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Distribution of methane in the Lena Delta and Buor Khaya Bay, Russia

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Abstract

The Lena River is one of the largest Russian rivers draining into the Laptev Sea. The permafrost areas surrounding the Lena are predicted to melt at increasing rates due to global temperature increases. With this melting, large amounts of carbon – either organic or as methane – will reach the waters of the Lena and the adjacent Buor Khaya Bay (Laptev Sea).

Methane concentrations and the isotopic signal of methane in the waters of the Lena Delta and estuary were monitored from 2008 to 2010. Meltwater run-off of permafrost soils produced hotspots for methane input into the river system (median concentration 1500 nM) compared with concentrations of around 100 nM observed in the main channels of the Lena. Within the river, especially at sites with meltwater input, microbiological experiments indicated strong in situ methane production but a very low methane oxidation potential. In the estuary of Buor Khaya Bay, methane concentrations decreased towards background levels of 20 nM. Here, the strong stratification of the water column permits the dilution of methane with seawater, and methane is released mainly by diffusion into the atmosphere.

1 Introduction

The Arctic Ocean is an intercontinental sea surrounded by the landmasses of Alaska, Canada, Greenland and Siberia/Russia. Large areas are shallow shelf seas into which about 10 % of global runoff flows (Lammers et al., 2001). Many Arctic rivers carry high concentrations of dissolved and particulate material and, in addition, eroding coastlines also contribute to a strong terrestric input (Dittmar and Kattner, 2003; Lantuit et al., 2011).

Although most regions of Earth have warmed over recent decades, that observed in the Arctic far exceeds the global average, and consequently, observed changes are also more extreme (Symon et al., 2005; IPCC et al., 2007). One of the most

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obvious and straightforward implications of the observed warming, is that river runoff will change/increase (Peterson et al., 2002). Additionally, one of the most profound future consequences of permafrost thaw is that the Arctic terrestrial freshwater system is likely to undergo a transition from a surface-water-dominated system to a groundwater-dominated system (Frey and McClelland, 2009). The disproportionate influence of rivers on the Arctic Ocean means that changes in the discharge or chemistry of Arctic rivers have potentially large implications for ocean physics, chemistry and biology (Holmes et al., 2011).

Because methane is an efficient greenhouse gas, understanding methane sources and sinks is important for studying local processes and determining global methane budgets. However, methane release from the Arctic area, including gas hydrate and marine permafrost, is not currently included in the IPCC methane budget calculations.

While it is clear that there are substantial stocks of carbon in the Arctic, there are also significant uncertainties associated with the magnitude of organic matter stocks contained in permafrost, and the storage of methane hydrates beneath both subterranean and submerged permafrost of the Arctic (McGuire et al., 2009). The Arctic is a substantial source of methane to the atmosphere (between 32 and 112 $Tg CH_4 yr^{-1}$), primarily because of the large area of wetlands throughout the region (McGuire et al., 2009).

The Lena River is the second-largest river ($530 km^3 yr^{-1}$) draining into the shallow Laptev Sea, and further into the Arctic Ocean (Peterson et al., 2002). The methane cycle in the Laptev and East Siberian Sea has been investigated intensively by Shakhova and Semiletov and co-workers. The rivers Indigirka and Kolyma transport significant amounts of methane into the East Siberian Sea (Shakhova et al., 2005), and even the atmospheric methane signal mirrored the strong methane import of this area (Shakhova and Semiletov, 2007). However, the influence of the Lena River on the methane budget of the ESS and Laptev Sea is not quite clear; some previous studies have suggested that it plays a minor role (Semiletov et al., 2011), whereas others reported high methane concentrations in its estuary (Shakhova et al., 2010).

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The aim of this study is a detailed analysis of methane distribution in the Lena River and its estuary. With the help of experiments and isotopic methane signature, we assess the further fate of this riverine methane, and process of dilution, oxidation and atmospheric flux.

2 Material and methods

2.1 Study site

Water samples were collected during three summer expeditions in 2008, 2009 and 2010 (Boike et al., 2009; Wetterich et al., 2011) within the main channels of the Lena River and Buor Khaya Bay (Fig. 1). River water was sampled from the "Puteyski 405" survey ship, via a Unite water sampler (Unite, Austria). Coastal water was sampled with Niskin bottles from the ship "TB 0012" during 2010. Additional parameters (temperature, salinity, oxygen, pH) were obtained either with a portable multi-parameter probe (Multi 350i, WTW) on board, or with a CTD probe (Eco 159, Sea and Sun) directly in the water. Water samples were collected from the surface, above and below the thermocline if applicable, and near the bottom.

Serum bottles (120 ml) were flushed extensively and closed with butyl stoppers; excess water could escape via a needle in the stopper. Samples were poisoned with 0.01 % $HgCl_2$. In the laboratory, 20 ml of nitrogen were added to extract methane from the water phase, and excess water could escape via a needle. The volumes of the water and gas phases were calculated by differential weighing.

Meltwater running off the permafrost soil in small streams was filled directly into 12 ml serum vials and analysed as described above.

2.2 Gas analysis

Headspace methane concentrations were analysed in the laboratory with a gas chromatograph (GC 2014, Shimadzu) equipped with a flame ionisation detector and

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a molecular sieve column (Hay Sep N, 80/100, Alltech). The temperatures of the oven, the injector and detector were 40 °C, 120 °C and 160 °C, respectively. The carrier gas (N_2) flow was 20 ml min^{-1}, with 40 ml min^{-1} H_2 and 400 ml min^{-1} synthetic air. Gas standards (Air Liquide) with methane concentrations of 10 and 100 ppm were used for calibration.

After measuring the methane concentration, the headspace was analysed for the isotopic composition of methane. Depending on the methane concentration of the headspace, 0.5–2.5 ml of the headspace was transferred into an evacuated 20-ml gas sampling tube. These data are only available for 2010.

The $\delta^{13}C_{CH_4}$ values were determined by a Finnigan DELTAplus XP mass spectrometer. The extracted gas was purged and trapped with a PreCon unit to pre-concentrate the sample. The reproducibility, as derived from duplicates, was 1.6 %. The isotope ratios are presented relative to the VPDB (Vienna Pee Dee Belemnite) standard using the conventional delta notation (Craig, 1957).

2.3 Calculation of methane oxidation

For the calculation of the fraction of methane oxidised, the maximum methane concentration of a sample group was set as fraction 1, and the other concentrations were related to this concentration accordingly. Starting from the data point with the maximum methane concentration and the corresponding $\delta^{13}C$, the theoretical $\delta^{13}C$ of a given methane fraction was calculated. A Rayleigh distillation model of the type discussed by (Coleman et al., 1981), i.e.

$$\delta^{13}C_{CH_4} = 1000 \cdot (1/\alpha - 1) \cdot \ln f + (\delta^{13}C_{CH_4})_0 \quad (1)$$

was used to determine fraction f of the methane remaining (thus, $1 - f$ is the methane consumed by oxidation), where α is the kinetic isotope fractionation factor and $(\delta^{13}C_{CH_4})_0$ is the starting stable isotopic composition. From the literature, fractionation factors for microbial methane oxidation range from 1.02 in freshwater (Bastviken

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et al., 2002) to 1.017 for Arctic marine water (Damm et al., 2007). When only diffusion is assumed, an α of 1.0009 is reported (Happell et al., 1995).

2.4 Mixing experiments

Microorganisms were separated from their original water (8 l) by filtration through 3- and 0.2-µm filters (Satorius). The filters were resuspended in 20–40 ml of the original water, and kept cool (4 °C) until further processing. The estuarine sample originated from station T1 1005 at 10 m water depth (Fig. 1), with an in situ temperature of 0.8 °C, a salinity of 15.2 and a methane concentration of 19 nM. The riverine sample originated from station T5 1003 surface (Fig. 1), with an in situ temperature of 19.5 °C, a salinity of 0 and a methane concentration of 363 nM. The "permafrost" sample was obtained from a meltwater stream at the foot of a melting permafrost cliff (72°20.169 N 126°17.750 E); no bacterial samples were taken.

For the experiment, the filters were vigorously mixed, and 1-ml subsamples were distributed into 12-ml glass vials. The concentrate was diluted with 3 ml of the filtered river or marine water. Incubation was initiated with the addition of methane to a final concentration of 335 nM. Twelve samples served as controls and were killed immediately after the addition of methane. Each combination was measured in triplicate. Samples were incubated in the dark and outside. Temperatures ranged from 6 to 16 °C. Incubation was stopped after 40 h by the addition of $HgCl_2$ (0.01 % final concentration). In the home laboratory, methane concentrations within the headspace were determined. Methane consumption or production rates (nmol l^{-1} h^{-1}) were related to the original bacterial concentration in the sample and the incubation time.

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3 Results

3.1 Hydrographic/oceanographic information

For 2008 and 2009, only data from the surface and the bottom are available, whereas data for 2010 cover the whole water column in Buor Khaya Bay. In the middle of the Bay and around Muostakh Island, temperatures ranged between 13 and 15 °C at the surface and 3–4 °C at the bottom in 2008. The corresponding salinities were around 1 at the surface and around 21 at the bottom. Otherwise, temperatures ranged from 10 to 18 °C with salinities < 1. In 2009, the Lena River was slightly colder, with an average temperature of 12 °C. Only one station was found to have brackish water (12 PSU and 2 °C). In 2010, the Olenekskaya Channel was relatively warm at 19 °C, compared with 16 °C in other channels. No differences were observed between surface and bottom temperatures, whereas distinct stratification was observed in Buor Khaya Bay (Fig. 2, example shows Transect 1). Cold, saline water was observed below a water depth of 12 m, while at the surface the warmer Lena River water extended far to the northeast (Bussmann, 2011).

3.2 Methane concentrations

In a first overview (Fig. 3), the median methane concentrations in the area were comparable between the three sampling years, even with a slight increase from a median of 28 nM in 2008 (n = 43) to 35 nM in 2009 (n = 35) and 41 nM (n = 63) in 2010. However, methane concentrations of permafrost meltwater were orders of magnitude higher (median of 1505 nM, n = 7, samples from 2009 and 2010).

In 2008, the outlet of the Bykovskaya Channel into Buor Khaya Bay, and the area around Muostakh Island were investigated (Fig. 4a). The highest methane concentrations were observed at the outlet of the Bykovskaya Channel (150–280 nM), while in Buor Khaya Bay the concentration decreased to background levels of around 20 nM. In 2009, the area around Muostakh Island and in Buor Khaya Bay – comparable to the

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area investigated in 2008 – also revealed only background levels of around 20 nM; however, the same low concentrations were also observed in the Lena Channels (Fig. 4b). Higher concentrations (of around 100 nM) were only observed in the Olenekskaya Channel. In 2010, maximal methane concentrations of 418 nM were also observed in the Olenekskaya Channel. Water temperature in this channel was rather high (19 °C), with oxygen content of 7.1 mg l^{-1}. The other main channels of the Lena revealed intermediate concentrations of around 100 nM, and concentrations decreased further to background levels in Buor Khaya Bay (Fig. 4c).

3.3 Isotopic signature of methane

The lightest carbon isotope signature of methane -42.3 ± 1.7 ($n = 3$, Fig. 5B) was recorded in the river water of the Olenekskaya Channel together with methane concentrations of 414 ± 52 nM. This isotopic signature of the Olenekskaya Channel was comparable to the signature of the meltwater (-39.4 ± 1.9, $n = 3$, Fig. 5A); however, here, much higher methane concentrations of 1891 ± 877 nM were measured. Unfortunately, the isotopic samples from the other Lena channels were lost and no information is available. Methane in the waters of Buor Khaya Bay was much heavier (-33.6 ± 0.8, $n = 30$, Fig. 5C). The corresponding methane concentrations covered a wide range, from approximately 10 to 100 nM. However, at two stations we detected a very heavy signature of -12.1 ± 2.1 ($n = 4$, Fig. 5D) together with low methane concentrations (21 ± 3 nM). These data points were excluded from further analysis and discussion, as they are likely of other, non-biogenic origin.

3.4 Mixing experiments

In order to estimate the influence of the mixing of the Lena River with the marine Laptev Sea water, we separated and concentrated the original riverine bacteria from their water and mixed them with the marine Laptev Sea water, and vice versa. Additionally, sterile filtered meltwater from permafrost was used as medium. The riverine bacteria

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revealed a strong potential of methane production (Fig. 6). The mixing of the riverine bacteria with the coastal water resulted in a reduced activity, while the addition of meltwater increased the methane production potential. The potential activity of the coastal bacteria was much lower than that of the riverine bacteria (Fig. 6). Thus, the mixing with the riverine freshwater showed no clear effect. However, the addition of meltwater switched the "neutral" methane production/consumption activity towards a clear signal of methane production. Additional simple incubations of natural river water with natural methane concentrations (ca. 400 nM, 24 h) revealed no potential of methane consumption.

4 Discussion

4.1 Processes within the river

Methane concentrations in the Lena River from a 3-yr period were around 100 nM, which is the same range of concentrations observed by Semiletov et al. (2011). These concentrations are within the lower range of 10 to 1400 nM reported for boreal rivers (Middelburg et al., 2002). Isotopic data of methane in meltwater and in the Olenekskaya Channel (−42 and −39‰) are rather heavy when compared to other Arctic lakes, which were reported to be within the range −59 to −79‰ (Walter et al., 2008), or −72‰ reported for permafrost sediment cores (Koch et al., 2009).

Methane concentrations within the channels of the Lena River can be altered by different processes. Higher methane concentrations can be due to input of meltwater and other tributaries, or in situ production. On the other hand, reduction of the methane content can be due to microbial oxidation and diffusion into the atmosphere.

The Olenekskaya Channel is characterized by high methane concentration and an isotopic signal similar to the meltwater; thus, we assume a high meltwater input here. Additionally, the experiments show that – especially in the Olenekskaya Channel – there is a strong potential for in situ methane production, which is further increased

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by the addition of meltwater-based organic matter. For the Trofimovskaya Channel, there seem to be further sources of methane along its flow, as seen by the increase of methane concentration with flow distance (Fig. 7). A similar increase in methane is also reported from boreal rivers (Angelis and Lilley, 1987). However, in the Bykovskaya Channel, methane concentrations did not change along the flow distances.

As discussed, declining methane concentrations within the river could be attributed to microbial oxidation or diffusion into the atmosphere. From simple incubation experiments and the mixing experiment, we found no indications of active methane oxidation (or it was below our detection threshold). Applying an isotopic model in the rivers appears to be infeasible, because basic assumptions (no other sources of methane) are not valid.

Gas exchange across an air-water interface can be described in general by the following function (Angelis and Lilley, 1987): $F = k \cdot (c_m - c_e)$, where F is the rate of gas flux per unit area, c_m is the methane concentration measured in surface water and c_e is the atmospheric gas equilibrium concentration based on Wiesenburg and Guinasso (1979). The gas exchange coefficient, k, is a function of water surface agitation.

In most rivers, stream turbulence is more important for gas exchange than wind stress (Angelis and Lilley, 1987). Therefore, for river flux calculations, a simplified turbulence model was applied, where the gas exchange coefficient is defined as $k = 1.46 \cdot (D \cdot V/h)^{0.5}$, where D is the molecular diffusion coefficient of methane at the respective temperature and salinity (Unisense gas tables; modified from Broecker and Peng, 1974), V is stream velocity and h stream depth. Data on stream velocity at the respective date and position were kindly provided by I. Fedorova (2012).

For the Olenekskaya Channel, which has high methane concentrations, we also calculated high methane fluxes ($x = 852\,\mu mol\,m^{-2}\,d^{-1}$, $n = 4$, or $14\,mg\,C\,m^{-2}\,d^{-1}$). In the Olenekskaya Channel, ebullition was also observed, which will thus increase the total methane flux (Baulch et al., 2011). For the Trofimovskaya and Bykovskaya Channels, the flux was lower, at 167–278 $\mu mol\,m^{-2}\,d^{-1}$ or 3–4 $mg\,C\,m^{-2}\,d^{-1}$. However, locally increased stream velocities in the Trofimovskaya Channel also increased the methane

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flux to 597 µmol $m^{-2} d^{-1}$ (10 mg C $m^{-2} d^{-1}$). At the nearby Samoylov research station, the "terrestric" methane flux ranged from 4 to 28 mg C $m^{-2} d^{-1}$ at the rim and the depression of polygon structures (Kutzbach et al., 2004). Eddy covariance indicated an average daily methane flux of 18.7 mg $m^{-2} d^{-1}$, mainly determined by wind velocity (Sachs et al., 2008). Thus, the methane flux from the Lena River and its various channels is slightly less than the terrestric emissions, but within the same range.

The warmest water temperatures in the Lena were previously recorded in July/August (ca. 15 °C) (Yang et al., 2002); as methanogenesis is strongly influenced by temperature (Eugster et al., 2011), we also propose highest methane production rates in July/August. Soil temperatures and therefore permafrost melting rates are also highest during these months (Boike et al., 2012). Thus, we assume that our methane flux represents the maximal flux on a seasonal scale. The strength of the methane flux following the spring ice-melt remains uncertain.

Thus, we can only conclude that the observed methane concentrations of around 100 nM are the result of a strong meltwater input and a strong in situ production of methane. As we had little evidence for methane oxidation activities, the decrease of riverine methane appears to be mainly governed by diffusion into the atmosphere.

4.2 Processes within Buor Khaya Bay

When the Lena River enters the coastal area of Buor Khaya Bay, methane concentrations decrease from 100 nM to around 20 nM, together with a slight shift of the isotopic values towards a heavier signature (Fig. 5). Within the bay, we have no indications of any methane sources, and only processes to reduce methane concentrations seem to be relevant.

4.2.1 Dilution

To determine whether the decrease in methane concentrations observed within Buor Khaya Bay was due to dilution, the methane data were plotted against salinity (Fig. 8).

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For the years 2008 and 2010, no correlation was observed between salinity and methane concentration. This was also true when only data with a salinity of > 1 were used. Thus, methane loss is not the result of mixing methane-rich river water with methane-poor marine water, as described for other estuaries (Angelis and Lilley, 1987). This may be due to the very strong stratification of the water column (Fig. 2), as in other river-dominated and stratified estuaries (Middelburg et al., 2002).

4.2.2 Production and oxidation (microbial processes)

Methane concentration may also be reduced due to microbial oxidation, and can be inferred from isotopic data. In Fig. 5, data from Buor Khaya Bay are related to the data set with the maximal methane concentration (102 nM); Rayleigh curves are plotted to show how the isotopic signal would change due to microbial oxidation with a fraction factor of 1.017, or due to diffusion from water into the atmosphere with fractionation factor of 1.0009 (see Material and Methods). As seen in Fig. 9, the observed decrease in methane concentration and the relatively minor shift towards heavier methane cannot be explained by methane oxidation; instead, diffusion seems to be the dominant process here.

The lack of methane oxidation is in contrast to other estuaries, where, during summer, oxidation removed a substantial proportion of methane from the freshwater and brackish areas of the estuary (Angelis and Scranton, 1993). This low methane oxidation potential is also supported by our experiments. However, it is not yet clear why the methane oxidation is so low.

The functions developed by Wanninkhof (1992) calculate the flux of dissolved methane into the atmosphere. In addition to our data on water temperatures, salinities and methane concentrations at transects 1 and 3, we used wind speeds of 2–4 $m\,s^{-1}$ (www.aari.ru, data for Tiksi). Consequently, the flux of dissolved methane from Buor Khaya Bay into the atmosphere ranged from 21 to 49 $\mu mol\,m^{-2}\,d^{-1}$ along the coast (or 0.33–0.79 $mg\,C\,m^{-2}\,d^{-1}$). At the northernmost station, the methane flux decreased to 5 $\mu mol\,m^{-2}\,d^{-1}$ (or 0.08 $mg\,C\,m^{-2}\,d^{-1}$). This is much lower than the minimum flux

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of 130 µmol $m^{-2} d^{-1}$ previously estimated for European estuaries (Middelburg et al., 2002). Shakhova and Semiletov (2007) reported 3.02–4.86 g C $cm^{-2} h^{-1}$ (or 0.07–0.12 mg C $m^{-2} d^{-1}$) methane flux from the East Siberian Sea and northern parts of Buor Khaya Bay in 2003 and 2004. These data correspond well with our northernmost station.

5 Conclusions

The concentration and isotopic signature of methane in the Lena River and Buor Khaya Bay reveal intermediate concentrations comparable to boreal estuaries. Within the river, especially at sites with meltwater input, experimental results suggest a strong in situ methane production, but a very low, non-detectable methane oxidation potential. Thus, methane is released from the river and estuary water mainly by diffusion into the atmosphere. Riverine methane fluxes are within the same order of magnitude as the "surrounding" terrestrial methane fluxes. Within Buor Khaya Bay, the strong stratification of the water column permits the dilution of methane and, again, methane is released mainly via diffusion into the atmosphere.

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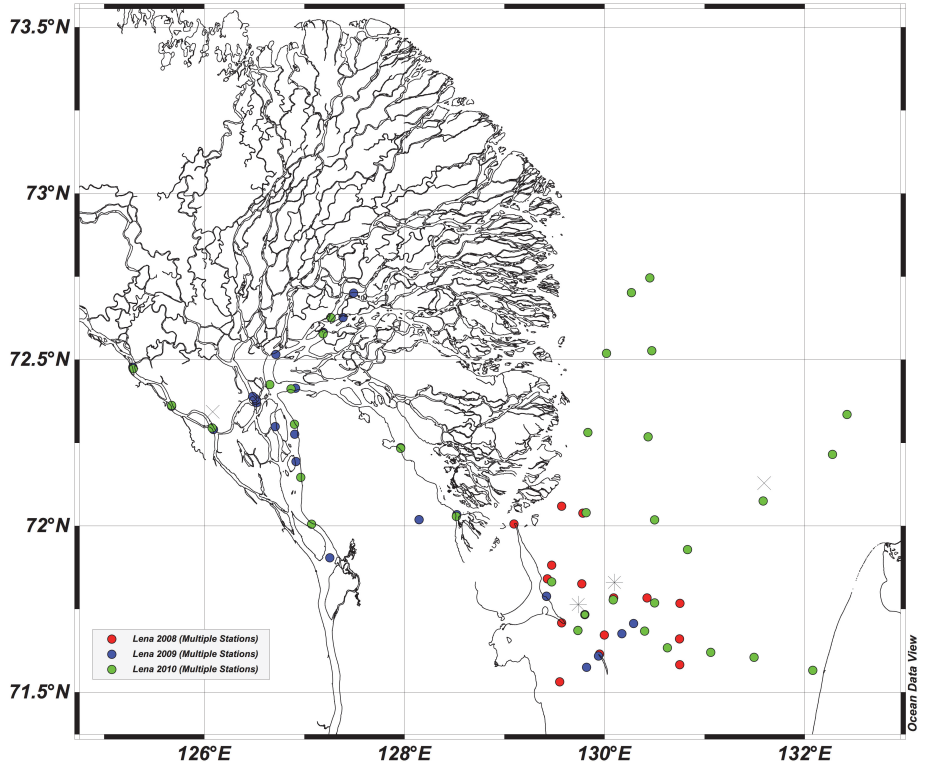

Fig. 1. Sampling stations in the years 2008 (red), 2009 (blue) and 2010 (green) within the Lena Delta and Buor Khaya Bay. Asterisks indicate stations with special isotopic signature, and Xs indicate stations where water samples were used for experiments.

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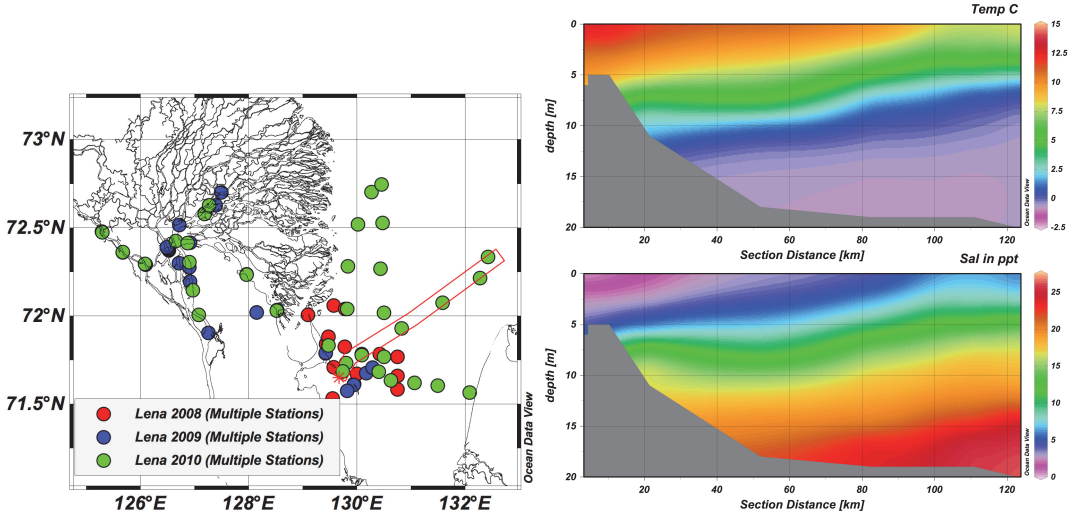

Fig. 2. Temperature and salinity for transect 1 in 2010.

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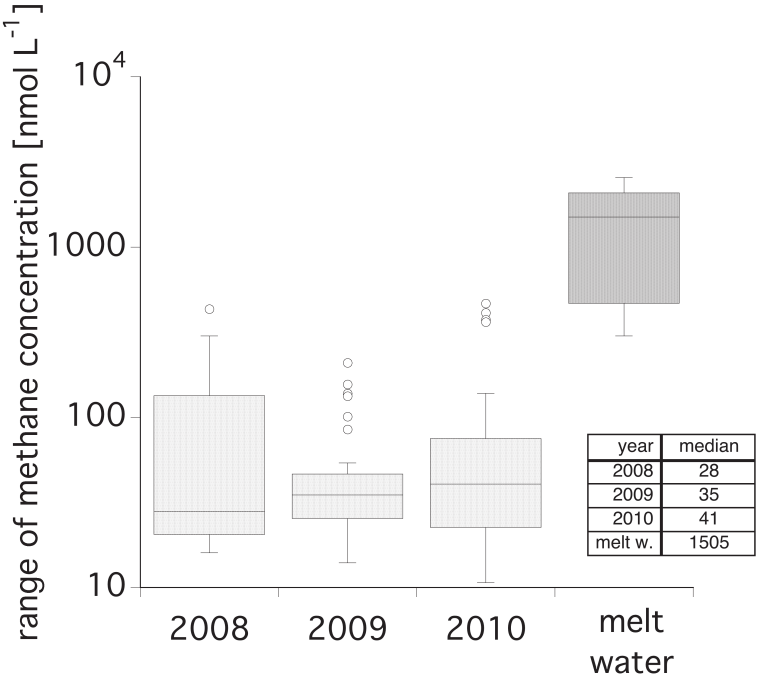

Fig. 3. Range of methane concentrations in the Lena River and Buor Khaya Bay in the years 2008, 2009 and 2010, as well as methane concentrations in permafrost meltwater. The box plot shows the median line, upper and lower quartiles, the line of maximum and minimum values and outliers (dots).

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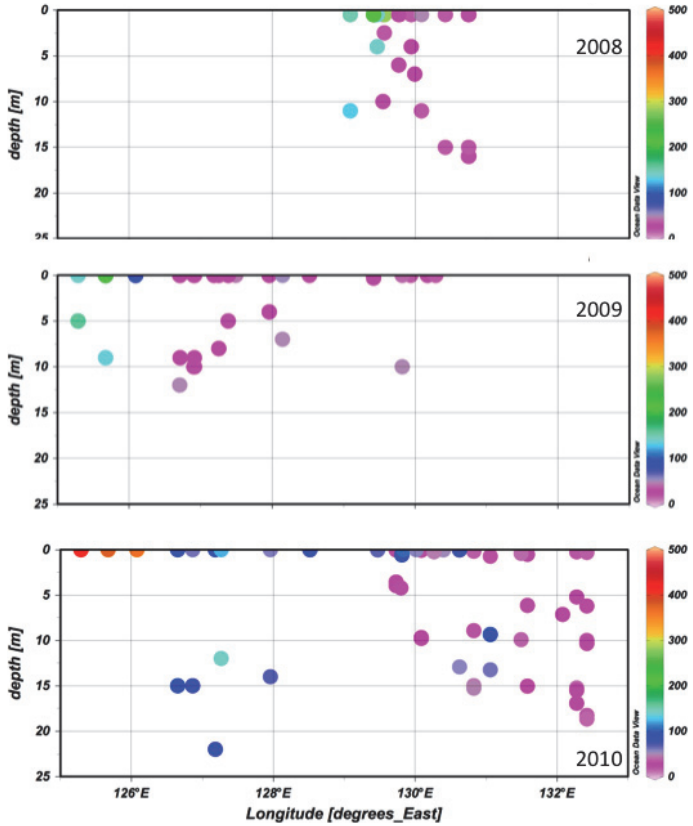

Fig. 4. Distribution of methane concentrations (in nmol L^{-1}) in 2008, 2009 and 2010 versus longitude. Locations of the sampling stations are shown in Fig. 1.

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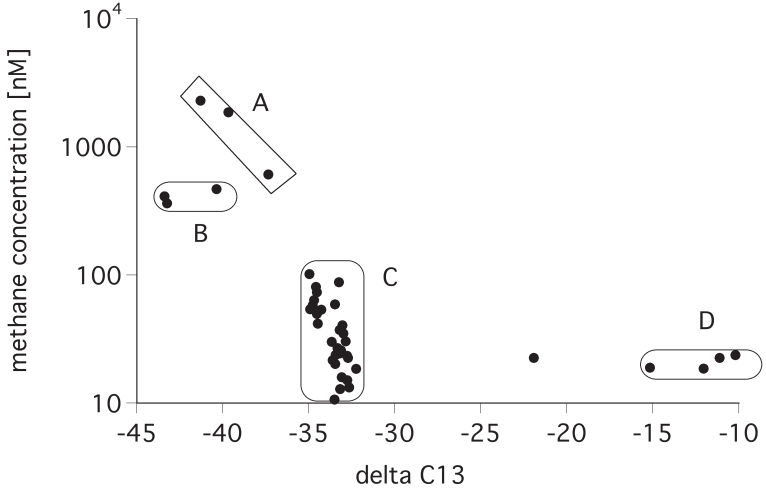

Fig. 5. Carbon isotope signature of methane in 2010, with corresponding methane concentrations in meltwater (A), Olenekskaya Channel (B), Buor Khaya Bay (C) and two separate stations in Buor Khaya Bay (D), indicated with an asterisk in Fig. 1.

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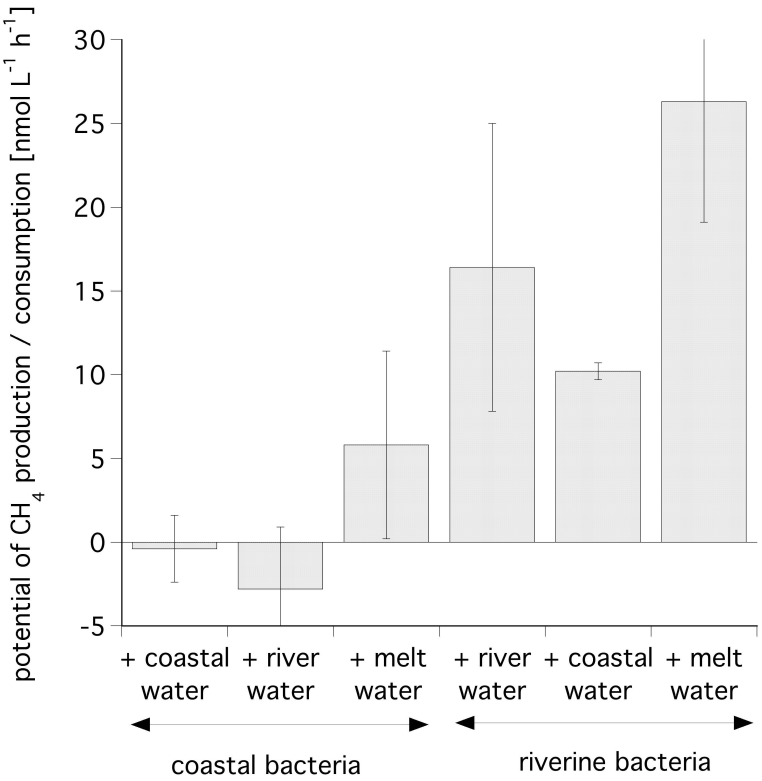

Fig. 6. Experiment with bacteria extracted from coastal or river water and mixed with coastal, river or permafrost meltwater; and corresponding potential methane turnover rates after adding methane to a concentrations of 335 nM. Results show the average of three samples with standard deviation.

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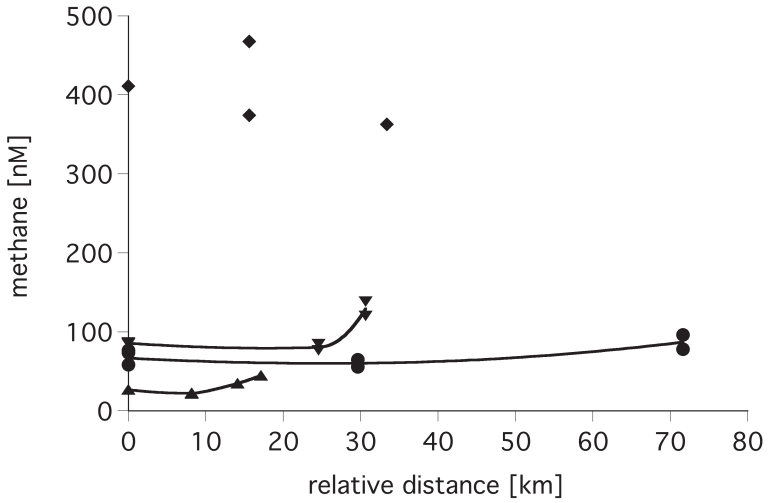

Fig. 7. Methane concentrations in the Olenekskaya Channel 2010 (diamonds), Trofimovskaya Channel 2008 (upward triangle) and 2010 (downward triangle) and the Bykovskaya Channel (circles), plotted versus flow direction and distances from the first station (set as zero km).

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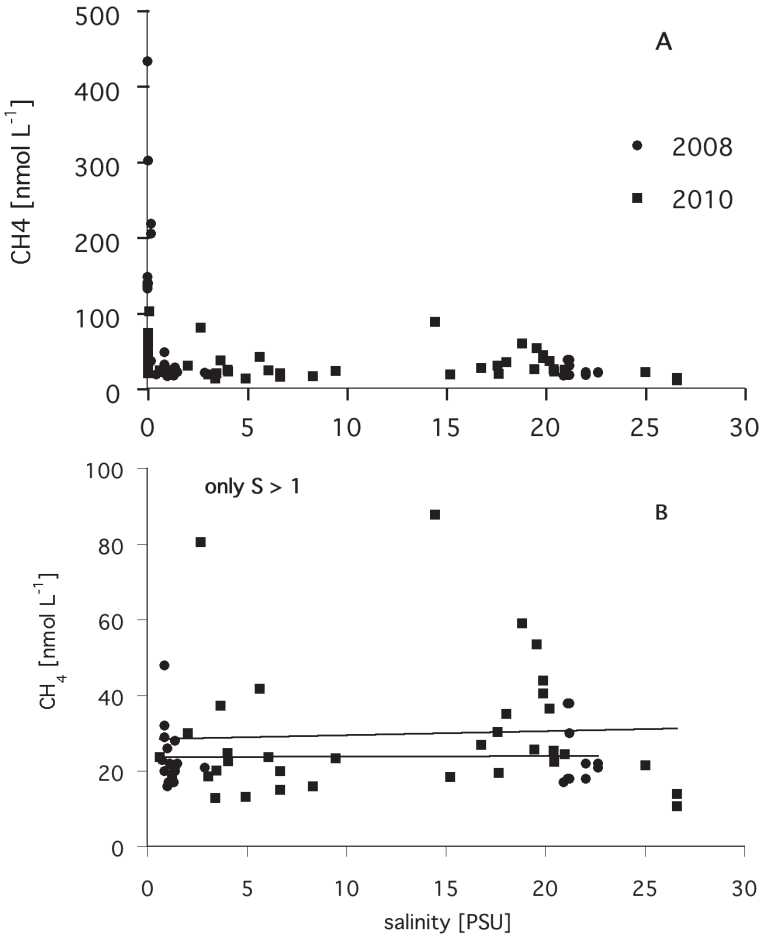

Fig. 8. Methane concentrations in Buor Khaya Bay versus salinity for 2008 (circles) and 2010 (squares). **(A)** all samples, **(B)** only samples with a salinity of > 1.

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Distribution of methane in the Lena Delta and Buor Khaya Bay, Russia

I. Bussmann

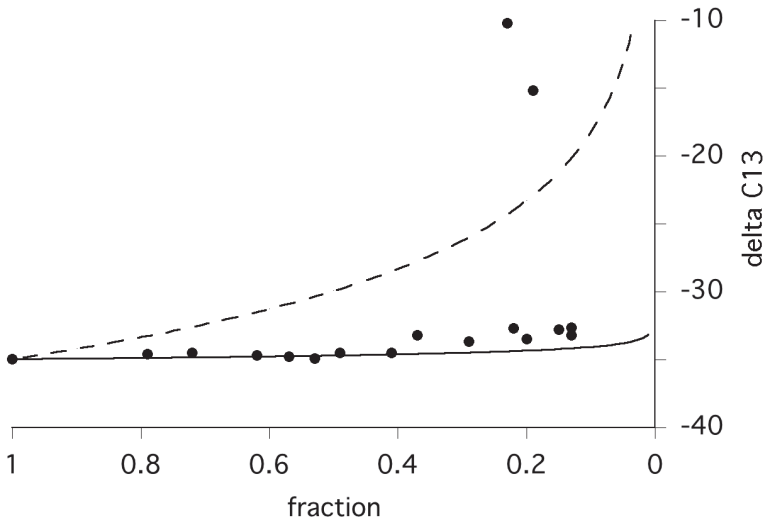

Fig. 9. Fraction of methane oxidised in waters of Buor Khaya Bay in 2010. The end member of the Rayleigh curve is based on the data set with the highest methane concentration (102 nM) and the corresponding isotope signature of −34.96‰ VPDB. The curves show the prospective $\delta^{13}C$ signatures modified by diffusion (a = 1.0009, solid line) or oxidation (a = 1.017, dashed line).

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