Seasonal methane dynamics in three different Siberian water bodies

5   Ingeborg Bussmann[1], Irina Fedorova[2], Bennet Juhls[3], Pier Paul Overduin[4], Matthias Winkel[5]

[1]Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research - Helgoland,
Germany

[2]St. Petersburg University, Russia

10   [3] Institute for Space Sciences, Department of Earth Sciences, Freie Universität Berlin, Berlin,
Germany

[4]Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research - Potsdam,
Germany

[5]German Research Centre for Geosciences, 3.5 Interface Geochemistry Group - Potsdam,

15   Germany

Correspondence to: Ingeborg Bussmann (ingeborg.bussmann@awi.de)

Abstract

20   Arctic regions and their water bodies are being affected by the most rapid climate warming on
Earth. Arctic lakes and small ponds are known to act as an important source of atmospheric
methane. 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

25   water bodies (Lena River, Tiksi Bay and Lake Golzovoye, Siberia, Russia) over a period of
two years. 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 was also investigated.

In the Lena River winter methane concentrations were a quarter of the summer concentrations

30   (8 vs 31 nmol L^{-1}) and mean winter MOX rate was low (0.023 nmol L^{-1} d^{-1}). In contrast, Tiksi
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

35   bay. The temperature had a strong influence on the MOX, (Q10 = 2.72 ± 0.69) compared to
temperate environments. 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-

40   times higher methane concentration in the water column.
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. Methane oxidation rate was not able to
counteract this winter time accumulation.

45

1 Introduction
Worldwide, the mixing ratio of methane (methane) has been increasing rapidly since 2005
(Hartmann et al., 2013; IPCC, 2013). The reasons for the observed increases in atmospheric
methane are unclear. A probable explanation is increased methane emissions from wetlands,

50   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). In the Arctic, mean atmospheric methane mixing
ratio increased by 6 ppb per year from 2001 to 2017 (Platt et al., 2018). Especially the Laptev
Sea, in eastern Siberia, is generally a source of methane to the atmosphere, with a great

55   interannual variability of sea-air fluxes of methane (Wåhlström 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., 2016).
        Lakes are important sources of atmospheric methane on a regional to global scale

60   (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
atmosphere via ebullition (Bastviken et al., 2004; Walter et al., 2007), a temperature-sensitive
transport mode with high spatial and temporal heterogeneity (DelSontro et al., 2015). The role

65   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).

One major drawback from most of these studies is that sampling was conducted in the
70 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
external oxygen supply, enhanced anaerobic degradation leads to accumulation of methane,
75 H_2S and NH_3 (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). 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
80 known for lakes with incomplete spring mixing and consequently a maximum gas exchange
in autumn during complete mixing (Deshpande et al., 2015).

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
85 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 in
90 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
95 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
100 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
105   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
110   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 in situ
sink.

115   ## 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; Schleusner et al., 2015).

120   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 June
125   (Angelopoulos et al., 2019). Due to its isolation behind Muostakh Island and Cape Muostakh,
sea ice tends to be preserved longer than in the Central Laptev Sea (Günther et al., 2015.).

        The Lena River has a mean annual discharge of 581 km^3/year. It is the 2nd largest
Arctic river by annual discharge and the 6th largest globally. There are no dams on the main-
stem of the Lena, but there is a dam on the Viluy River, one of the Lena's main tributaries
130   (Holmes et al., 2013). The Lena's watershed is 2.6 million km^2, of which 70.5% is underlain
by continuous permafrost (Juhls et al., in review). 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-
Khaya Bay through the northern and eastern part of the delta and through he Bykovskaya
135   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).

140

3 Material and Methods

3.1 Study sites

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).

145   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 diameter with a maximal depth of 10 m, surrounded by Yedoma uplands at various stages of

150   degradation and with no ice grounding in its center (Spangenberg, 2018; Strauss et al., 2018). 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

155   same locations (Lena River near Samoylov, Tiksi Bay and Lake Golzovoye) with open water (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).

160   *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

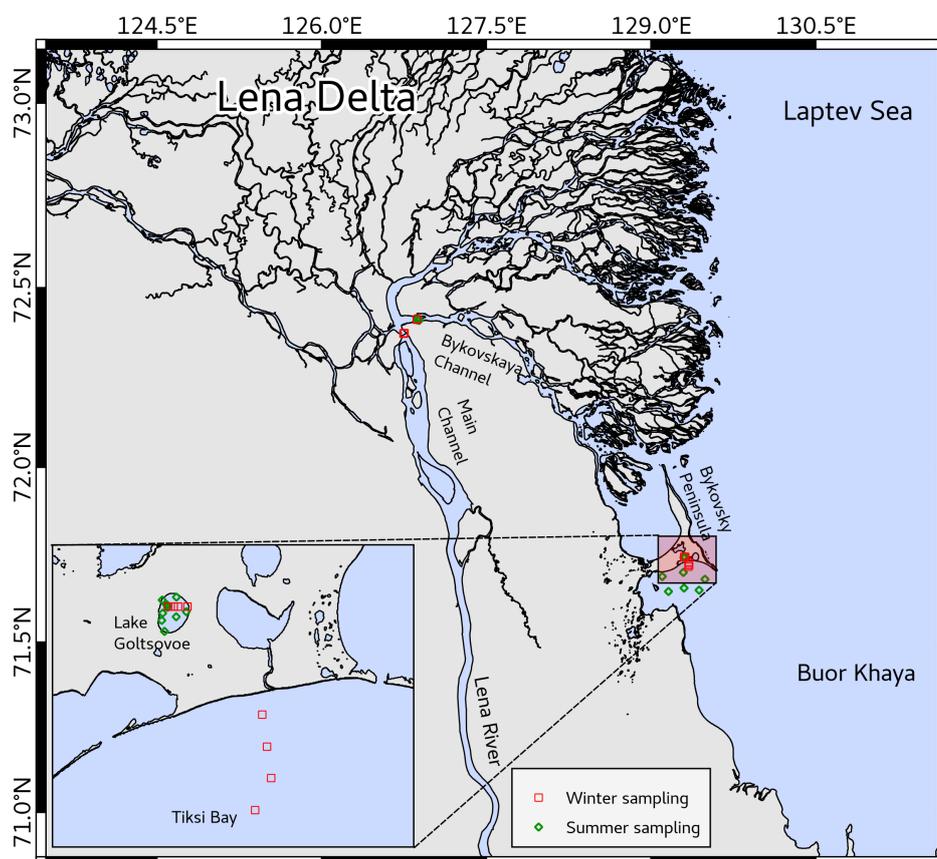



Figure 1: Map of the study area and sampling locations (winter sampling locations in red, summer sampling locations in green).

3.2 Water sampling for methane analysis

In winter, water samples from below the ice 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% H_2SO_4 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.

175 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 .

180 ### 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

185 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

190 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 H_2SO_4) for analysis of methane concentration (Strauss et al., 2018).

3.4 Methane analysis
195 Methane concentrations were determined via head-space method by adding 20 ml of N_2 as headspace. Headspace methane concentrations were analyzed in the home laboratory with a gas chromatograph (GC 2014, Shimadzu) equipped with a flame ionization detector and a Shinycarbon column (Restek, USA). The temperatures of the oven, the injector, and detector were 100, 120, and 160 °C, respectively. The carrier gas (N_2) flow was 20 mL min^{-1}, with 40

200 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%.

205 ### 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

210   multiplied with the in-situ methane concentration. Radioactive, tritiated methane (0.1 ml of

^{3}H-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

ml of 25% H$_2$SO$_4$. Abiotic controls were poisoned before adding the tracer. Radioactivity was

determined with a liquid scintillation counter (Triathler, Finnland) and Ultima Gold (Perkin

215   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;

4; 7 and 10 °C show the temperature curve of the MOX reaction. At a previous cruise off

220   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

process according to (Raven and Geider, 1988):

225
$$Q10 = \exp(-10 * \frac{m}{Tis^2})$$

where Tis is the in-situ temperature and "m" the slope of the regression line of the Arrhenius

plot (the inverse of the absolute temperature vs. the natural logarithm of the MOX rate).

230   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

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

235   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).

4 Results

4.1 Methane concentrations and hydrochemistry

In the Lena River in summer 2016, median methane concentrations were 37 nM (n = 21) in 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 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 µS 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 µS 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 µS 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., (in review).

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

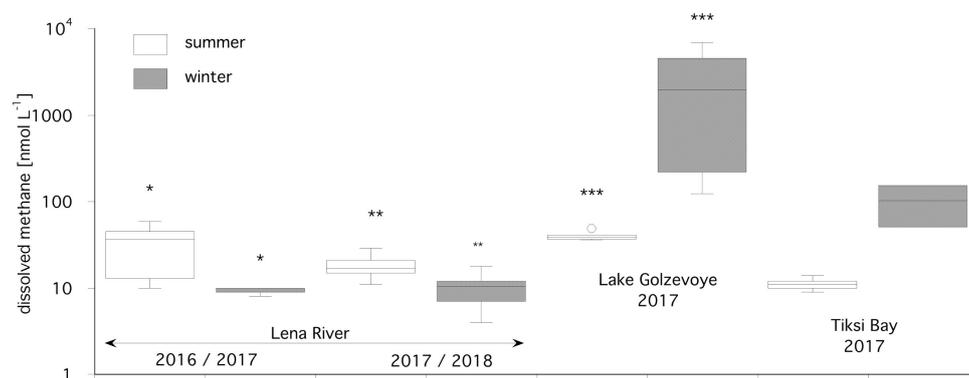
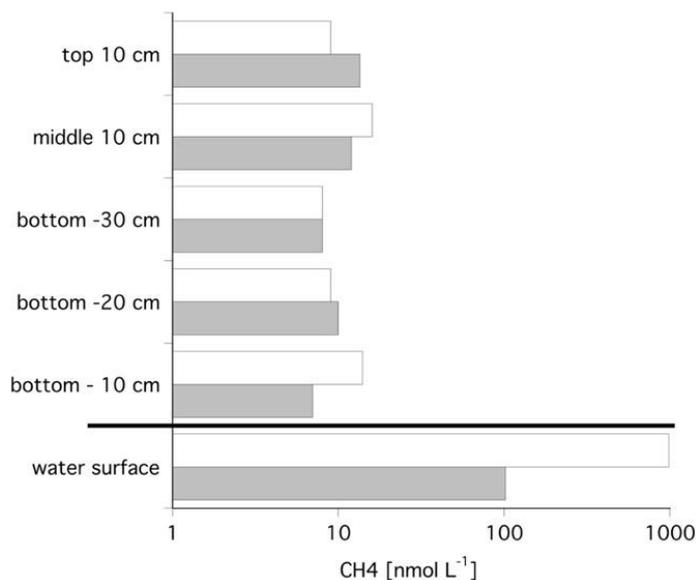
275   (n = 2). Due to low sample numbers, no statistical test was possible. In summer 2017, water temperature ranged from 4.3 – 10.9 °C, electric conductivity from 20 – 6200 µS cm[-1]. In winter, bottom water had a temperature of 3.3 °C and 0.3 °C at the top 1 m, with a thermocline at 1 – 2 meters.

280

Figure 2: Median summer and winter methane concentrations at the Lena River, Lake Golzovoye 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.

285   **4.2 Ice cores**
Ice cores were taken at Lake Golzovoye 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 Golzovoye and Tiksi Bay were rather low (both with a median of 9 nM). No depth gradients from the ice surface, middles section and the three lower most sections were

290   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 Golzovoye, 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 11times and 109times higher than in the ice cores, for Tiksi Bay and Lake Golzovoye, respectively, with a median

295   of 102 nM and 985 nM (Bussmann and Fedorova, 2019). However, in one core of Lake Golzovoye (core #24), concentrations were orders of magnitude higher throughout the core (854 – 11091 nM) and 6954 nM in the water below (Figure 4).

300 *Figure 3: Median methane concentrations in the ice cores and in the water below the ice; for Lake Golzovoye (without core #24) in white and Tiksi Bay in grey. Note the logarithmic scale.*

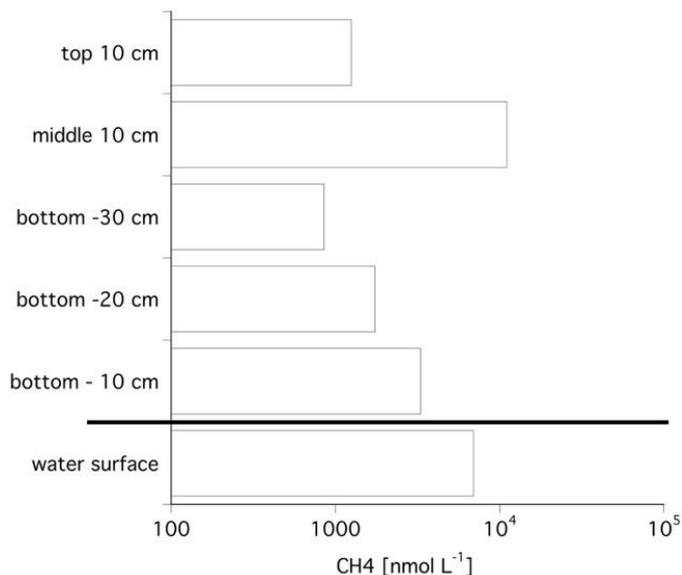



Figure 4: Methane concentrations in the ice core #24 and in the water below the ice in Lake Golzovoye. Note the logarithmic
305 *scale.*

4.3 Methane oxidation rates (MOX)

Methane oxidation rates were determined in the melted water from the ice cores and in water

from the Lena River. Due to logistic restraints at the field sites, no direct measurements of

310   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

315   could be detected (on average 0.011 nmol L^{-1} 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
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
from Lena River water samples with the respective in-situ methane concentration of the water

320   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 nmol L^{-1} d^{-1}), where also very high
methane concentrations were observed (Figure 5). The median MOX in the other water
samples of Lake Golzovoye was about one order of magnitude lower (2.95 nmol L^{-1} d^{-1}). In
Tiksi Bay and in the Lena River MOX rates decreased by further orders of magnitude

325   (median of 0.305 and 0.023 nmol L^{-1} d^{-1}, Figure 5).

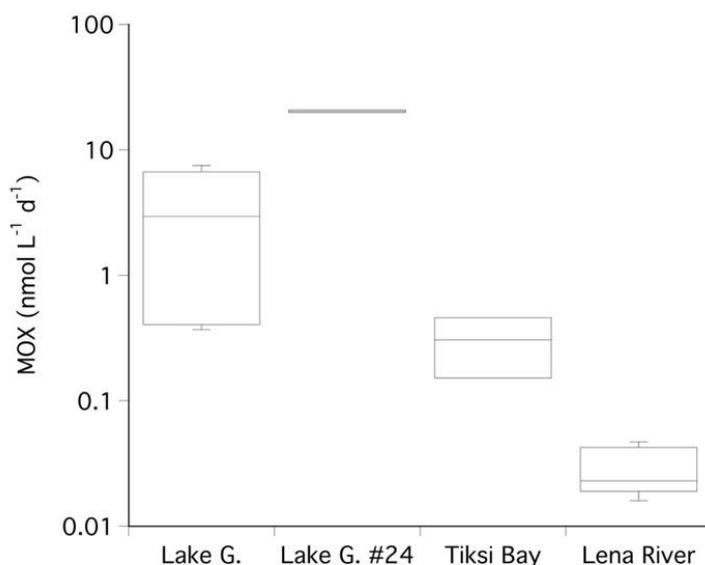



Figure 5: Box plot of the methane oxidation rates in water under ice cover at Lake Golzovoye, at the location of ice core #24, Tiksi Bay and the Lena River. Note the logarithmic scale.

330

4.4 Temperature influence on MOX

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 6). The Q10 calculated for these water samples from the

335   Lena River was 2.72 ± 0.69. For polar and marine waters off Svalbard a Q10 of 2.99 ± 0.86 was calculated.

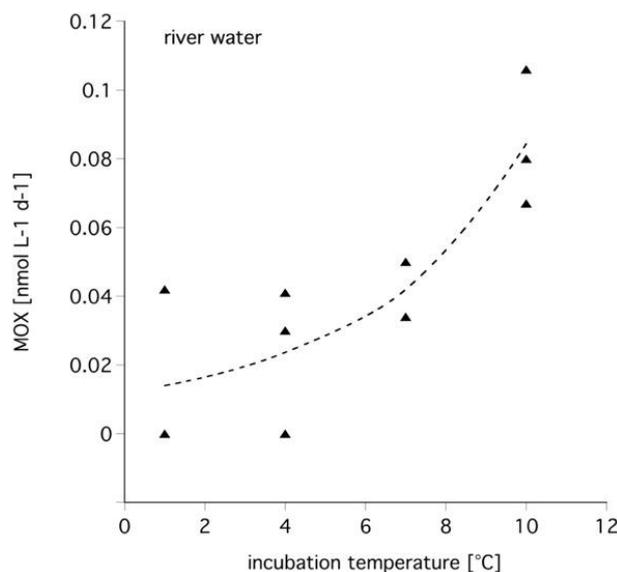



Figure 6: Influence of incubation temperature on methane oxidation rate for Lena River water, winter 2017.

340

5 Discussion

5.1 Methane concentrations and hydrochemistry

In this study we compared the methane concentration under ice cover (winter) with open

345   water situations (summer) in three different water bodies.

In winter the methane concentrations in the Lena River were 4times lower as in 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 m^3 sec^{-1}) was about 10-times lower than in summer (July and August with 29.800 and

350   26.000 m^3 sec^{-1}, (Shiklomanov et al., 2018).

In winter there are only a few possible sources of methane left. The surrounding soils 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
355 production. Especially in the Lena River a substantial amount of POC originates from
thermokarst-induced, abrupt collapse of Pleistocene Ice Complex deposits. These events
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
360 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
365 information yet available.

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
370 on Tiksi Bay will lead to a decrease in tidal amplitudes and velocities (Fofonova et al., 2014).
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) by
further degradation of organic material. However, as aerobic methane oxidation in the water
375 column is impaired by low temperatures, methane concentrations in water increase.

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
380 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
385 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 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

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

5.2 Ice cores

395   The median methane concentration of all ice cores for Lake Golzovoye and Tiksi Bay was 9 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

400   means that, in terms of methane, a complete separation of the water body from the 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

405   methane release to the atmosphere.

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

410   freshwater lakes reduce the total bacterial cell number at least 100,000-fold. In addition, methanotrophic bacteria are particular 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

415   other biological processes in the bottom layer of the ice cores. Thus, we conclude that the 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)

420   In this study we determined the *in situ* methane oxidation rate with tritiated methane as tracer. The 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 *in situ* rates. The fractional turnover rate k' was determined only in the ice cores and in river water; within these k' was evenly distributed. Therefore, this factor was applied to all water samples, i.e. even to the

425   brackish water samples from Tiksi Bay.

Our data span three orders of magnitude, ranging from 0.02 nmol L^{-1} d^{-1} in the Lena River, 0.31 in Tiksi Bay and 2.95 nmol L^{-1} d^{-1} in Lake Golzovoye. Another polar study finds a MOX of 0.004–1.09 mg C m^{-3} d^{-1} (= 0.33 – 91 nmol L^{-1} d^{-1}) at the water-ice interface of a Swedish lake (Ricão Canelhas et al., 2016). Also (Bastviken et al., 2002) report a MOX of

430   similar range (0.001–39 mg C m^{-3} d^{-1} = 0.08 – 3250 nmol L^{-1} $d^{-1)}$ 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 – 0.28 mg methane L^{-1} d^{-1} = 1875 – 17500 nmol L^{-1} d^{-1}) with a kinetic approach to determine MOX (Martinez-Cruz et al., 2015). In marine polar waters off Svalbard MOX was determined with the same tracer method and

435   ranged from 1.6 – 2.2 nmol L^{-1} 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 same method as in this study. Therefore, we can approach a seasonal comparison (winter vs.

440   summer), assuming interannual variability is negligible. 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 nmol L^{-1} d^{-1} (n = 8), the summer MOX was about 3 orders of magnitude higher than in winter (0.023 nmol L^{-1} d^{-1}). For MOX in lakes, summer rates from small lakes near Research Station Samoylov Island were 36-

445   times higher (median 107 nmol L^{-1} d^{-1}, n = 6) than winter rates (2.95 nmol L^{-1} 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). 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

450   nmol L^{-1} d^{-1}, Table 2).

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

Location	[CH₄]winter / [CH₄]summer*	Under ice accumulation**	Water velocity winter	[MOX] winter	[MOX]summer*** / [MOX]winter
Lena River	S > W, x 5	n.a§	strong	low	S > W, x 991
Tiksi Bay	W > S, x 7	Wa > I, x 11	medium	medium	S = W
Lake Golzovoye	W > S, x 40	Wa > I, x 109	minor	high	S > W, x 36

§ n.a. not available

455 *comparing dissolved methane concentrations in water in winter and summer

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

*** winter data from this study, summer data from (Osudar et al., 2016) and (Bussmann et al., 2017)

460   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

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

    But 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

470 (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 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

475 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* 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

480   and 1.75, respectively (Figure 7). Although the substrate concentration in the temperate

waters was higher, MOX from polar regions seems to react more sensitively to a temperature

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

485   increases while methanotrophic bacteria from temperate waters are nearer to their optimum

temperature and do not react as sensitively.

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

490   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

communities and food webs structure change based on carbon availability (Kellogg et al.,

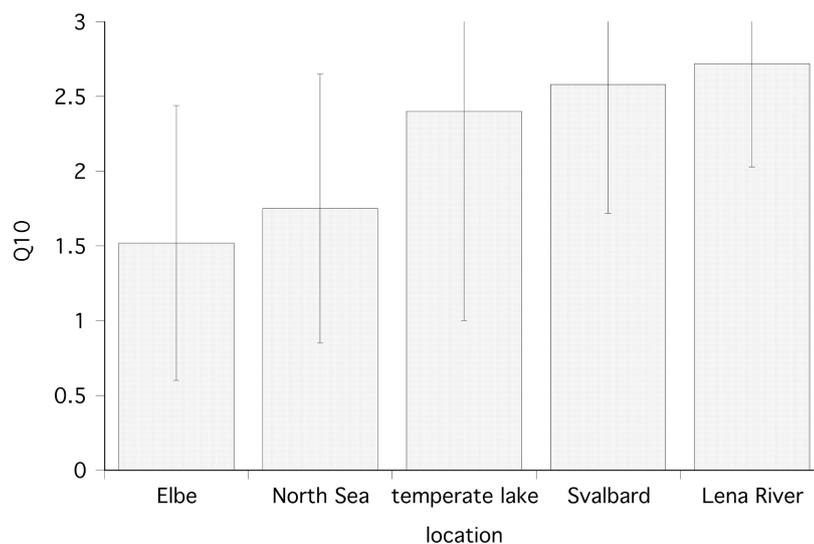
2019).

495

Figure 7: Temperature dependence (Q10) 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).

500   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.

505 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 in/decrease in a linear way. Thus, the duration of ice cover is important and

510 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

515 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).

520

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

525 annual cycle, following the ice-covered and summer periods. Other processes during autumn mixing, ice-on, ice off are not considered.

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

530 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

535 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
540   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
545   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. Thus, an extended time of ice-free conditions could reduce the methane flux from the Arctic water
550   bodies.

Data availability:

Data on methane concentration and MOX will be submitted to the Pangea database.
555

Acknowledgements:

This study was part of the Helmholtz program PACES, Topic 1.3. We are thankful to the logistics department of the Alfred Wegener Institute, particularly Waldemar Schneider.
560   Logistical support for the fieldwork was provided by the Russian Hydrographic Service (Hydrobase Tiksi). We acknowledge support by the Open Access Publication Funds of Alfred-Wegener-Institut Helmholtz-Zentrum für Polar- und Meeresforschung.

565

References

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
570   2019.

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 nethanotroph *Methylovulum psychrotolerans*, Front. Microbiol., doi: 10, 589, 2019.

575

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.

Bastviken, D., Cole, J., Pace, M., and Tranvik, L.: Methane emissions from lakes:
580   Dependence of lake characteristics, two regional assessments, and a global estimate, Glob. Biogeochem. Cycl., 18, GB4009, doi:10.1029/2004GB002238, 2004.

Bednařík, A., Blaser, M., Matoušů, A., Tušer, M., Chaudhary, P. P., Šimek, K., and Rulík, M.: Sediment methane dynamics along the Elbe River, Limnologica, 79, 125716, 2019.

585

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:
590   10.1016/j.quascirev.2015.08.014, 2016.

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.

595   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.

Bussmann, I., Hackbusch, S., Schaal, P., and Wichels, A.: Methane distribution and oxidation
600   around the Lena Delta in summer 2013, Biogeosciences, 14, 4985-5002, doi.org/10.5194/bg-14-4985-2017, 2017.

Bussmann, I., and Fedorova, I.: Dissolved methane concentrations under ice cover in the Lena Delta area., 2019. https://doi.pangaea.de/10.1594/PANGAEA.905776
605

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,
610   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.

615 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, doi: 10.1002/2017JG004047, 2018.

DelSontro, T., McGinnis, D. F., Wehrli, B., and Ostrovsky, I.: Size does matter: Importance
620 of large bubbles and small-scale hot spots for methane transport, Environmental Science & Technology, 49, 1268-1276, doi: 10.1021/es5054286, 2015.

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
625 Geophysical Research G: Biogeosciences, 121, 1924-1933, doi: 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
630 Geophysical Research: Biogeosciences, 123, 2527-2540, doi: 10.1029/2018JG004622, 2018.
.

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.,
635 60, 1656-1670, doi: 10.1002/lno.10126, 2015.

Fedorova, I., Chetverova, A., Bolshiyanov, 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
640 field observations, Biogeosciences, 12, 345-363, doi: 10.5194/bg-12-345-2015, 2015.

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-
645 409,

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, doi: 10.1016/j.csr.2013.11.010, 2014.
650

Green, P. N., and Woodford, S. K.: Preservation studies on some obligatory methanotrophic bacteria, Lett. Appl. Microbiol., 14, 158-162, 1992.

Günther, F., Overduin, P. P., Yakshina, I. A., Opel, T., Baranskaya, A. V., and Grigoriev, M.
655 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.

Gurevich, E. V.: Influence of air temperature on the river runoff in winter (the Aldan river
660 catchment study), Russian Meteorol. Hydrol., Russian Meteorol. Hydrol., 34, 628–633, 2009.

Hoefman, S., Van Hoorde, K., Boon, N., Vandamme, P., De Vos, P., and Heylen, K.: Survival orrevival: Long-term preservation induces a reversible viable but non-culturable state in methane-oxidizing bacteria, PLoS ONE, 7, e34196, doi:
665 10.1371/journal.pone.0034196, 2012.

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.

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.

Juhls, B., A., Stedmon S. C., Morgenstern A., M., Meyer H., Höhlemann J., Heim B., Povazhnyi V, and Overduin, P. P.: Identifying drivers of seasonality in Lena River biogeochemistry and dissolved organic matter fluxes, Front. Environ. Sci. - Biogeochemical Dynamics, in review.

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, doi: 10.1134/S0026261719060043, 2019.

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.

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, doi: 10.1029/2006jg000336, 2007.

Kellogg, C. T. E., McClelland, J. W., Dunton, K. H., and Crump, B. C.: Strong seasonality in arctic estuarine microbial food eebs, Front. Microbiol., 10, 2628, 2019.

King, G. M., and Adamsen, A. P.: Effects of temperature on methane consumption in a forest soil and in pure cultures of the methanotroph *Methylomonas rubra*, Appl Environ Microbiol, 58, 2758-2763, 1992.

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, doi: 10.1038/s41598-017-05783-2, 2017.

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.

Kruse, S., Bolshiyanov, 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.

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.

720

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.

725   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.

Magen, C., Lapham, L. L., Pohlman, J. W., Marshall, K., Bosman, S., Casso, M., and
730   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
735   Alaskan lakes, Biogeosciences, 12, 4595-4606, doi:10.5194/bg-12-4595-2015, 2015.

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,
740   doi:10.1016/j.scitotenv.2017.06.187, 2017.

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,
745   2017a.

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
750   Reports, 7, 42997, doi:10.1038/srep42997, 2017b.

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,
755   Polar Science, doi: 10.1016/j.polar.2019.01.005, 2019.

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, doi:
10.1002/2015GL066235, 2015.

760

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.

765   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, doi: 10.5194/tcd-9-3741-2015, 2015.

770 Overduin, P., Blender, F., Bolshiyanov, 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.

Peterson, B. J., Holmes, R. M., McClelland, J. W., Vörösmarty, C. J., Lammers, R. B.,
775 Shiklomanov, A. I., Shiklomanov, I. A., and Rahmstorf, S.: Increasing river discharge to the Arctic Ocean, Science, 298, 2171-2173, doi: 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.,
780 Vadakkepuliyambatta, S., Mienert, J., and Lund Myhre, C.: Methane at Svalbard and over the European Arctic Ocean, Atmos. Chem. Phys., 18, 17207-17224, doi: 10.5194/acp-18-17207-2018, 2018.

Prowse, T., Alfredsen, K., Beltaos, S., Bonsal, B., Duguay, C., Korhola, A., McNamara, J.,
785 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.

Raven, J. A., and Geider, R. J.: Temperature and algal growth, New Phytol., 110, 441-461, doi: 10.1111/j.1469-8137.1988.tb00282.x, 1988.
790
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.

795 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, doi: 10.1016/j.ecss.2005.08.009, 2006.

Santibáñez, P. A., Michaud, A. B., Vick-Majors, T. J., D'Andrilli, J., Chiuchiolo, A., Hand, K.
800 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.

Saunois, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J. G., Dlugokencky,
805 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, 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.,
810 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., Voulgarakis, A., van Weele, M., van der Werf, G. R., Weiss, R., Wiedinmyer, C., Wilton, D.
815 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, doi: 10.5194/essd-8-697-2016, 2016.

820 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.

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 825 transect in Alaska, Biogeosciences, 12, 3197-3223, doi: 10.5194/bg-12-3197-2015, 2015.

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, doi: 10.1038/s41467-019-09592-1, 2019.
830
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.

835 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.
840
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, submitted.

845 Strauss, J., Boike, J., Bolshiyanov, 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 850 under warming conditions. , Environmental Research Letters, 10, 1-9, doi: 10.1088/1748-9326/10/5/054016, 2015.

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, 10,764-710,772, doi: 855 10.1002/2016GL070796, 2016.

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, doi: 10.1002/2014GL061814, 2014.
860
Thottathil, S. D., Reis, P. C. J., and Prairie, Y. T.: Methane oxidation kinetics in northern freshwater lakes, Biogeochem., 143, 105-116, doi: 10.1007/s10533-019-00552-x, 2019.

Townsend-Small, A., Åkerström, F., Arp, C. D., and Hinkel, K. M.: Spatial and temporal 865 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.

870

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.

875 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, doi.org/10.1002/lno.11172, 2019.

Wåhlström, I., Dieterich, C., Pemberton, P., and Meier, H. E. M.: Impact of increasing inflow
880 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, doi: 10.1002/2015JG003307, 2016.

Walter Anthony, K., Daanen, R., Anthony, P., Schneider von Deimling, T., Ping, C.-L.,
885 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.

890 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.

895 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.

900 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.

905 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.

910 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.

Winkel, M., Mitzscherling, J., Overduin, P. P., Horn, F., Winterfeld, M., Rijkers, R.,
915 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.

920   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.

925   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.