

The role of the Siberian rivers in increasing dissolved methane in the East Siberian shelf

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Abstract. Based on the climatic picture of circulation the problem of modeling the dissolved methane in the Arctic Ocean with a drain of the Siberian Rivers is considered. The regional large-scale ocean model, developed in the ICM&MG, SB RAS is used. The distribution of the dissolved methane in the sea water is obtained with the advection-diffusion equation with parameterization of the oxidation process included.

As a result of numerical modeling accumulation processes and stability with respect to time of high concentration of the dissolved methane in the Laptev Sea, known from the observational data are reproduced. It is obtained that the drain of the Lena River can bring a powerful contribution to an abnormal high concentration of methane in the Dm. Laptev Strait water. It is shown that methane oxidation controls the CH₄ release in the atmosphere.

Introduction

The atmospheric methane is a greenhouse gas that plays an important role in the chemistry of the Earth's atmosphere [1]. The ocean constitutes a vast methane reservoir, mainly, in the form of hydrates, although they are thought to play minor role in the modern methane cycle. A massive methane release from the sea-floor sediments due to decomposition of methane hydrate is a potential contributor to the global warming. Estimates of the global oceanic methane emissions considerably differ and range from 0.4–0.8 Tg/yr for the open ocean [2] up to 16–40 Tg/yr including the coastal areas and marine geological sources [3].

Interest to studying a cycle of carbon in the Arctic regions is caused by the climatic changes which occur in the north high latitudes and are expressed in increasing the surface air temperature, permafrost thawing, and an increase in a drain of the rivers. The climatic changes in the Arctic may affect the changes in the atmospheric greenhouse gas concentrations (by altering release and uptake of carbon dioxide and methane).

The shelf of the Arctic Ocean plays the role of estuary of the Siberian rivers, watersheds being located on the territory of storing frozen stocks of perennial species and being available for the direct discharge of methane. The recent studies have shown that an increase in the air temperature in northern latitudes since 1985 has averaged 0.68 °C per decade [4]. At the same, the freshwater discharge over the course of the past century in

some Siberian rivers has increased [5, 6]. The destruction of permafrost and changes in hydrological conditions, in particular, an increase in runoff, contribute to the Arctic basin in the global methane cycle.

A number of studies have shown that the role of the Arctic Ocean in the global methane cycle is substantially underestimated [7–10]. Thus, as a result of the field research in 2003–2008 in the East Siberian Sea and in the Laptev Sea revealed a high content of dissolved methane plumes [9, 10]. It was experimentally established that the bottom water layer in these areas is saturated with methane in its content in the atmosphere of high latitudes of the north hemisphere by 2,200–3,000 %, and in the surface water — up to 700–2,000 %. The extreme methane anomalies in the plume areas indicate to the presence of both the surface and bottom methane sources.

Concentrations of dissolved methane in the surface water layer in the estuaries of the Siberian Rivers ranged from 7 to 700 nmol/L [11]. According to the results obtained [11], the concentration of dissolved methane increased from west to east. The lowest methane values were detected in the estuary of the Ob River (7.4–41.3 nmol/L) increasing in the estuary of the Yenisey River (7.1–130.8 nmol/L). The highest methane values were found in the Lena River estuary (the mean value being 61.6–651.2 nmol/L). The authors suggested that the west-to-east increase in the dissolved methane distribution reflects the stage of permafrost degradation. Indeed, the Ob basin is located in the region partially underlain by discontinuous and sporadic permafrost. The area of the Yenisey River is underlain by discontinuous and island permafrost and the Lena watershed is totally in the zone of continuous permafrost.

The data obtained by measuring the methane concentration in the surface layer of the atmosphere, have shown that the anomalously high values of methane in the air (up to 8 ppm) of space combined with patches of anomalously high methane concentrations in the sea water (up to 500 nmol/L [9]).

It has been hypothesized to enhance transport in the atmosphere released from methane hydrates offshore due to the formation of cross-cutting taliks, including those caused by the current warming, and increase the gas permeability of the permafrost sediments [9]. An alternative view is that an increase in the flow of methane into the atmosphere could be due to increased biogenic methane in the surface sediments of the bottom layer due to an increase in the removal of organic matter on the shelf of the rivers and an increasing temperature of the bottom waters [12]. In this context, it becomes relevant to identify an individual contribution of freshwater and marine ecosystems of the Arctic as a source of methane in the Arctic shelf.

In this paper, we examine the contribution of the Siberian Rivers to the removal of high concentrations of dissolved methane in the Arctic Basin using numerical modeling. The main objectives of this study are the following:

1. Studying the distribution of dissolved methane supplied by the river runoff on the Arctic shelf.
2. Assessing the role of oxidation of dissolved methane in the seawater.
3. Quantifying the flow of methane from the waters of the East Siberian shelf into the atmosphere.

1. The ocean model

This paper considers the distribution of dissolved methane coming from Siberian Rivers into the waters of the shelf zone of the Arctic Ocean. The oceanic processes are described using a numerical model of the ocean dynamics that can be traced back to the model of the world ocean circulation (developed in the Institute of Computational Mathematics and Mathematical Geophysics of the Siberian Branch of the Russian Academy of Sciences) adapted to the region of the Arctic and north Atlantic and modified in the course of further investigations. The system of complete nonlinear equations of ocean thermohydrodynamics is written in curvilinear orthogonal coordinates with conventional hydrostatic and the Boussinesq approximations. On the ocean surface, the “rigid lid” approximation is used. The dynamic equations are solved by the method of separating barotropic and baroclinic modes. The problem is solved with respect to time by a hybrid explicit–implicit scheme with the method of splitting to physical processes and spatial coordinates [13,14]. For the diffusion equation, an implicit (in time) numerical scheme combined with the method of splitting to one-dimensional finite difference equations is used. The equation describing the transport by currents is approximated using the QUICKEST scheme [15].

The sea-ice model proposed (known as an elastic viscous–plastic model) is a modification of a standard viscous–plastic model of the ice dynamics. A detailed description of this model can be found in [16–18].

The interaction between the Arctic Basin and north Atlantic is simulated for the domain of the Arctic Ocean and north Atlantic starting with 20°S. The grid resolution for the northern Atlantic is chosen to be 1°. On the circle of 65°N, the spherical grid adopted for the Atlantic is naturally coupled with another orthogonal grid of a finer resolution. The second grid is constructed in the following way: a one-degree spherical grid of coordinate lines with groundbased poles is constructed on a small hemisphere with a diameter equal to the diameter of the circle of 65°N, and the resulting grid is projected onto the polar area limited by the circle of 65°N. In this case, all coordinate lines of the second grid are orthogonal and turn at the latitude of 65°N to the corresponding coordinate lines of the spherical system. A maximum resolution is 35 km. On average, the nodes of the numerical grid of the Arctic Ocean area are located at a distance of about 50 km. The vertical

distribution consists of 33 horizontal levels becoming denser near to the surface, where the resolution is equal to 10 m. A minimum depth of the shelf zone is taken to be 50 m.

The initial distributions of temperature and salinity have been derived from a combination of the Levitus data and a series of oceanographic data collected by sensory investigations of the Arctic and adjacent areas.

2. Methane processing

As a result of the numerical modeling, with the use of re-analysis data NCEP/NCAR, the system of interaction of the North Atlantic water and the Arctic ocean, the including the circulation of waters in the seas of the Siberian shelf has been restored [19].

Based on the climatic circulation picture problem of the simulation of the dissolved methane transport into the Arctic Ocean due to the drain of the Siberian Rivers is considered. By analogy with all other tracers, the transport of methane concentration C is simulated as an advection-diffusion equation with local sources and sinks

$$\frac{\partial C}{\partial t} + u \cdot \nabla C = D - C_{\text{ox}},$$

where u is the velocity of the ocean currents, D describes the process of horizontal and vertical diffusion of methane with the use of second order operators, C_{ox} describes the flow of methane, in particular, due to its oxidation.

The lateral boundaries for the river freshwater inflow are given according to the existing data on the climatic mean seasonal variation of the runoff of 13 major rivers of the Arctic. The values of their flows are taken from the AOMIP database (<http://www.whoi.edu/page.do?pid=29836>). On the lateral boundary of the points corresponding to the deltas of the rivers Ob, Yenisey, Lena, Yana, Kolyma, Indigirka, the concentrations of dissolved methane, corresponding to the measurements [11] are set.

Let us set the flow of rivers as accompanied by additional fresh water (zero salinity) and the methane concentration using the following boundary conditions:

$$-\mu_s \frac{dC}{dn} + (u \cdot n)C = -\frac{C \cdot T_r}{A},$$

where n is normal to the interface through which the flow moves, $u_n = (u \cdot n)$ is the speed in this direction, T_r is a flow rate of the river, A is a lateral surface area equal to the product section of the border length and depth of the pool in this area, μ_s is a horizontal diffusion coefficient.

3. Results of modeling

As part of the described numerical model four of the numerical experiments for the time period from 2002 to 2009 were carried out. In all the experiments, the sources of dissolved methane were specified in the estuaries of the Siberian rivers (Figure 1) ranging from 30 to 300 nmol/L, according to the measurement data [11]. In addition, the following values were adopted, respectively: 30 nmol/L for the Ob estuary; 70 nmol/L for Yenisey, 300 nmol/L for Lena, and 200 nmol/L for Indigirka, Kolyma and Yana.

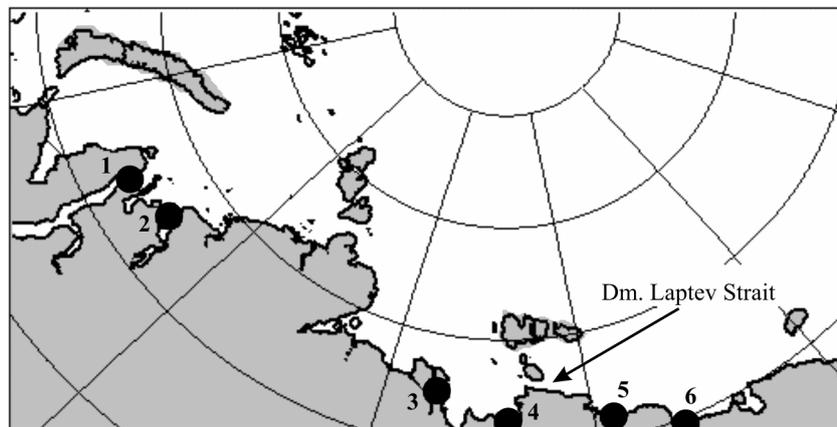


Figure 1. The simulated region. The places of setting methane sources are associated with river flows: 1 Ob, 2 Yenisey, 3 Lena, 4 Yana, 5 Indigirka, and 6 Kolyma

1. In the first numerical experiment, the oxidation of methane $C_{ox} = 0$ is not taken into account.

The spatial distribution of the concentrations of dissolved methane coming from the watersheds of the rivers in the surface layer of the ocean resulting in a numerical experiment for 2005, 2007 is shown in Figure 2. The flow of the river Lena makes a significant contribution to the formation of abnormally high concentrations of methane in the offshore waters of the Laptev Sea. According to the dynamics of the river runoff, a maximum concentration (90 nmol/L) occurs in May. Further extension is in accordance with a system of currents in the cyclonic direction, defined by the system of winds and by the Coriolis force effect deflecting to the right. By September, the highest values (up to 90 nmol/L) are determined in the Dm. Laptev Strait, which corresponds to the measurement data [9].

In the model proposed, the removal of dissolved methane in the central part of the Arctic and its accumulation in the water column occur due to the absence in the model of the oxidation of methane. The dissolved methane is not conserved, being lost through biological oxidation. The methane arrived in the sea water is consumed partly in the ocean by microbial oxidation

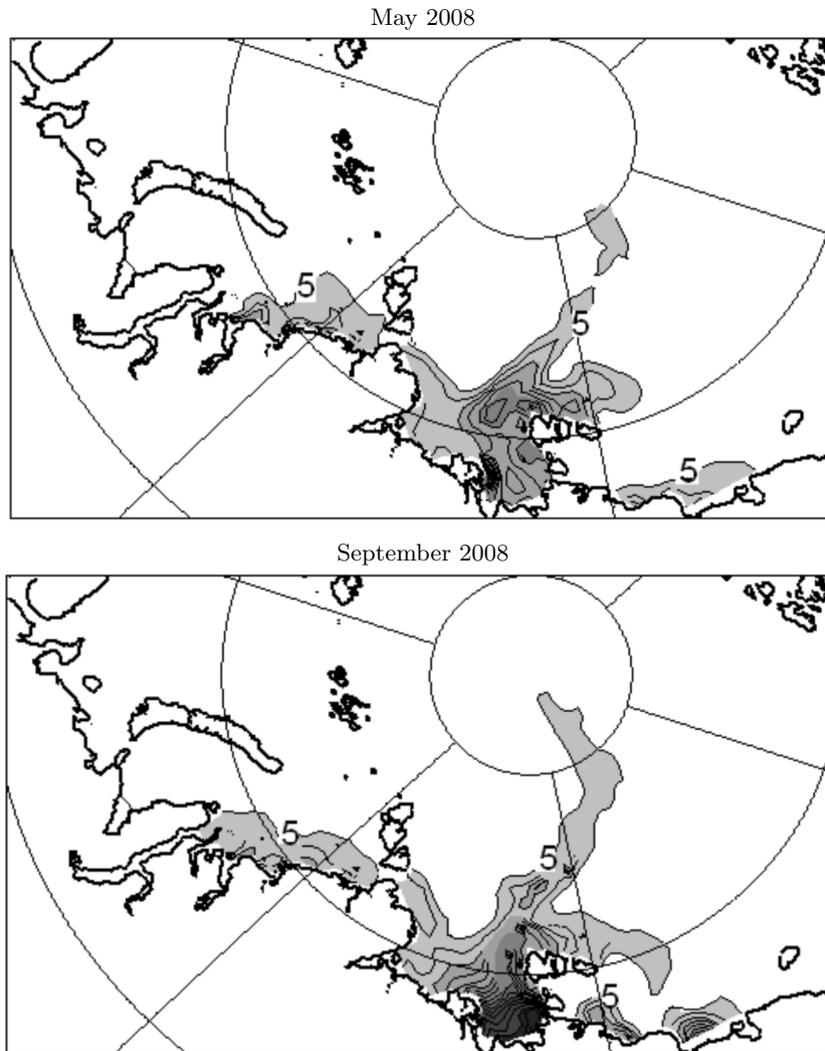


Figure 2. The spatial distribution of dissolved methane in nmol/L obtained in Experiment 1. The step between contours is 5 nmol/L

reducing the portion reaching the atmosphere. The process of methane oxidation was studied in the Black, Baltic, Bering and Kara Seas [20]. The rate of methane oxidation in the water is 0.1–200 nL/L per day.

Various parameterizations of this process are discussed in the literature [21–23]. The main objective of the following experiments is to quantify a fraction of methane oxidized in the water column in relation to the total of discharging, dispersed in the water column and escaping into the atmosphere. In the present paper, two ways of the account of methane oxidation are used.

2. In the second experiment, the oxidation rates of methane correspond to the data of measurements made in the Bering Sea [23]. When the relative methane oxidation rates observed in water samples collected from the same areas were seasonally compared, there were found many differences. According to these data, the rate of oxidation of CH_4 is defined as follows: 1.4 nL/L per day from December to March, 1.3 nL/L per day from April to July, and 2 nL/L per day from August to November.

The resulting distribution of methane in 2005 is presented in Figure 3. As in the previous experiment, a maximum amount of methane appears in May, with maximum concentrations decreased (as compared to the first experiment), by 17 % and amounted to 75 nmol/L. Further distribution of methane occurs according to a system of currents in the cyclonic direction, and by September, the maximum values of 55 nmol/L are determined in the Dm. Laptev Strait.

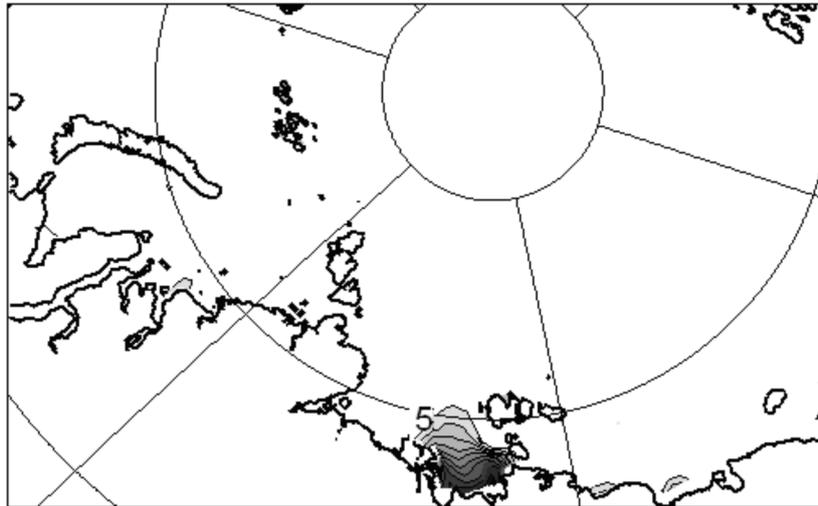


Figure 3. The spatial distribution of dissolved methane in nmol/L obtained for September 2008 in Experiment 2 (the maximum concentration of 55 nmol/L)

High concentrations of methane are stored in the shelf waters, but the transport in the central part of the Arctic does not happen. Methane isn't accumulated during a year because the methane oxidation processes are taken into account. So in May, high values of methane are defined only in the vicinity of the Lena river discharge, whereas in the first experiment, this period is characterized by the accumulation of methane in the whole area of the shelf.

3. In the third experiment, the methane oxidation was taken into account with allowance of the approach proposed in [21], where the term “turnover times” was introduced, as a measure characterizing the lifetime of methane

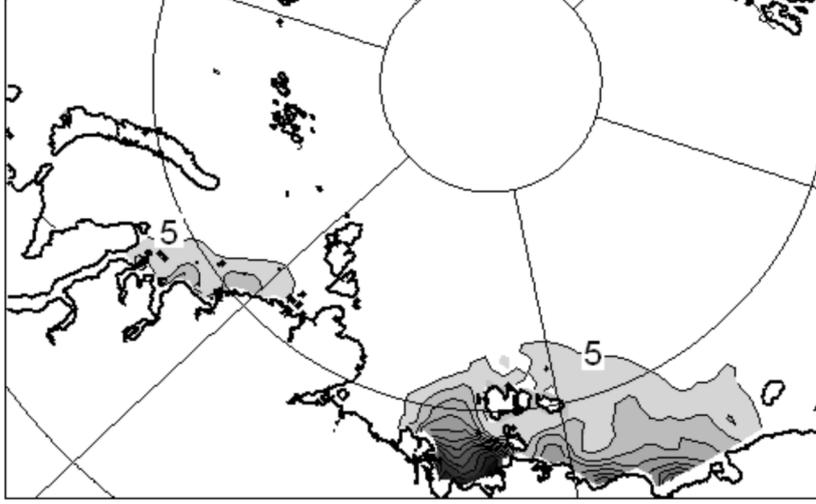


Figure 4. The spatial distribution of dissolved methane in nmol/L obtained for September 2008 in Experiment 3 (the maximum concentration of 65 nmol/L)

dissolved in the seawater. The rates of generation of methane in the deep seawater have been measured by several methods. The results of measurements of methane concentrations in the subbottom discharge area [21] show that its “lifetime” in the seawater is relatively constant: approximately 1.5 years for the deep layers of the ocean with high concentrations of CH_4 and tens of years, at shallow depths, where its concentration is much lower.

This parameterization was considered in [24]: the methane stock in the model in terms of its lifetime [21], which was 1.5 year at a depth of over 370 m and 10 years for a shallower ocean. In the experiment, we used a similar approach, namely, as follows:

$$C_{\text{ox}} = \frac{C}{\tau}, \quad \tau = \begin{cases} 1.5 \text{ year}, & z \geq 370 \text{ m}, \\ 10 \text{ years}, & z < 370 \text{ m}. \end{cases}$$

The resulting distribution of methane produced in this experiment in 2005 is presented in Figure 4. The concentration of dissolved methane in the surface layer reached 65 nmol/L in plume areas in September 2005. As in the second experiment, high concentrations are determined in the inflow area of the Lena River. In contrast to Experiment 2, a higher concentration of methane in the East Siberian Sea has grown up to 30 nmol/L due to the effect of the Indigirka and Kolyma.

4. Reasons for an apparent scaling of the removal time of methane have not been analyzed extensively in the literature. Although the spread of the methane removal time data is large for any concentration, typical time constants can reasonably be well defined empirically.

In [22], the model lifetime and removal are as follows:

$$\log_{10} \tau = 1 - \log_{10} C, \quad C_{\text{ox}} = \frac{C}{\tau} = 0.1C^2.$$

In these expressions, the units of measurements of τ and methane C are days and micromoles, respectively.

Figure 5 shows the distribution of dissolved methane in the surface layer for the Septembers of 2005 and 2008, obtained in Experiment 4. The con-

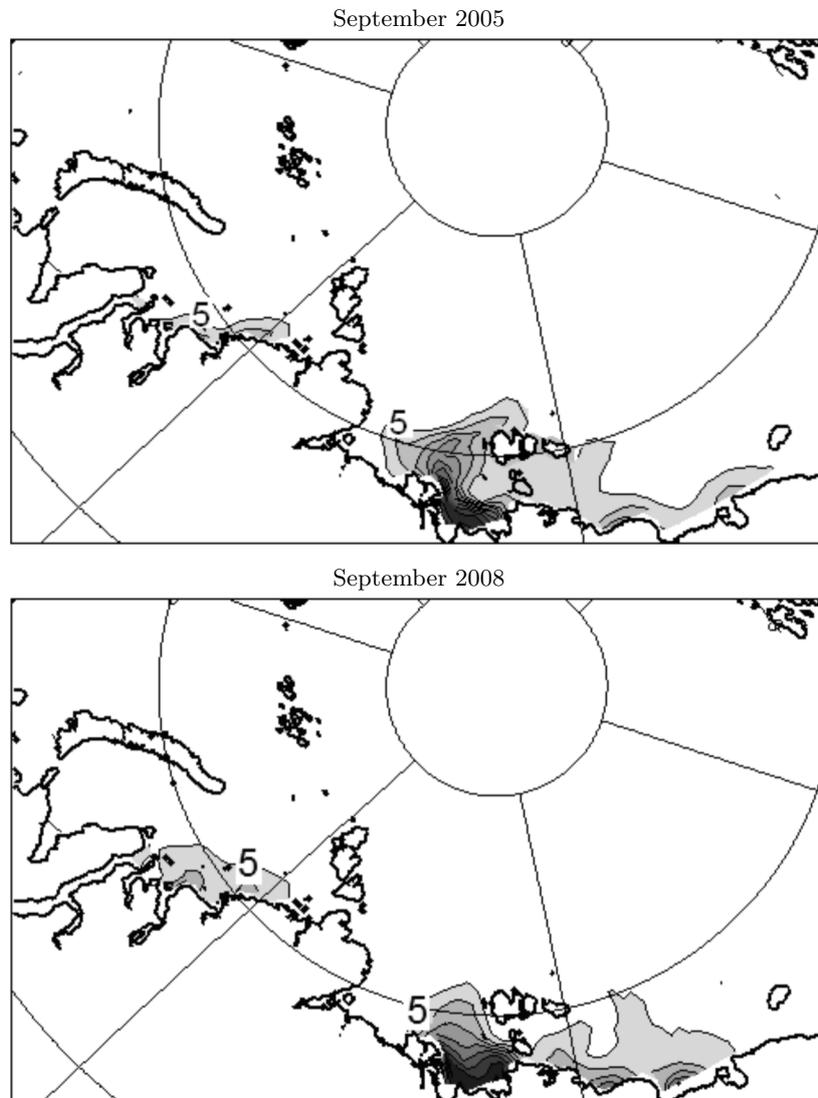


Figure 5. The spatial distribution of dissolved methane obtained in Experiment 4

centration of dissolved methane reached 55 nmol/L in the plume areas in September 2005 and 40 nmol/L in September 2008. The concentration has insignificantly decreased and the resulting distribution repeats that of Experiment 3.

5. Methane flux estimate. Saturation of surface waters by the dissolved methane allows calculating a methane flux in the atmosphere. We calculate the methane sea-air flux (F , g/(km²h)) from the equation proposed by Wanninkhof [25]: the rate of gas exchange between the ocean surface and the atmosphere is controlled by wind speed (V , m/s), sea-surface temperature (T , °C) and a difference between measured the methane concentration in the surface seawater (C_w , nmol/L) and equilibrium of methane solubility in the atmosphere (C_a , nmol/L)

$$F = 0.31V^2 \left(\frac{Sc}{660} \right)^{-0.5} (C_w - C_a).$$

The value of the Schmidt number for the methane dissolved in seawater was calculated for real temperature:

$$Sc = 2039.2 - 120.31 \cdot T + 3.4209 \cdot T^2 - 0.040437 \cdot T^3.$$

The atmospheric equilibrium solubility equation [26] is given by

$$\ln C_a = \ln f_G + A_1 + A_2 \cdot (100/T) + A_3 \ln(T/100) + A_4 \cdot (T/100) + S \cdot [B_1 + B_2 \cdot (T/100) + B_3 \cdot (T/100)^2],$$

where C_a is the equilibrium solubility in nmol/L, T is the surface absolute temperature (°C), S is salinity in ‰, f_G is molecular fractions in the dry air for methane ($1.41 \cdot 10^{-6}$), A_i and B_i are constants:

$$\begin{aligned} A_1 &= -415.2807, & B_1 &= -0.05916, \\ A_2 &= 596.8104, & B_2 &= 0.032174, \\ A_3 &= 379.2599, & B_3 &= -0.00482, \\ A_4 &= -62.0757. \end{aligned}$$

The above equilibrium can be used to calculate the atmospheric equilibrium solubility of any dissolved gas for different values of temperature, salinity, and atmospheric concentration.

The methane-water fluxes, with allowance for methane concentration field in the sea water in the period of the ice-free water were calculated. Below we present the fluxes obtained in Experiment 4 for September. There were obtained the fluxes of methane up to 63 g/(km²h) in 2003; up to 36 g/(km²h) in 2004; up to 50 g/(km²h) in 2005 and up to 40 g/(km²h)

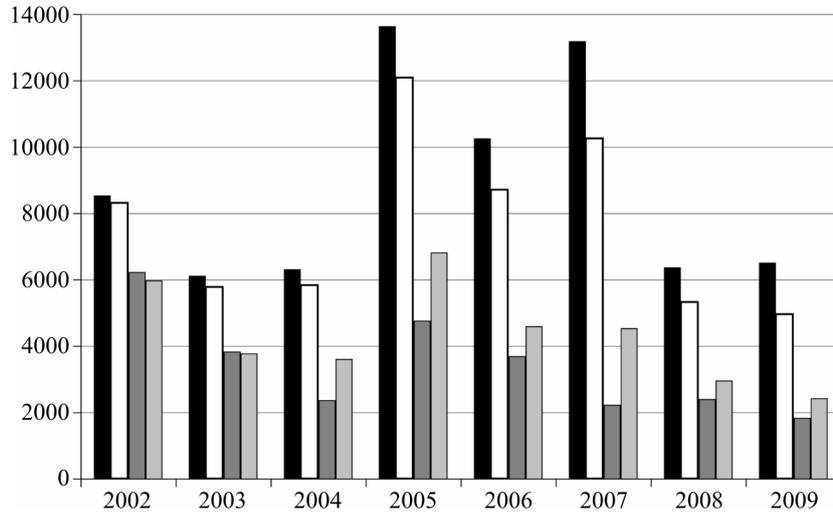


Figure 6. The total methane flux into the atmosphere (in tones) produced in all experiments for the summertime: black color — Experiment 1, grey color — Experiment 2, white color — Experiment 3, light-grey color — Experiment 4

in 2008, which is comparable with the observational data. The methane fluxes into the atmosphere of the East Siberian shelf, obtained with the use of the measured methane on the water surface are given in [9]: the average values for the Septembers of 2003 and 2004 are 4.86 and 3.02 g/(km²h), respectively. For the plume areas the flux can 3–15 times increase.

To estimate the contribution of the river runoff to the total methane emissions the integral fluxes of methane for the entire modeling domain were calculated. Figure 6 shows a diagram with the estimates obtained for the whole summer period, when there is no ice cover for all the experiments. It was found that the emission of methane into the atmosphere could be from 6,000 to 14,000 tons in Experiment 1, from 2,000 up to 6,000 tons in Experiment 2, from 5,000 up to 12,000 tons in Experiment 3 and from 2,000 up to 7,000 tons in Experiment 4, depending on the year under study. When taking into account the decreasing the flux of methane into the atmosphere oxidation process, was about 60 % (Experiment 2), about 15 % (Experiment 3), and 50 % (Experiment 4) as compared to Experiment 1. The results of Experiments 2 and 4 have shown that the use of the empirical relation [22] for the calculation of methane oxidation is consistent with accounting the oxidation rates, typical of this region.

The integrated flux of methane obtained in Experiment 4 for the entire period of the simulation is shown in Figure 7. A maximum flux obtained for the period of 2005–2007 was 4.5–7 kilotons. It can be seen that a significant contribution to the total methane flux is given by the Lena River runoff.

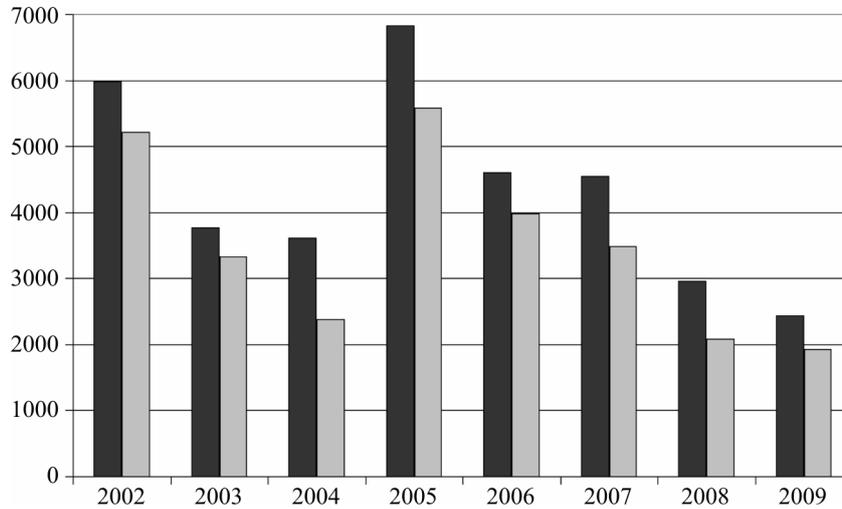


Figure 7. The total methane flux into the atmosphere (in tones) produced in Experiment 4 for the summertime: black color for all the rivers, grey color for the Lena River

4. Conclusion

We have managed to simulate the picture of dissolved methane accumulation processes, and time stability of its high concentration in the Laptev Sea, which is in accordance with the available observational data.

It is obtained that the drain of the Lena River can bring a powerful contribution to an abnormal high concentration of methane in the Dm. Laptev Strait water. The reason for the accumulation of methane in this area is a system of currents and the coastline. According to the dynamics of the river runoff it was found that the visible values of methane are in May, further distribution being in accordance with a system of currents, and by September, maximum values are determined near the Dm. Laptev Strait. This corresponds to the measurement data.

It is shown that methane oxidation controls the methane release into the atmosphere. The methane oxidation in the water column provides an efficient loss within the zone of distribution of a dissolved gas, which may essentially reduce its flux into the atmosphere. On the basis of the numerical experiments performed it was found that the simulation results may significantly vary depending on the parameterization of the oxidation processes.

The experiments allowed us to assess a possible emission of methane into the atmosphere, which can range from 2,000 up to 14,000 tons per year, depending on the considered oxidation parameterization.

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