (Bussmann and Fedorova, 2019). However, in one core of Lake Golzevoye (core #24), concentrations were orders of magnitude higher throughout the core (854 – 11091 nM) and 6954 nM in the water below (Figure 3).

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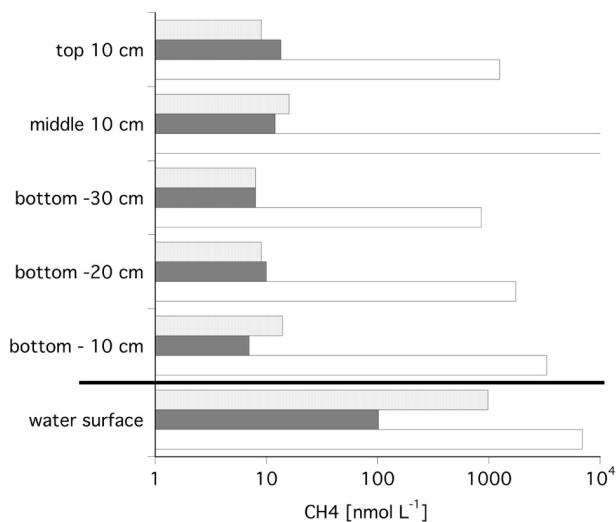


Figure 3: Median methane concentrations in the ice cores and in the water below the ice; for Lake Golzovoye (in light grey), with core #24 separately (in white), and Tiksi Bay in dark grey. Note the logarithmic scale.

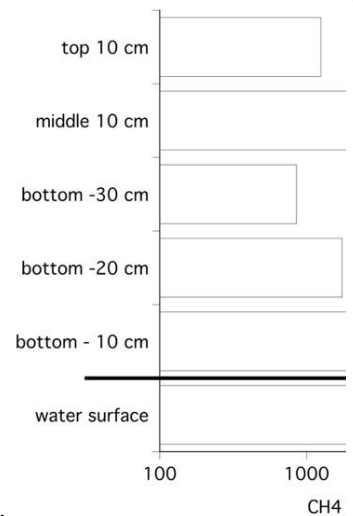
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4.3 Methane oxidation rates (MOX)

Methane oxidation rates were determined in the melted water from the ice cores from the different locations and in water from the Lena River. Due to logistic restraints at the field sites, no direct measurements of MOX in the waters of Lake Golzovoye and Tiksi Bay was possible. In the first step we determined the fractional turnover rate k' . In the ice cores, k' was very patchy distributed and 73% of all samples were below the detection limit. In all positive samples, the value for k' was rather stable with a median of 0.003 per day ($n = 31$). Even though very patchy and many samples being below the detection limit, in some samples of the ice cores a low MOX rate could be detected (on average $0.011 \text{ nmol L}^{-1} \text{ d}^{-1}$). In the water samples below the ice, the fractional turnover rate k' was never below the detection limit. The median k' determined for all water samples (Lena River) was also 0.003 ($n = 14$). To calculate the MOX for all water samples, we multiplied the median k' of 0.003 obtained from Lena River and ice cores with the respective in-situ methane concentration of the water columns of Lake Golzovoye, Tiksi Bay and Lena River.

Highest MOX rates were found in the water below station #24 in Lake Golzovoye ($20.36 \text{ nmol L}^{-1} \text{ d}^{-1}$), where also very high methane concentrations were observed (Figure 4). The median MOX in the other water samples of Lake Golzovoye was about one order of



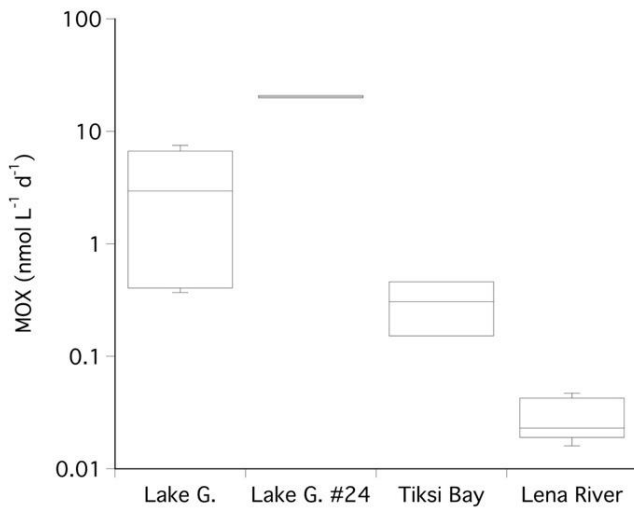
hat gelöscht: Figure 4: Methane concentrations in the ice core #24 and in the water below the ice in Lake Golzovoye. Note the logarithmic scale. ¶

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magnitude lower ($2.95 \text{ nmol L}^{-1} \text{ d}^{-1}$). In Tiksi Bay and in the Lena River MOX rates decreased by further orders of magnitude (median of 0.305 and $0.023 \text{ nmol L}^{-1} \text{ d}^{-1}$, Figure 4).



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| *Figure 4: Box plot of the calculated methane oxidation rates in water under ice cover at Lake Golzovoye, at the location of ice core #24, Tiksi Bay and the Lena River. Note the logarithmic scale.*

4.4 Temperature influence on MOX

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To assess the influence of temperature on the MOX, we incubated water samples at different temperatures and determined their MOX rate. As expected, with increasing temperature the MOX rate also increased (Figure 5). The Q10 calculated for these water samples from the Lena River was 2.72 ± 0.69 . For polar and marine waters off Svalbard a Q10 of 2.99 ± 0.86 was calculated.

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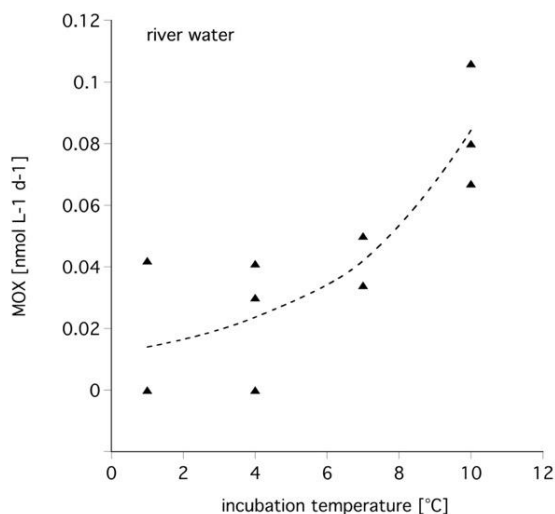


Figure 5: Influence of incubation temperature on methane oxidation rate for Lena River water, winter 2017.

395 5 Discussion

5.1 Methane concentrations and hydrochemistry

In this study we compared the methane concentration under ice cover (winter) with open water situations (summer) in three different water bodies.

In winter the methane concentrations in the Lena River were 4times lower as in
 400 summer (Table 2). The Lena River displays a reduced but still substantial water flow /
 discharge under ice cover. In 2017, the discharge in winter (March and April with 2.830 and
 2.185 m³ sec⁻¹) was about 10-times lower than in summer (July and August with 29.800 and
 26.000 m³ sec⁻¹, (Shiklomanov et al., 2018).

In winter there are only a few possible sources of methane left. The surrounding soils
 405 of the drainage basins are all frozen. The ground below the main channels of the Lena is still
 unfrozen (Fedorova et al., 2019), however the sediment consists of coarse grain sizes and is
 poor in organic material (Rivera et al., 2006) and we do not expect any in situ methane
 production. Especially in the Lena River a substantial amount of POC originates from
 thermokarst-induced, abrupt collapse of Pleistocene Ice Complex deposits. These events
 410 mainly occur in late summer, the signal however, is still visible in winter (Wild et al., 2019).
 With these collapses [also](#), methane could be imported to the river. So, we could only detect
 low methane concentrations in winter. In contrast, in summer, the active layer soils from the

415 drainage basin allow for several sources of methane and thus increased methane concentrations in summer. Also, methane could be transported from the southern Lena catchment towards our study area, as is suggested for particulate organic matter (Winterfeld et al., 2015). At least during the warm season, methane production from (temperate) river sediments is possible (Bednařík et al., 2019). About the river-ice on the Lena there is no information yet available.

420 In Tiksi Bay, we observed a 7-times increase of dissolved methane under ice cover, compared to open water conditions (Table 2). Tiksi Bay is part of Buor Khaya Bay and via the central Laptev Sea perennially connected to the Arctic Ocean. Not much is known about tidal surge or water movement in Tiksi Bay under ice cover. It is anticipated that the ice cover on Tiksi Bay will lead to a decrease in tidal amplitudes and velocities (Fofonova et al., 2014).
435 The structure of ice formation in Tiksi Bay also suggest that even in winter it is still an open system connected to the outer bay (Spangenberg et al., submitted). Sources of methane could be through diffusion of methane from the underlying sediment (Bussmann et al., 2017), where methane is produced by the degradation of organic material. However, as aerobic methane oxidation in the water column is impaired by low temperatures, methane concentrations in water increase.

440 In Lake Golzovoye, dissolved methane concentrations increased by 40-times from summer to winter (Table 2). Lake Golzovoye is an isolated freshwater lake with presumably only a weak thermohaline circulation (Leppäranta, 2015; Spangenberg et al., submitted). A similar, seasonal pattern of methane among lake waters of the Mackenzie Delta has been observed, ranging from very high concentrations at the end of winter beneath lake-ice (> 200.000 nM) to considerably lower concentrations during open water, particularly by late summer (approx. 1000 nM) (Cunada et al., 2018). For Yedoma lakes in Alaska, the same pattern was observed with high methane concentrations in winter, and a decrease of more than one order of magnitude in summer, (even though the actual concentrations > 900,000 nM were much higher than in this study) (Martinez-Cruz et al., 2015; Sepulveda-Jauregui et al., 2015). The same pattern is reported for boreal Swedish lakes (Denfeld et al., 2018). The source of methane in the water column of Lake Golzovoye is presumably the sediment, where high concentrations and active methane production, sulfate reduction and anaerobic methane oxidation have been observed in winter 2017 (S. Liebner, C. Knoblauch, unpublished data).

445 The role of water velocity and water column mixing is not clear, but our data suggest that in a stagnant water body such as a lake more methane accumulates under ice than in a water body with running water (river). Water column turbulent diffusivity has a major

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influence on the methane cycle, where higher turbulence potentially leads to greater
450 proportion of methane being oxidized and lower turbulence leads to a greater proportion
being stored (Vachon et al., 2019).

5.2 Ice cores

The median methane concentration of all ice cores for Lake Golzovoye and Tiksi Bay was 9
455 nM, which was supersaturated compared to atmospheric concentrations for which equilibrium
concentration would be 5 nM. More details on the ice formation in the different water bodies
are given in (Spangenberg et al., submitted). Compared to the methane concentrations in the
water, the concentrations in the ice were 1 - 2 orders of magnitude lower (Table 2). This
means that, in terms of methane, a complete separation of the water body from the
460 atmosphere can be assumed. As mentioned earlier in this study and in (Spangenberg et al.,
submitted), core #24 in Lake Golzovoye had much higher methane concentrations throughout
the core and visible inclusions of (methane) bubbles. We assume that core #24 was located
above an active ebullition site, which might have slowed ice formation and prolonged direct
methane release to the atmosphere.

465 In the ice itself, 28% of the samples showed methane oxidation capability. During ice
formation most free-living bacteria are lost from the liquid phase through incorporation into
the ice, while bacterial aggregates remain in the water (Santibáñez et al., 2019). In an
experimental setup, (Wilson et al., 2012) show that multiple freeze-thaw cycles in water from
freshwater lakes reduce the total bacterial cell number at least 100,000-fold. In addition,
470 methanotrophic bacteria are particularly sensitive to freezing and thawing (Green and
Woodford, 1992; Hoefman et al., 2012). These findings could explain the reduced activity of
methanotrophic bacteria within the ice cores.

Also, we did not detect any discolorations or other indications of photosynthesis or
other biological processes in the bottom layer of the ice cores. Thus, we conclude that the
475 ecosystem of freshwater ice and its lower margin do not reach the richness observed in polar
sea ice (Leppäranta, 2015).

5.3 Methane oxidation rates (MOX)

In this study we determined the methane oxidation rate with tritiated methane as tracer. The
480 advantage of the tracer injection method is that natural low concentrations are hardly altered
and thus we assume that our values are close to the actual rates. The fractional turnover rate k'
was determined in ice cores from the lake and Tiksi Bay, and in river water. Within these
locations k' was evenly distributed. However, k' may vary between different environments

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490 (river, lake and brackish water) as well as between ice cores and underlying water. The
fractional turnover rate is influenced by temperature, methane and oxygen concentrations
(Steinle et al., 2017). Thus, in our calculations an error is embedded. However, as temperature
495 was low at all locations, and the methanotrophic population of the ice cores is similar to the
ones in the water below, we assume that the application of one k' to all samples is a good
assumption.

Our data span three orders of magnitude, ranging from $0.02 \text{ nmol L}^{-1} \text{ d}^{-1}$ in the Lena
River, 0.31 in Tiksi Bay and $2.95 \text{ nmol L}^{-1} \text{ d}^{-1}$ in Lake Golzovoye. Another polar study finds a
MOX of $0.004\text{--}1.09 \text{ mg C m}^{-3} \text{ d}^{-1}$ ($= 0.33\text{--}91 \text{ nmol L}^{-1} \text{ d}^{-1}$) at the water-ice interface of a
500 Swedish lake (Ricão Canelhas et al., 2016). Also (Bastviken et al., 2002) report a MOX of
similar range ($0.001\text{--}39 \text{ mg C m}^{-3} \text{ d}^{-1} = 0.08\text{--}3250 \text{ nmol L}^{-1} \text{ d}^{-1}$). However, the methods of
determining MOX were quite different, and more of a potential rate. Another study in
Yedoma lakes in Alaska also reports higher MOX ($0.03\text{--}0.28 \text{ mg methane L}^{-1} \text{ d}^{-1} = 1875\text{--}$
 $17500 \text{ nmol L}^{-1} \text{ d}^{-1}$) with a kinetic approach to determine MOX (Martinez-Cruz et al., 2015).
505 In marine polar waters off Svalbard MOX was determined with the same tracer method and
ranged from $1.6\text{--}2.2 \text{ nmol L}^{-1} \text{ d}^{-1}$ in summer (Mau et al., 2017b). Thus, our data are within
the very low range of reported MOX rates in polar regions, probably due to methodological
differences.

In previous years we determined MOX in the study area during summer, applying the
510 same method as in this study. Therefore, we can approach a seasonal comparison (winter vs.
summer), assuming interannual variability is negligible and neglecting spring and autumn
mixing. To estimate the importance of ice cover on the overall MOX, we assume an ice
coverage of 270 days for Lake Golzevoye and Tiksi Bay (Cortés and MacIntyre, 2020) and
160 days for the Lena River (Shiklomanov and Lammers, 2014). By multiplying the
515 respective winter and summer MOX with the days of ice cover and days of open water, we
can calculate the amount of methane oxidized during ice cover versus ice off time.

For the Lena river and permafrost lakes we compare our winter data with summer data
obtained in July 2012 (Osudar et al., 2016). For the Lena River, with a median MOX of 22.8
 $\text{nmol L}^{-1} \text{ d}^{-1}$ ($n = 8$), the summer MOX was about 3 orders of magnitude higher than in winter
520 ($0.023 \text{ nmol L}^{-1} \text{ d}^{-1}$). The amount of methane oxidized during ice off (4674 nmol/L) was about
1270-times more than the amount oxidized during ice cover (4 nmol/L).

For MOX in lakes, summer rates from small lakes near Research Station Samoylov
Island were 36-times higher (median $107 \text{ nmol L}^{-1} \text{ d}^{-1}$, $n = 6$) than winter rates (2.95 nmol L^{-1}

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d⁻¹). Also, for Yedoma lakes in Alaska summer values of MOX were about 10-times higher than winter values (Martinez-Cruz et al., 2015). The amount of methane oxidized during open water (10165 nmol/L) was about 13-times more than the amount oxidized during ice cover (797 nmol/L).

For Tiksi Bay there are also summer values of MOX available (Bussmann et al., 2017). However, with a median summer rate of 0.419 nmol L⁻¹ d⁻¹ for surface, riverine water there is little difference when compared to our winter data (0.31 nmol L⁻¹ d⁻¹, Table 2). The amount of methane oxidized during open water (40 nmol/L) was about 2-times less than the amount oxidized during ice cover (84 nmol/L).

Table 2: Comparison of methane concentration in water and ice, as well as methane oxidation rates (MOX) at different sites and different seasons.

Location	[CH ₄]winter / [CH ₄]summer*	Under ice accumulation**	Water velocity winter	[MOX] winter	MOX _{during ice off} / MOX _{during ice coverage} ***
Lena River	S > W, x 5	n.a.§	strong	Low, S >> W	4674 / 4 nmol/L 1270
Lake Golzovoye	W > S, x 40	Wa >> I, x 109	minor	High, S > W	10165 / 797 nmol/L 13
Tiksi Bay	W > S, x 7	Wa > I, x 11	medium	Medium S = W	40 / 84 nmol/L 0.5

§ n.a. not available

*comparing dissolved methane concentrations in water in winter and summer

**comparing methane concentration in lowest ice core layer with underlying water

*** MOX winter data from this study, summer data from (Osudar et al., 2016) and (Bussmann et al., 2017); assuming 270 days of ice coverage for Lake Golzovoye and Tiksi Bay, and 160 day of ice coverage for Lena River.

There still seems to be no clear consensus on the factors limiting MOX in winter. In several boreal lakes MOX was restricted to lakes, where the phosphate concentrations were highest (Denfeld et al., 2016). Another study reports that in winter MOX is mainly controlled by the dissolved oxygen concentration, while in the summer it was controlled primarily by the methane concentration (Martinez-Cruz et al., 2015). The stratification of lakes determining

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the availability of methane and oxygen for the methanotrophic bacteria also strongly influences MOX (Kankaala et al., 2006; Kankaala et al., 2007).

555 Temperature is also an important factor affecting winter MOX. MOX is observed at temperatures of 2 °C, mostly by *Methylococcaceae* (Ricão Canelhas et al., 2016). A recent study with *in situ* concentrations in a northern temperate lake observes a Q10 of 2.4 ± 1.4 (Thottathil et al., 2019), which is a bit lower than the Q10 of this study with 2.7 and 2.9 for polar, fresh and marine water, respectively. In addition, there are co-correlations between
560 temperature and methane concentration. At substrate (methane) saturation, temperature has a strong influence, while under substrate limiting conditions, temperature has a minor influence (Lofton et al., 2014; King and Adamsen, 1992). In contrast, (Thottathil et al., 2019), observed a strong temperature response for MOX across the entire range of ambient methane concentrations. Measuring MOX with tritiated methane and thus at close to *in situ*
565 concentrations, we can compare the Q10 from this study with data from temperate environments. The Q10 for polar environments was higher than Q10 values obtained from temperate waters (Elbe River, Germany and North Sea, (Bussmann et al., 2015).) with 1.52 and 1.75, respectively (Figure 6). Although the substrate concentration in the temperate waters was higher, MOX from polar regions seems to react more sensitively to a temperature
570 increase. One explanation could be that the temperature optimum of psychrotolerant methanotrophs is below 20 °C (Bale et al., 2019). Thus, the polar methanotrophs are further away from their optimum temperature and react with increased activity to temperature increases while methanotrophic bacteria from temperate waters are nearer to their optimum temperature and do not react as sensitively.

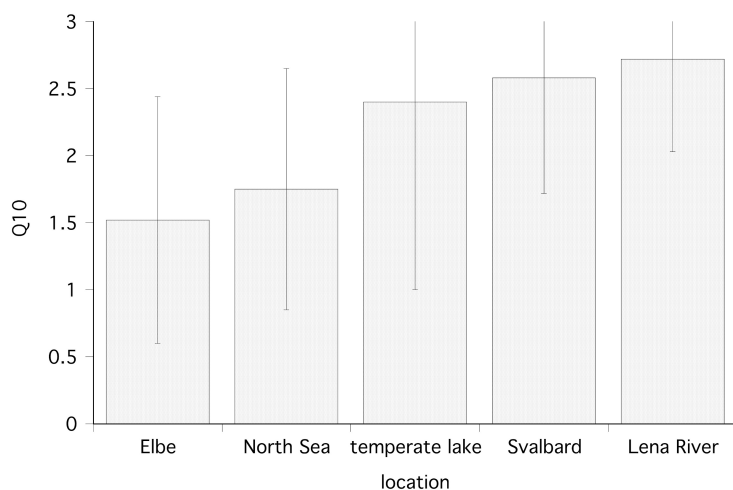
575 Environmental conditions between winter and summer conditions certainly differ and may also affect the population structure of methanotrophs. Some psychrophilic strains adapt to colder temperatures (20 °C versus 4 °C) by modifying their fatty acid composition (Bale et al., 2019), while others may have only limited abilities, resulting in different population structures. This is supported by a recent study revealing that wintertime Arctic bacterial
580 communities and food webs structure change based on carbon availability (Kellogg et al., 2019).

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585 Figure 6: Temperature dependence (Q_{10}) of the methane oxidation rate at different locations; shown are the mean and standard deviation. Data for Elbe and North Sea are from Bussmann et al. (2015), data for the temperate lake are from Thottathil et al. (2019).

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In this study we compared the methane inventory (concentrations) and the biological sink (methane oxidation) of three polar aquatic environments under summer and winter conditions. For a complete budget, the methane sources should be known, as there is methane input from the sediment, either by diffusion or ebullition and lateral input by groundwater, river flow or water circulation in the bay. Additional sinks for the systems are methane flux from the water into the atmosphere and lateral output by water circulation. In the following we apply our results on the methane cycle of the three different environments:

595

In the river we find higher methane concentrations and higher MOX in summer. The low concentration in winter are probably due to low methane input from the frozen borders and a reduced but still effective dilution of methane by the water flow (Fedorova et al., 2019). MOX is low, and thus will not contribute to the removal of methane from the river. The ice off on the river will probably not increase methane emissions, as only minor amounts are accumulated under the ice.

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In the lake, we observed a strong accumulation of methane in winter under ice cover. Thus, either the methane sources are strengthened and/ or the sinks have weakened. In winter there is an active cycle of methanogenesis and (anaerobic) methane oxidation in the sediment (S. Liebner, unpublished data). However, we assume that this activity is the same or less than in summer. Ebullition does occur in winter (as shown for ice core #24) and thus will lead to locally increased methane concentrations. The methane sink, flux from the water into the

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In winter when the water bodies are ice covered and methane emissions are stopped, MOX in the water would be the only sink left. However, MOX under winter conditions is reduced, either by low temperatures (this study), low oxygen or phosphate content (Martinez-Cruz et al., 2015; Denfeld et al., 2016). The turnover time of MOX (the time needed to consume all of the present methane) is the inverse of our determined median k' , i.e. 333 days. Thus, with an estimated ice coverage of 200 days, MOX will not contribute significantly to a methane reduction, and about 60% of the methane inventory can be consumed. If we consider the winter and summer conditions as two extremes (low concentration and high MOX in summer versus high concentrations and low MOX in winter); we assume that after ice-on, both parameters increase in a linear way. Thus, the duration of ice cover is important and we assume that a shorter ice cover results in less high methane concentrations under ice and subsequently in a reduced pulse of methane emissions at ice-off. In a changing, warming Arctic, a shortened time of ice coverage on lakes and rivers is predicted (Prowse et al., 2011). In respect to our study this would imply a shortened time for methane accumulation under ice and a shortened time for the less efficient winter-MOX. Thus, an extended time of ice-free conditions could reduce the methane emissions from Arctic water bodies. However, it has to be kept in mind that not much is known about the MOX during water column mixing in spring or autumn. At least one study reports MOX as the dominant methane sink at ice-off (Utsumi et al., 1998b).

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640 atmosphere is cut off by the ice cover, thus the only remaining sink is MOX, which is reduced by low temperature and other environmental factors as discussed above. During and after ice-off, altered or weakened water column stratification will allow a mixing of the water column. This results in increased methane emission, but also to enhanced MOX as more oxygen and nutrients will become available (Utsumi et al., 1998b). In summer increased MOX and methane flux from the water lead to reduced methane concentrations in the water.

645 In the bay, we observed an accumulation of methane under ice and higher concentrations in winter. Thus, we assume that the sinks have weakened, with a stable or reduced methane input. Our comparison shows that MOX does not change significantly between the seasons, thus the other main sink, transport via water exchange of the bay with the shelf water is reduced during winter because of the ice cover (Fofonova et al., 2014) and direct flux from open water is reduced by the ice cover. There probably is still an input of methane from the sediments, which results than in a slight accumulation of methane. The ice off in the bay will result in increased methane emissions and also reduced methane concentrations when water circulation in the bay re-starts.

655 In a changing, warming Arctic, a shortened time of ice coverage on lakes and rivers is predicted (Prowse et al., 2011); (Newton and Mullan, 2020; Benson et al., 2012). This could be -6 days per 1°degree of temperature increase (Newton and Mullan, 2020). Another scenario is given by (Benson et al., 2012) with -17 days / 100 years. For the lake we observed the greatest difference between the ice-covered winter situation and open water. Especially in the lake, the duration of ice cover is important and we assume that a shorter ice cover results in less high methane concentrations under ice and subsequently in a reduced pulse of methane emissions at ice-off. In respect to our study this would imply a shortened time for methane accumulation under ice and a shortened time for the less efficient winter-MOX. For lakes this would result in increased MOX during ice off with a ratio of $MOX_{\text{during ice off}} / MOX_{\text{during ice coverage}}$ of 14 and 16, versus 13 today. For rivers the same trend can be assumed with the ratio increasing from today's 1270 towards 1358 or 1539 for the two scenarios.

665 Thus, an extended time of ice-free conditions could reduce the methane emissions from Arctic water bodies. However, it has to be kept in mind that not much is known about the MOX during water column mixing in spring or autumn.

Feldfunktion geändert

hat gelöscht: ¶

6 Conclusions

Our work on an eastern Siberian lake, river and marine bay showed that methane accumulates under ice cover during the winter and is consumed differently in the three water bodies. Our study was restricted to late winter and mid-summer, which represents two extremes of the annual cycle. Other processes during autumn mixing, ice-on, ice off are not considered.

hat gelöscht: , following the ice-covered and summer periods

Two main physical factors affecting the methane cycle in the water bodies under ice cover are the water velocity and the ice cover itself. In most of our ice cores no concentration gradient between the bottom of the ice cores and the top was obvious. As we could hardly detect any MOX within the ice cores, we assume that methane is not integrated into the ice during freeze up. Therefore, the ice cover seems to effectively prevent any methane flux from the highly accumulated methane concentrations in the water towards the atmosphere. In the river with running water under the ice cover, only a minor accumulation of methane was observed. In the bay with a restricted but still present water movement, dilution or mixing with other water bodies, allowed for a moderate accumulation of methane. In the small lake, we assume a stagnant water body with a subsequent accumulation of high amounts of methane.

The biotic counterpart of the observed methane accumulation is microbial methane oxidation (MOX). In most cases, MOX in summer was much higher than in winter. We observed a strong dependence of MOX on the temperature, and with in-situ temperatures of only 1 °C in winter subsequently low rates were observed. Higher methane concentrations in winter indicate the methanotrophic bacteria were not limited by substrate (methane) concentrations. However, especially in Lake Golzovoye and its stagnant water body oxygen could become a limiting factor. Other factors could be nutrient limitation or shift in the population structure.

In a changing, warming Arctic, a shortened time of ice coverage on the water bodies is predicted. In respect to our study this would imply a shortened time for methane to accumulate below the ice and a shorter time for the less efficient winter-MOX. Especially for lakes, an extended time of ice-free conditions could reduce the methane flux from the Arctic water bodies.

hat gelöscht: predicted.

hat gelöscht: Thus

Data availability:

Data on methane concentration and MOX are available at the [Pangea](#) database.

[Bussmann, I; Fedorova, I; Juhls, B et al. \(2020\): Dissolved methane concentrations and oxidation rates in ice cores from the Lena Delta area, 2016-2018.](https://doi.pangaea.de/10.1594/PANGAEA.920013)
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710 [Bussmann, I; Fedorova, I; Juhls, B et al. \(2020\): Dissolved methane concentrations and oxidation rates in water samples from the Lena Delta area, 2016-2018.](https://doi.pangaea.de/10.1594/PANGAEA.919986)
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715 Author contribution:
All authors carried out field work and measurements and collected samples. IB performed the methane and MOX analyzes. IB, BJ, PPO and MW contributed to the initial and final versions of the manuscript.

720 Competing interests:
The authors declare that they have no conflict of interest.

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References

- 740 Angelopoulos, M., Westermann, S., Overduin, P., Faguet, A., Olenchenko, V., Grosse, G.,
and Grigoriev, M. N.: Heat and salt flow in subsea permafrost modeled with CryoGRID2,
Journal of Geophysical Research - Earth Surface, 124, 920-937, doi: 10.1029/2018JF004823
2019.
- 745 Bale, N. J., Rijpstra, W. I. C., Sahonero-Canavesi, D. X., Oshkin, I. Y., Belova, S. E.,
Dedysh, S. N., and Sinninghe Damsté, J. S.: Fatty acid and hopanoid adaption to cold in the
methanotroph *Methylovulum psychrotolerans*, Front. Microbiol., 10, 589, 2019.
- Bastviken, D., Ejlertsson, J., Sundh, I., and Tranvik, L.: Measurement of methane oxidation in
lakes: a comparison of methods, Environ. Sci. Technol., 36, 3354-3361, 2002.
- 750 Bastviken, D., Cole, J., Pace, M., and Tranvik, L.: Methane emissions from lakes:
Dependence of lake characteristics, two regional assessments, and a global estimate, Glob.
Biogeochem. Cycl., 18, GB4009, doi:10.1029/2004GB002238, 2004.
- 755 Bednařík, A., Blaser, M., Matoušů, A., Tušer, M., Chaudhary, P. P., Šimek, K., and Rulík,
M.: Sediment methane dynamics along the Elbe River, Limnologia, 79, 125716,
<https://doi.org/10.1016/j.limno.2019.125716>, 2019.
- 760 Benson, B. J., Magnuson, J. J., Jensen, O. P., Card, V. M., Hodgkins, G., Korhonen, J.,
Livingstone, D. M., Stewart, K. M., Weyhenmeyer, G. A., and Granin, N. G.: Extreme events,
trends, and variability in Northern Hemisphere lake-ice phenology (1855–2005), Clim.
Change, 112, 299-323, 10.1007/s10584-011-0212-8, 2012.
- 765 Biskaborn, B. K., Subetto, D. A., Savelieva, L. A., Vakhrameeva, P. S., Hansche, A.,
Herzschuh, U., Klemm, J., Heinecke, L., Pestryakova, L. A., Meyer, H., Kuhn, G., and
Diekmann, B.: Late Quaternary vegetation and lake system dynamics in north-eastern Siberia:
Implications for seasonal climate variability, Quaternary Science Reviews, 147, 406-421, doi:
10.1016/j.quascirev.2015.08.014, 2016.
- 770 Bussmann, I.: Distribution of methane in the Lena Delta and Buor Khaya Bay, Russia,
Biogeosciences, 10, 4641–4465, doi:10.5194/bg-10-4641-2013, 2013.
- Bussmann, I., Matousu, A., Osudar, R., and Mau, S.: Assessment of the radio 3H-CH4 tracer
technique to measure aerobic methane oxidation in the water column Limnol. Oceanogr.:
Methods, 13, 312-327, doi:10.1002/lom3.10027, 2015.
- 775 Bussmann, I., Hackbusch, S., Schaal, P., and Wichels, A.: Methane distribution and oxidation
around the Lena Delta in summer 2013, Biogeosciences, 14, 4985-5002, doi.org/10.5194/bg-
14-4985-2017, 2017.
- 780 Bussmann, I., and Fedorova, I.: Dissolved methane concentrations under ice cover in the Lena
Delta area., 2019.
- 785 Charkin, A. N., Dudarev, O. V., Semiletov, I. P., Kruhmalev, A. V., Vonk, J. E., Sánchez-
García, L., Karlsson, E., and Gustafsson, Ö.: Seasonal and interannual variability of
sedimentation and organic matter distribution in the Buor-Khaya Gulf: the primary recipient
of input from Lena River and coastal erosion in the southeast Laptev Sea, Biogeosciences, 8,
2581-2594, 2011.

- Cole, J. J., Prairie, Y. T., Caraco, N. F., McDowell, W. H., Tranvik, L. J., and Striegl, R. G.: Plumbing the global carbon cycle: Integrating inland waters into the terrestrial carbon budget, *Ecosystems*, 10, 172–185, doi: 10.1007/s10021-006-9013-8, 2007.
- 790 Cortés, A., and MacIntyre, S.: Mixing processes in small arctic lakes during spring, *Limnol. Oceanogr.*, 65, 260-288, 10.1002/lno.11296, 2020.
- 795 Cunada, C. L., Lesack, L. F. W., and Tank, S. E.: Seasonal dynamics of dissolved methane in lakes of the Mackenzie Delta and the role of carbon substrate quality, *Journal of Geophysical Research: Biogeosciences*, 123, 591-609, 10.1002/2017JG004047, 2018.
- 800 DelSontro, T., McGinnis, D. F., Wehrli, B., and Ostrovsky, I.: Size does matter: Importance of large bubbles and small-scale hot spots for methane transport, *Environmental Science & Technology*, 49, 1268-1276, 10.1021/es5054286, 2015.
- 805 Denfeld, B. A., Ricão Canelhas, M., Weyhenmeyer, G. A., Bertilsson, S., Eiler, A., and Bastviken, D.: Constraints on methane oxidation in ice-covered boreal lakes, *Journal of Geophysical Research G: Biogeosciences*, 121, 1924-1933, 10.1002/2016JG003382, 2016.
- Denfeld, B. A., Klaus, M., Laudon, H., Sponseller, R. A., and Karlsson, J.: Carbon dioxide and methane dynamics in a small boreallake during winter and spring melt events, *Journal of Geophysical Research: Biogeosciences*, 123, 2527-2540, 10.1029/2018JG004622, 2018.
- 810 Deshpande, B. N., MacIntyre, S., Matveev, A., and Vincent, W. F.: Oxygen dynamics in permafrost thaw lakes: Anaerobic bioreactors in the Canadian subarctic, *Limnol. Oceanogr.*, 60, 1656-1670, 10.1002/lno.10126, 2015.
- 815 Fedorova, I., Chetverova, A., Bolshiyarov, D., Makarov, A., Boike, J., Heim, B., Morgenstern, A., Overduin, P. P., Wegner, C., Kashina, V., Eulenburg, A., Dobrotina, E., and Sidorina, I.: Lena Delta hydrology and geochemistry: long-term hydrological data and recent field observations, *Biogeosciences*, 12, 345-363, 10.5194/bg-12-345-2015, 2015.
- 820 Fedorova, I., Bobrov, N., Pankova, D., Konosavskii, P., and Alekseeva, N.: Modeling of thermic process of the arctic ecosystems, *International Multidisciplinary Scientific GeoConference Surveying Geology and Mining Ecology Management, SGEM*, 2019, 401-409,
- 825 Fofonova, V., Androsov, A., Danilov, S., Janout, M., Sofina, E., and Wiltshire, K.: Semidiurnal tides in the Laptev Sea Shelf zone in the summer season, *Cont. Shelf Res.*, 73, 119-132, 10.1016/j.csr.2013.11.010, 2014.
- Green, P. N., and Woodford, S. K.: Preservation studies on some obligatory methanotrophic bacteria, *Lett. Appl. Microbiol.*, 14, 158-162, 1992.
- 830 Günther, F., Overduin, P. P., Yakshina, I. A., Opel, T., Baranskaya, A. V., and Grigoriev, M. N.: Observing Muostakh disappear: permafrost thaw subsidence and erosion of a ground-ice-rich island in response to arctic summer warming and sea ice reduction, *The Cryosphere*, 9, 151–178, 2015.
- 835 Gurevich, E. V.: Influence of air temperature on the river runoff in winter (the Aldan river catchment study), *Russian Meteorol. Hydrol.*, Russian Meteorol. Hydrol., 34, 628–633, 2009.

- 840 Hoefman, S., Van Hoorde, K., Boon, N., Vandamme, P., De Vos, P., and Heylen, K.:
Survival or revival: Long-term preservation induces a reversible viable but non-culturable
state in methane-oxidizing bacteria, *PLoS ONE*, 7, e34196, doi:
10.1371/journal.pone.0034196, 2012.
- 845 Holmes, R. M., McClelland, J. W., Peterson, B. J., Tank, S. E., Bulygina, E., Eglinton, T. I.,
Gordeev, V. V., Gurtovaya, T. Y., Raymond, P. A., and Repeta, D. J.: Seasonal and annual
fluxes of nutrients and organic matter from large rivers to the Arctic Ocean and surrounding
seas, *Estuar. Coast.*, 35, 369-382, 2012.
- 850 Holmes, R. M., Coe, M. T., Fiske, G. J., Gurtovaya, T., McClelland, J. W., Shiklomanov, A.
I., Spencer, R. G. M., S. E. Tank, and Zhulidov, A. V.: Climate change impacts on the
hydrology and biogeochemistry of Arctic Rivers, in: *Global Impacts of Climate Change on
Inland Waters*, edited by: C. R. Goldman, M. K., and R. D. Robarts, Wiley, 2013.
- 855 Janout, M. A., Hölemann, J., Laukert, G., Smirnov, A., Krumpen, T., Bauch, D., and
Timokhov, L.: On the variability of stratification in the freshwater-influenced Laptev Sea
Region, *Front. Mar. Sci.*, 7, 10.3389/fmars.2020.543489, 2020.
- 860 Juhls, B., Stedmon, C. A., Morgenstern, A., Meyer, H., Hölemann, J., Heim, B., Povazhnyi,
V., and Overduin, P. P.: Identifying Drivers of Seasonality in Lena River Biogeochemistry
and Dissolved Organic Matter Fluxes, *Frontiers in Environmental Science*, 8,
10.3389/fenvs.2020.00053, 2020.
- 865 Kallistova, A. Y., Savvichev, A. S., Rusanov, I. I., and Pimenov, N. V.: Thermokarst lakes,
ecosystems with intense microbial processes of the methane cycle, *Microbiology*, 88, 649-
661, 10.1134/S0026261719060043, 2019.
- 870 Kankaala, P., Huotari, J., Peltomaa, E., Saloranta, T., and Ojala, A.: Methanotrophic activity
in relation to methane efflux and total heterotrophic bacterial production in a stratified, humic,
boreal lake, *Limnol. Oceanogr.*, 51, 1195–1204, 2006.
- 875 Kankaala, P., Taipale, S., Nykänen, H., and Jones, R. I.: Oxidation, efflux, and isotopic
fractionation of methane during autumnal turnover in a polyhumic, boreal lake, *J. Geophys.
Res.*, 112, G02033, 10.1029/2006jg000336, 2007.
- 880 Kellogg, C. T. E., McClelland, J. W., Dunton, K. H., and Crump, B. C.: Strong seasonality in
arctic estuarine microbial food webs, *Front. Microbiol.*, 10, 2628, 2019.
- 885 King, G. M., and Adamsen, A. P.: Effects of temperature on methane consumption in a forest
soil and in pure cultures of the methanotroph *Methylobacterium rubrum*, *Appl Environ Microbiol.*,
58, 2758-2763, 1992.
- 890 Kohnert, K., Serafimovich, A., Metzger, S., Hartmann, J., and Sachs, T.: Strong geologic
methane emissions from discontinuous terrestrial permafrost in the Mackenzie Delta, Canada,
Scientific Reports, 7, 5828, 10.1038/s41598-017-05783-2, 2017.
- 895 Kohnert, K., Juhls, B., Muster, S., Antonova, S., Serafimovich, A., Metzger, S., Hartmann, J.,
and Sachs, T.: Towards understanding the contribution of waterbodies to the methane
emissions of a permafrost landscape on a regional scale – a case study from the Mackenzie
Delta, Canada, *Global Change Biol.*, 24, 3976– 3989, doi: 10.1111/gcb.14289, 2018.

- 890 Kruse, S., Bolshiyarov, D., Grigoriev, M. N., Morgenstern, A., Pestryakova, L., Tsibizov, L.,
and Udke, A.: Russian-German Cooperation: Expeditions to Siberia in 2018, Alfred Wegener
Institute for Polar and Marine Research, Bremerhaven, 257, 2019.
- 895 Lantuit, H., Atkinson, D., Overduin, P. P., Grigoriev, M., Rachold, V., Grosse, G., and
Hubberten, H.-W.: Coastal erosion dynamics on the permafrost-dominated Bykovsky
Peninsula, north Siberia, 1951-2006, 2011, 30, 7341, DOI: 10.3402/polar.v30i0.7341, 2011.
Leppäranta, M.: Freezing of lakes and the evolution of their ice cover, Springer, Berlin
Heidelberg, 2015.
- 900 Lininger, K. B., and Wohl, E.: Floodplain dynamics in North American permafrost regions
under a warming climate and implications for organic carbon stocks: A review and synthesis,
Earth-Sci. Rev., 193, 24-44, <https://doi.org/10.1016/j.earscirev.2019.02.024>, 2019.
- 905 Lofton, D., Whalen, S., and Hershey, A.: Effect of temperature on methane dynamics and
evaluation of methane oxidation kinetics in shallow Arctic Alaskan lakes, Hydrobiologia,
721, 209-222, doi:10.1007/s10750-013-1663-x, 2014.
- 910 Magen, C., Lapham, L. L., Pohlman, J. W., Marshall, K., Bosman, S., Casso, M., and
Chanton, J. P.: A simple headspace equilibration method for measuring dissolved methane,
Limnol. Oceanogr.: Methods, 12, 637-650, doi:10.4319/lom.2014.12.637, 2014.
- Martinez-Cruz, K., Sepulveda-Jauregui, A., Walter Anthony, K., and Thalasso, F.:
Geographic and seasonal variation of dissolved methane and aerobic methane oxidation in
Alaskan lakes, Biogeosciences, 12, 4595-4606, 10.5194/bg-12-4595-2015, 2015.
- 915 Martinez-Cruz, K., Leewis, M. C., Herriott, I. C., Sepulveda-Jauregui, A., Anthony, K. W.,
Thalasso, F., and Leigh, M. B.: Anaerobic oxidation of methane by aerobic methanotrophs in
sub-Arctic lake sediments, Sci Total Environ, 607-608, 23-31,
10.1016/j.scitotenv.2017.06.187, 2017.
- 920 Mau, S., Römer, M., Torres, M. E., Bussmann, I., Pape, T., Damm, E., Geprägs, P.,
Wintersteller, P., Hsu, C.-W., Loher, M., and Bohrmann, G.: Methane data and stable carbon
isotopic composition of methane along the continental margin off Svalbard. In: PANGAEA,
2017a.
- 925 Mau, S., Römer, M., Torres, M. E., Bussmann, I., Pape, T., Damm, E., Geprägs, P.,
Wintersteller, P., Hsu, C. W., Loher, M., and Bohrmann, G.: Widespread methane seepage
along the continental margin off Svalbard - from Bjørnøya to Kongsfjorden, Scientific
Reports, 7, 42997, 10.1038/srep42997, 2017b.
- 930 Morozumi, T., Shingubara, R., Murase, J., Nagai, S., Kobayashi, H., Takano, S., Tei, S., Fan,
R., Maximov, T. C., and Sugimoto, A.: Usability of water surface reflectance for the
determination of riverine dissolved methane during extreme flooding in northeastern Siberia,
Polar Science, <https://doi.org/10.1016/j.polar.2019.01.005>, 2019.
- 935 Newton, A. M. W., and Mullan, D.: Climate change and Northern Hemisphere lake and river
ice phenology, The Cryosphere Discuss., 2020, 1-60, 10.5194/tc-2020-172, 2020.

- O'Reilly, C. M., Sharma, S., Gray, D. K., Hampton, S. E., and Read, J. S.: Rapid and highly variable warming of lake surface waters around the globe, *Geophys. Res. Lett.*, 42, 1–9, <https://doi.org/10.1002/2015GL066235>, 2015.
- 940
- Osudar, R., Liebner, S., Alawi, M., Yang, S., Bussmann, I., and Wagner, D.: Methane turnover and methanotrophic communities in arctic aquatic ecosystems of the Lena Delta, Northeast Siberia, *FEMS Microbiol Ecol*, 92, doi: 10.1093/femsec/fiw116, 2016.
- 945
- Overduin, P., Wetterich, S., Günther, F., Grigoriev, M. N., Grosse, G., Schirrmeister, L., Hubberten, H. W., and Makarov, A.: Coastal dynamics and submarine permafrost in shallow water of the central Laptev Sea, East Siberia, *Cryosphere Discussions*, 9, 3741-3775, 10.5194/tcd-9-3741-2015, 2015.
- 950
- Overduin, P., Blender, F., Bolshiyarov, D. Y., Grigoriev, M. N., Morgenstern, A., and Meyer, H.: Russian-German Cooperation: Expeditions to Siberia in 2016, Alfred Wegener Institute for Polar and Marine Research, Bremerhaven, 295, 2017.
- 955
- Peterson, B. J., Holmes, R. M., McClelland, J. W., Vörösmarty, C. J., Lammers, R. B., Shiklomanov, A. I., Shiklomanov, I. A., and Rahmstorf, S.: Increasing river discharge to the Arctic Ocean, *Science*, 298, 2171-2173, 10.1126/science.1077445, 2002.
- Platt, S. M., Eckhardt, S., Ferré, B., Fisher, R. E., Hermansen, O., Jansson, P., Lowry, D., Nisbet, E. G., Pisso, I., Schmidbauer, N., Silyakova, A., Stohl, A., Svendby, T. M., Vadakkepuliambatta, S., Mienert, J., and Lund Myhre, C.: Methane at Svalbard and over the European Arctic Ocean, *Atmos. Chem. Phys.*, 18, 17207-17224, 10.5194/acp-18-17207-2018, 2018.
- 960
- Prowse, T., Alfredsen, K., Beltaos, S., Bonsal, B., Duguay, C., Korhola, A., McNamara, J., Pienitz, R., Vincent, W. F., Vuglinsky, V., and Weyhenmeyer, G. A.: Past and future changes in Arctic lake and river ice, *Ambio*, 40, 53-62, doi: 10.1007/s13280-011-0216-7, 2011.
- 965
- Raven, J. A., and Geider, R. J.: Temperature and algal growth, *New Phytol.*, 110, 441-461, 10.1111/j.1469-8137.1988.tb00282.x, 1988.
- 970
- Ricão Canelhas, M., Denfeld, B. A., Weyhenmeyer, G. A., Bastviken, D., and Bertilsson, S.: Methane oxidation at the water-ice interface of an ice-covered lake, *Limnol. Oceanogr.*, 61, S78–S79, doi: 10.1002/lno.10288, 2016.
- 975
- Rivera, J., Karabanov, E. B., Williams, D. F., Buchinskyi, V., and Kuzmin, M.: Lena River discharge events in sediments of Laptev Sea, Russian Arctic, *Estuar. Coast. Shelf Sci.*, 66, 185-196, 10.1016/j.ecss.2005.08.009, 2006.
- 980
- Santibáñez, P. A., Michaud, A. B., Vick-Majors, T. J., D'Andrilli, J., Chiuchiolo, A., Hand, K. P., and Priscu, J. C.: Differential incorporation of bacteria, organic matter, and inorganic ions into lake ice during ice formation, *Journal of Geophysical Research: Biogeosciences*, 124, 585-600, doi: 10.1029/2018JG004825, 2019.
- 985
- Saunois, M., Bousquet, P., Poulter, B., Peregón, A., Ciais, P., Canadell, J. G., Dlugokencky, E. J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F. N., Castaldi, S., Jackson, R. B., Alexe, M., Arora, V. K., Beerling, D. J., Bergamaschi, P., Blake, D. R., Brailsford, G., Brovkin, V., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Curry,

- 990 C., Frankenberg, C., Gedney, N., Höglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H. S., Kleinen, T., Krummel, P., Lamarque, J. F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., McDonald, K. C., Marshall, J., Melton, J. R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F. J. W., Patra, P. K., Peng, C., Peng, S., Peters, G. P., Pison, I., Prigent, C., Prinn, R., Ramonet, M., Riley, W. J., Saito, M., Santini, M., Schroeder, R., Simpson, I. J., Spahni, R., Steele, P., Takizawa, A., Thornton, B. F., Tian, H., Tohjima, Y., Viovy, N.,
- 995 Voulgarakis, A., van Weele, M., van der Werf, G. R., Weiss, R., Wiedinmyer, C., Wilton, D. J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X., Yoshida, Y., Zhang, B., Zhang, Z., and Zhu, Q.: The global methane budget 2000–2012, *Earth Syst. Sci. Data*, 8, 697-751, 10.5194/essd-8-697-2016, 2016.
- 1000 Saunio, M., Stavert, A. R., Poulter, B., Bousquet, P., Canadell, J. G., Jackson, R. B., Raymond, P. A., Dlugokencky, E. J., Houweling, S., Patra, P. K., Ciais, P., Arora, V. K., Bastviken, D., Bergamaschi, P., Blake, D. R., Brailsford, G., Bruhwiler, L., Carlson, K. M., Carrol, M., Castaldi, S., Chandra, N., Crevoisier, C., Crill, P. M., Covey, K., Curry, C. L., Etiope, G., Frankenberg, C., Gedney, N., Hegglin, M. I., Höglund-Isaksson, L., Hugelius, G.,
- 1005 Ishizawa, M., Ito, A., Janssens-Maenhout, G., Jensen, K. M., Joos, F., Kleinen, T., Krummel, P. B., Langenfelds, R. L., Laruelle, G. G., Liu, L., Machida, T., Maksyutov, S., McDonald, K. C., McNorton, J., Miller, P. A., Melton, J. R., Morino, I., Müller, J., Murguia-Flores, F., Naik, V., Niwa, Y., Noce, S., O'Doherty, S., Parker, R. J., Peng, C., Peng, S., Peters, G. P., Prigent, C., Prinn, R., Ramonet, M., Regnier, P., Riley, W. J., Rosentreter, J. A., Segers, A., Simpson, I. J., Shi, H., Smith, S. J., Steele, L. P., Thornton, B. F., Tian, H., Tohjima, Y., Tubiello, F. N.,
- 1010 Tsuruta, A., Viovy, N., Voulgarakis, A., Weber, T. S., van Weele, M., van der Werf, G. R., Weiss, R. F., Worthy, D., Wunch, D., Yin, Y., Yoshida, Y., Zhang, W., Zhang, Z., Zhao, Y., Zheng, B., Zhu, Q., Zhu, Q., and Zhuang, Q.: The Global Methane Budget 2000–2017, *Earth Syst. Sci. Data*, 12, 1561-1623, 10.5194/essd-12-1561-2020, 2020.
- 1015 Schleusner, P., Biskaborn, B. K., Kienast, F., Wolter, J., Subetto, D., and Diekmann, B.: Basin evolution and palaeoenvironmental variability of the thermokarst lake E l'gene-K yuele, Arctic Siberia., *Boreas*, 44, 2016-2229, 2015.
- 1020 Sepulveda-Jauregui, A., Walter Anthony, K. M., Martinez-Cruz, K., Greene, S., and Thalasso, F.: Methane and carbon dioxide emissions from 40 lakes along a north-south latitudinal transect in Alaska, *Biogeosciences*, 12, 3197-3223, 10.5194/bg-12-3197-2015, 2015.
- 1025 Serikova, S., Pokrovsky, O. S., Laudon, H., Krickov, I. V., Lim, A. G., Manasypov, R. M., and Karlsson, J.: High carbon emissions from thermokarst lakes of Western Siberia, *Nature Communications*, 10, 10.1038/s41467-019-09592-1, 2019.
- Shiklomanov, A. I., and Lammers, R. B.: River ice responses to a warming Arctic—Recent evidence from Russian rivers, *Environ. Res. Lett.*, 9, 035008, doi:10.1088/1748-9326/9/3/035008., 2014.
- 1030 Shiklomanov, A. I., Holmes, R. M., McClelland, J. W., Tank, S. E., and Spencer, R. G. M.: Arctic Great Rivers Observatory. Discharge Dataset, Version 20191112., 2018.
- Spangenberg, I.: Methane distribution and hydrochemistry in lake and sea ice from a region of thawing permafrost, Siberia, Master, University of Potsdam, 2018.
- 1035 Spangenberg, I., Overduin, P. P., Damm, E., Bussmann, I., Liebner, S., Meyer, H., Angelopoulos, M., Biskaborn, B. K., Grigoriev, M. N., and Grosse, G.: Methane pathways in winter ice of thermokarst lakes, lagoons and coastal waters in North Siberia, in revision.

- 1040 Steinle, L., Maltby, J., Treude, T., Kock, A., Bange, H. W., Engbersen, N., Zopfi, J., Lehmann, M. F., and Niemann, H.: Effects of low oxygen concentrations on aerobic methane oxidation in seasonally hypoxic coastal waters, *Biogeosciences*, 14, 1631-1645, 10.5194/bg-14-1631-2017, 2017.
- 1045 Strauss, J., Boike, J., Bolshiyarov, D. Y., Grigoriev, M. N., El-Hajj, H., Morgenstern, A., Overduin, P. P., and Udke, A.: Russian-German Cooperation: Expeditions to Siberia in 2017, Alfred Wegener Institute for Polar and Marine Research, Bremerhaven, 296 p, 2018.
Tan, Z., and Zhuang, Q.: Arctic lakes are continuous methane sources to the atmosphere under warming conditions. , *Environmental Research Letters*, 10, 1-9, <https://doi.org/10.1088/1748-9326/10/5/054016>, 2015.
- 1050 Tananaev, N. I., Makarieva, O. M., and Lebedeva, L. S.: Trends in annual and extreme flows in the Lena River basin, Northern Eurasia, *Geophys. Res. Lett.*, 43, 764-772, 10.1002/2016GL070796, 2016.
- 1055 Thibodeau, B., Bauch, D., Kassens, H., and Timokhov, L. A.: Interannual variations in river water content and distribution over the Laptev Sea between 2007 and 2011: The Arctic Dipole connection, *Geophys. Res. Lett.*, 41, 7237-7244, 10.1002/2014GL061814, 2014.
- 1060 Thottathil, S. D., Reis, P. C. J., and Prairie, Y. T.: Methane oxidation kinetics in northern freshwater lakes, *Biogeochem.*, 143, 105-116, 10.1007/s10533-019-00552-x, 2019.
- 1065 Townsend-Small, A., Åkerström, F., Arp, C. D., and Hinkel, K. M.: Spatial and temporal variation in methane concentrations, fluxes, and sources in lakes in Arctic Alaska, *Journal of Geophysical Research: Biogeosciences*, 122, 2966-2981, doi: 10.1002/2017JG004002, 2017.
Triputra, P. W.: Greenhouse gases in Siberian ice cores, Master thesis, Faculty of Science, Center of Environmental Sciences., Eötvös Loránd University, Budapest, 2018.
- 1070 Utsumi, M., Nojiri, Y., Nakamura, T., Nozawa, T., Otsuki, A., Takamura, N., Watanabe, M., and Seki, H.: Dynamic of dissolved methane and methane oxidation in dimictic Lake Nojiri during winter, *Limnol. Oceanogr.*, 43, 10-17, 1998b.
- 1075 Vachon, D., Langenegger, T., Donis, D., and McGinnis, D. F.: Influence of water column stratification and mixing patterns on the fate of methane produced in deep sediments of a small eutrophic lake, <https://doi.org/10.1002/lno.11172>, 2019.
- 1080 Wählström, I., Dieterich, C., Pemberton, P., and Meier, H. E. M.: Impact of increasing inflow of warm Atlantic water on the sea-air exchange of carbon dioxide and methane in the Laptev Sea, *Journal of Geophysical Research G: Biogeosciences*, 121, 1867-1883, 10.1002/2015JG003307, 2016.
- 1085 Walter Anthony, K., Daanen, R., Anthony, P., Schneider von Deimling, T., Ping, C.-L., Chanton, J. P., and Grosse, G.: Methane emissions proportional to permafrost carbon thawed in Arctic lakes since the 1950s, *Nature Geosci*, advance online publication, 10.1038/ngeo2795 <http://www.nature.com/ngeo/journal/vaop/ncurrent/abs/ngeo2795.html#supplementary-information>, 2016.
- 1090 Walter, K. M., Smith, L. C., and Stuart Chapin, F.: Methane bubbling from northern lakes: present and future contributions to the global methane budget, *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 365, 657-1676.

- , <https://doi.org/10.1098/rsta.2007.2036>, 2007.
- 1095 Wegner, C., Bauch, D., Hölemann, J. A., Janout, M. A., Heim, B., Novikhin, A., Kassens, H., and Timokhov, L.: Interannual variability of surface and bottom sediment transport on the Laptev Sea shelf during summer, *Biogeosciences*, 10, 1117-1129, 10.5194/bg-10-1117-2013, 2013.
- 1100 Wik, M., Johnson, J. E., Crill, P. M., DeStasio, J. P., Erickson, L., Halloran, M. J., Fahnestock, M. F., Crawford, M. K., Phillips, S. C., and Varner, R. K.: Sediment characteristics and methane ebullition in three subarctic lakes, *Journal of Geophysical Research: Biogeosciences*, 123, 2399-2411, doi: 10.1029/2017JG004298, 2018.
- 1105 Wild, B., Andersson, A., Bröder, L., Vonk, J., Hugelius, G., McClelland, J. W., Song, W., Raymond, P. A., and Gustafsson, Ö.: Rivers across the Siberian Arctic unearth the patterns of carbon release from thawing permafrost, *Proceedings of the National Academy of Sciences*, 116, 10280, 10.1073/pnas.1811797116, 2019.
- 1110 Wilson, S. L., Frazer, C., Cumming, B. F., Nuin, P. A. S., and Walker, V. K.: Cross-tolerance between osmotic and freeze-thaw stress in microbial assemblages from temperate lakes, *FEMS Microbiol. Ecol.*, 82, 405-415, 10.1111/j.1574-6941.2012.01404.x, 2012.
- 1115 Winkel, M., Mitzscherling, J., Overduin, P. P., Horn, F., Winterfeld, M., Rijkers, R., Grigoriev, M. N., Knoblauch, C., Mangelsdorf, K., Wagner, D., and Liebner, S.: Anaerobic methanotrophic communities thrive in deep submarine permafrost, *Scientific Reports*, 8, 10.1038/s41598-018-19505-9, 2018.
- 1120 Winterfeld, M., Goñi, M. A., Just, J., Hefter, J., and Mollenhauer, G.: Characterization of particulate organic matter in the Lena River delta and adjacent nearshore zone, NE Siberia – Part 2: Lignin-derived phenol compositions, *Biogeosciences*, 12, 2261-2283,, doi.org/10.5194/bg-12-2261-2015, 2015.
- 1125 Yang, D., Kane, D. L., Hinzman, L. D., Zhang, X., Zhang, T., and Ye, H.: Siberian Lena River hydrologic regime and recent change, *J. Geophys. Res.*, 107, 4694, 10.1029/2002jd002542, 2002.

