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Methane fluxes at the water–atmosphere boundary in the waters of the Russian sector of the Eastern Arctic

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Abstract. The average value of methane fluxes from the sea surface in the Chukchi Sea (4±4 mol/(km^2·day)) and parameters of methane supersaturation of the surface water layer (78±39 %) were lower than in the East Siberian Sea (32±24 mol/(km^2·day) and 346±247 %, respectively). In 50 % of cases, the concentrations of dissolved methane in the surface layer of sea waters were two times higher than the equilibrium values with the atmosphere. The heterogeneous distribution of methane in seawater causes a change in the direction and magnitude of methane fluxes at the water–atmosphere boundary under given experimental hydrometeorological conditions. Data analysis showed that the flux was predominantly determined by wind speed (correlation coefficient Q = 0.8), concentration of dissolved methane (Q = 0.6), parameter of methane supersaturation of waters (Q = 0.6), and temperature of the surface water layer (Q = –0.6). A negative correlation coefficient with temperature indicates that as the temperature decreases, the solubility of methane in water increases, the difference in concentrations with the atmosphere decreases, and the intensity of methane flux decreases.

Keywords: methane fluxes, East Siberian Sea, Chukchi Sea, Arctic

Потоки метана на границе вода–атмосфера на акватории российского сектора Восточной Арктики

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Резюме. Средние значения потоков метана с морской поверхности в Чукотском море (4±4 моль/(км^2·сут)) и показателей пересыщения поверхностного водного слоя метаном (78±39 %) были ниже, чем в Восточно-Сибирском море (32±24 моль/(км^2·сут) и 346±247 % соответственно). В 50 % случаев концентрации растворенного метана в поверхностном слое морских вод в 2 раза превышали равновесные с атмосферой значения. Неоднородное распределение метана в морской воде вызывает изменения направления и величины потоков метана на границе вода–атмосфера при данных экспериментальных гидро- и метеоусловиях. Анализ данных показал, что поток определялся главным образом скоростью ветра (коэффициент корреляции Q = 0,8), концентрацией растворенного метана (Q = 0,6), показателем пересыщения вод метаном (Q = 0,6), температурой в поверхностном водном слое (Q = –0,6). Отрицательный коэффициент корреляции с температурой указывает на то, что при снижении температуры увеличивается растворимость метана в воде, уменьшается разность концентраций с атмосферой, и интенсивность потока метана снижается.

Ключевые слова: потоки метана, Восточно-Сибирское море, Чукотское море, Арктика

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Introduction

Methane, despite its insignificant concentration in the atmosphere, plays an important role in shaping the climatic conditions of planet Earth. The average concentration of methane in the atmosphere of the Earth's northern hemisphere is currently 1.8 ppm, with a tendency of constant increase. Moreover, methane is an indicator of hydrocarbon deposits and subsurface gas saturation. Tectonic faults in the Earth's crust in the waters of the Eastern Arctic form permeable zones for the migration of deep gases such as methane, hydrogen, helium, and others. According to the literature, the Arctic region is divided into two macro-regions, western and eastern. In the western sector, including the Barents and Kara Seas and the greater part of the Laptev Sea, the concentration of methane in the atmosphere in 2010–2022 was increased compared to the eastern sector, including the East Siberian and Chukchi Seas. This is due to natural factors, primarily the geological structure of the region [1].

The sources of methane in the Arctic are commonly known: oil and gas deposits; coal-bearing layers; gas hydrates; anthropogenic methane in oil and gas production areas; and methane carried by rivers and produced by microbial communities.

The Arctic seas are predicted to contain enormous hydrocarbon resources, which have not yet been estimated objectively. The considerable thickness of the sedimentary cover and high prospects for predicting the generation of catagenetic gases (including methane homologs) in the subsurface imply a significant contribution of lateral and advective methane flux from the lithosphere to the hydrosphere and atmosphere.

Methane resources of the Eastern Arctic and Eastern Russia with potential for industrial exploitation reach 4.4 trillion m^3 [2]. The areas of predicted oil and gas deposits on the shelf of the East Siberian Sea are characterized by the maximum thickness (greater than 3 km) of the sedimentary cover within the North Chukchi (greater than 10 km), South Chukchi, and Ayon Basins [3]. Methane concentration in coal fields in eastern Russia reaches 11 trillion tons [4]. More than 15 lignite and brown coal seams with thickness up to 7–10 m and methane content up to 2 m^3/t at depths of 200–250 m have been identified in the Chaun Depression of the Ayon Basin [2]. A large area of gas release in the form of bubble plumes was recorded in the East Siberian Sea [5]. Gas-saturated sediments stretch northward from Wrangel Island [6]. The 560-km gas geochemical sampling profile from Cape Billings to the Mendeleev Ridge, carried out in 2008, showed high methane content in bottom sediment cores for all 56 stations. Background methane concentration in bottom sediments was 13 ppm, which is four times higher than in the Sea of Okhotsk. In the central part of the profile, an area of gas-saturated sediments with hurricane methane content up to 24,000 ppm (2.4 vol.%) stands out [7]. Gas hydrates were found on the border of the East Siberian and Chukchi Seas [8]. The potential amount of gas in the subsurface of the Chukchi Sea in the form of hydrates is estimated to be from $7 \cdot 10^{11}$ to $11.8 \cdot 10^{13}$ m^3 [9].

The calculation of methane fluxes at the water–atmosphere boundary is particularly relevant in the study of climatic changes. Although studies of methane concentration and emission to the atmosphere in the Arctic have become one of the major areas of research, there is still substantial uncertainty in the magnitude of emission due to the spatial inhomogeneity of fluxes and the use of data based on different methodologies.

According to the 2011 studies, the average concentration of dissolved methane in the surface water layer in the Laptev Sea and the East Siberian Sea was 17.6±0.18 nmol/L; in the Chukchi Sea and the Bering Sea, it was 8.05±0.05 nmol/L

[10]. In the Chukchi Sea, based on the data from 2004, methane concentrations in surface water layer varied from 3 to 18 nmol/L, with a range of methane fluxes from 5 to 57 μmol/(m^2·day) [11]. Methane fluxes from the waters of the Alaskan Chukchi Sea shelf and the Central Trog of Chukchi Sea were 10.08 μmol/(m^2·day) [12]. The lowest fluxes of 1.9±1.4 μmol/(m^2·day) for the summer and fall of 2015 were recorded in the waters of the Bering Strait and the Alaskan Chukchi Sea shelf [13]. According to one of the recent estimates, fluxes from the surface of the Laptev, East Siberian and Chukchi Seas were 4.58, 1.74, and 0.14 mg/(m^2·day), corresponding to annual fluxes of 0.83, 0.62, and 0.03 Tg per year, respectively [14].

Annual estimates imply extrapolation of fluxes to the entire shelf and deep-water areas, although the data are often insufficient or non-existent, especially for ice-covered waters and gas seeping areas. Experimental data on the Chukchi Sea inner shelf and the eastern part of the East Siberian Sea are still insufficient to study the patterns and specific features of the contribution of regional fluxes.

This study aims to calculate methane fluxes at the water–atmosphere boundary using experimental data obtained in the summer of 2013 and to investigate the spatial distribution of the methane exchange rate for the inner shelf waters of the eastern sector of the Russian Arctic.

Materials and methods

To calculate methane fluxes at each point of surface water layer sampling, experimental data on dissolved methane concentrations, temperature, salinity in the surface seawater layer, methane content in the near-water atmosphere layer, and real wind speeds during sampling were used. These data were obtained in the waters of the inner shelf of the eastern sector of the Russian Arctic in the course of the research and educational expedition on the training vessel *Professor Khlyustin* (July–September 2013).

The data obtained on the R/V *Akademik M.A. Lavrentiev* (LV45, 2008) were used in the discussion of the results.

Sampling of water, sediment, and air and analytical studies were carried out in accordance with the certified methodology adopted by the Laboratory of gas geochemistry of the POI FEB RAS (Certificate of Rosstandart No. 41 to the Laboratory Passport PS 1.047–18). In the Eastern Arctic, surface water samples were collected underway using an intake device at a depth of 5 m from the sea surface. At four hydrological stations, water samples were taken using Niskin bottles from the surface, near-bottom, and intermediate horizons. A hydrostatic sampler with a length of 350 cm was used to sample bottom sediments. The sediment was sampled using syringes, with a sampling step of 10 cm.

Gas concentrations in water and in sediments were determined by the equilibrium concentration method.

Chromatographic analysis of gas composition was performed on the Crystallux-4000M chromatograph (RPC "Meta-chrom", Co. Ltd, Yoshkar-Ola), equipped with a flame ionization detector and two thermal conductivity detectors; helium was used as a carrier gas.

Concentrations of methane dissolved in seawater were estimated using the calculated constants of methane solubility [15] in modification [16].

Methods for calculating methane fluxes at the water–atmosphere boundary. Methane fluxes at the water–atmosphere boundary in the East Siberian and Chukchi Seas were calculated taking into account the influence of surface microlayer properties on the gas exchange mechanism using the methodology described in the studies [17–19]:

$$F = \Delta C \cdot K, \quad (1)$$

where F is intensity of the methane flux, ΔC is the difference between the dissolved methane concentration and the methane concentration in equilibrium with the atmosphere, and K is the gas exchange coefficient at the water–atmosphere boundary.

The degree of supersaturation N (%) was calculated for each sample according to the following equation:

$$N = (\Delta C / C^*) \cdot 100, \quad (2)$$

where C^* is the equilibrium methane concentration.

Due to the strong variability of methane fluxes at the water–atmosphere boundary, ten gradations of flux values were proposed [20] as follows, mol/(km²·day): strong runoff (–6… –1), weak runoff (–1… –0.01), equilibrium (–0.01… 0.01), low-intensity emission (0.01–1), moderate-intensity emission (1–4), medium-intensity emission (4– 10), high-intensity emission (10–20), very high-intensity emission (20–50), abnormal-intensity emission (50–150), and hurricane emission values (150–500) [20].

Results and discussion

Uneven distribution of methane fluxes was observed from the sea surface of the inner shelf along the ship route in the summer of 2013 (Fig. 1, Table 1).

The studied water area is a source of methane input to the atmosphere. Within the proposed gradation, methane emissions varied from equilibrium values to values of abnormal intensity.

At the stations located in the water area of the Chaun Bay of the East Siberian Sea, fluxes of very high and abnormal intensity of 39 and 62 mol/(km²·day) were recorded, the supersaturation was 258 and 316 %. Near the Long Strait, the high-intensity flux was 20 mol/(km²·day), and the supersaturation was 694 %. Local zones of increased methane fluxes from the water area agree with high concentrations of dissolved methane in the surface water layer. Dissolved methane concentrations in the surface water layer in the study area varied from 4.1 to 27.1 nmol/L. The difference between the dissolved and equilibrium methane concentrations was positive, from 1 to 24 nmol/L. The supersaturation of the surface waters with dissolved methane relative to the atmosphere was from 33 to 694 %.

The average values and variation range of methane flux intensity at the water–atmosphere boundary, dissolved methane concentrations in the surface water layer, methane supersaturation parameters, and wind speeds for the East Siberian and Chukchi Seas are presented in Table 2.

The average value and variation range of methane fluxes at the water–atmosphere boundary in the East Siberian Sea were higher than in the Chukchi Sea. The range of methane fluxes from the Chukchi Sea water area was less than that given in [11] for 2004. The methane flux from the sea surface of the inner shelf of the Chukchi Sea in the summer of 2013 was two times lower, whereas the flux of the East Siberian Sea was three times lower than that given in [14] for 2014.

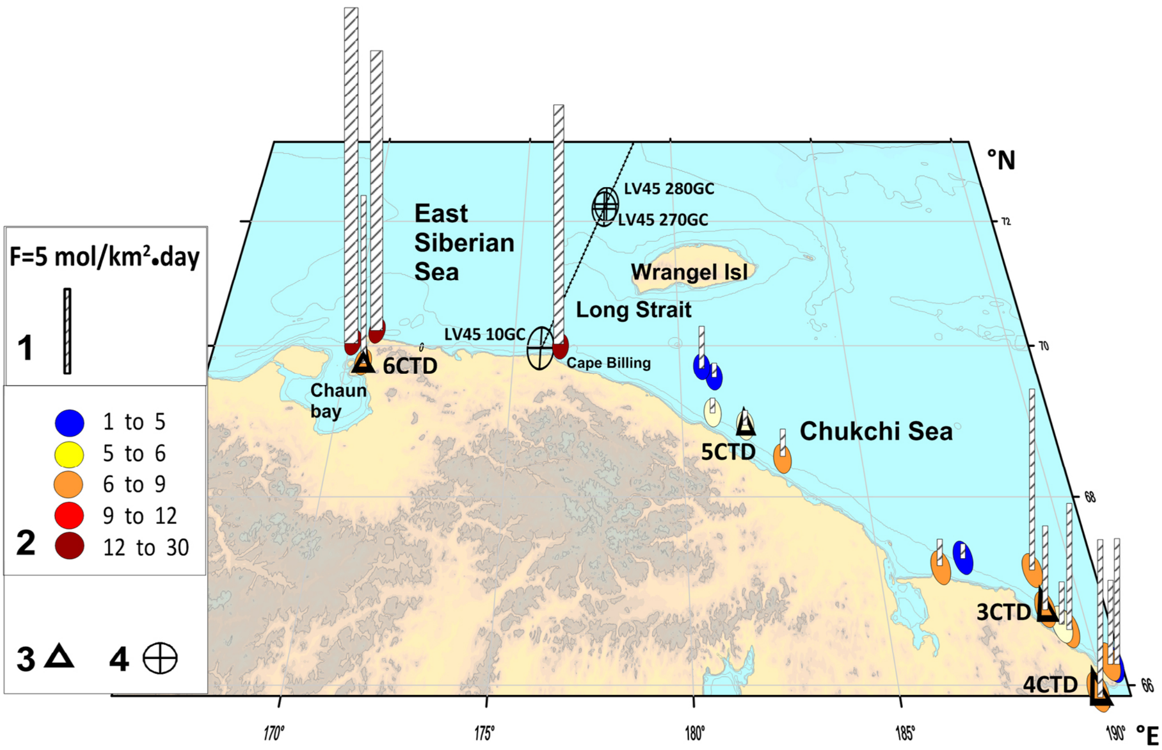

Fig. 1. The distribution of dissolved methane concentrations and methane fluxes on the water–atmosphere boundary within the studied area. 1 – methane fluxes on the water–atmosphere boundary, *F* (mol/(km²·day)); 2 – concentrations of dissolved methane (nmol/L); 3 – locations of CTD stations; 4 – locations of sediment sampling stations.

Pairwise correlation coefficients (Q) of the studied values are presented in Table 3.

Data analysis showed that the flux was primarily determined by wind speed *U* (correlation coefficient Q = 0.8); flux correlation coefficients with dissolved methane concentration *C* (Q = 0.6), methane supersaturation parameter *N* (Q = 0.6), and temperature *t* in the surface water layer (Q = –0.6) were also statistically significant. That is, as the wind speed, dissolved methane concentration, and water supersaturation parameter increase, the flux intensity increases. The correlation coefficient has an opposite relationship with temperature, indicating that as temperature decreases, the methane solubility in water increases, the concentration difference with the atmosphere decreases, and the methane flux intensity decreases.

The vertical distribution of dissolved methane concentrations at stations CTD6 in the East Siberian Sea, CTD5 and CTD3 in the Chukchi Sea, and CTD4 in the Bering Strait is shown in Fig. 2.

Table 1. Characteristics of methane fluxes and related parameters for the studied water area

Water area	*F*	*C*	*t, C°*	*S*	*C**	*ΔC*	*N*	*U*	Station No.
The Bering Strait	10.3	6.1	5.7	30.0	3.3	2.8	85	9	CTD 4
	7.1	4.1	7.1	31.4	3.0	1.1	36	13	
	5.1	6.1	6.0	31.7	3.1	3.0	97	5	
	7.2	8.1	4.3	31.9	3.4	4.7	137	5	
The Chukchi Sea	6.3	7.0	4.9	31.3	3.2	3.9	121	5	CTD 3
	1.1	5.2	2.5	29.0	3.5	1.7	48	2	CTD 5
	3.2	5.1	5.2	31.7	3.2	1.9	60	5	
	12.1	8.1	3.9	32.4	3.3	4.8	148	7	
	0.6	4.1	6.1	30.6	3.1	1.0	33	1	
	1.9	6.2	6.3	29.1	3.1	3.0	97	1	
	2.3	6.2	1.3	25.4	3.7	2.5	68	3	
	0.6	4.1	6.5	30.4	2.9	1.2	42	0	
	1.1	5.2	4.8	27.6	3.3	1.9	58	1	
	2.8	4.1	6.5	30.5	2.9	1.2	42	6	
The East-Siberian Sea	7.7	6.3	8.8	21.8	2.9	3.4	116	6	CTD 6
	19.7	27.1	0.7	30.5	3.4	23.7	694	2	
	38.9	12.1	0.8	31.8	3.4	8.7	258	10	
	61.6	14.1	0.7	31.8	3.4	10.7	316	12	

Notes. Here and in the tables 2 and 3: *F* – intensity of the methane flux on the water–atmosphere boundary, mol/(km^2·day); *C* – concentration of dissolved methane in the surface water layer, nmol/L; *t* – temperature, C°; *S* – salinity of the surface water layer, psu; *C** – equilibrium methane concentration, nmol/L; Δ*C* – difference between the dissolved methane concentration and the equilibrium methane concentration, nmol/L; *N* – parameter of methane supersaturation of water layer, %; *U* – wind speed, m/s.

Table 2. Average values of characteristics of the methane fluxes in 2013

Water area	Average value (variation range)			
	F, mol/(km^2·day)	*C*, nmol/L	*N*, %	*U*, m/s
The East-Siberian Sea	32±24 (62–8)	15±9 (27–6)	346±247 (694–116)	8±4 (12–2)
The Chukchi Sea	4±4 (12–1)	6±1 (8–4)	78±39 (148–33)	4±3 (9–0)

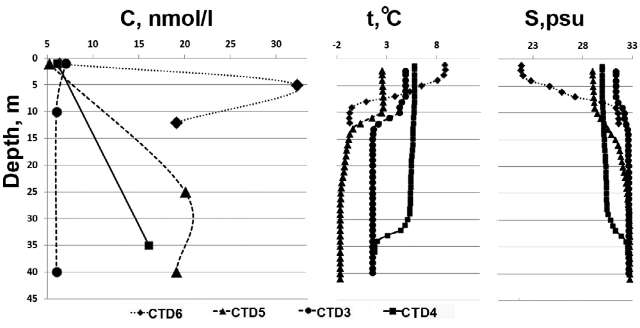

Fig. 2. Vertical distribution of dissolved methane concentrations (C, nmol/L), temperature (t, C°), and salinity (S, psu) in the East Siberian Sea (station CTD6), Chukchi Sea (stations CTD3, CTD5), and in the Bering Strait (station CTD4).

Inhomogeneity in the distribution of dissolved methane concentrations was observed on vertical profiles. At all stations, except for the station CTD3 in the Chukchi Sea, increased concentrations in the near-bottom layer were recorded. The maximum dissolved methane concentration at the 6 m horizon at the shallow-water station CTD6 in the coastal zone of the East Siberian Sea was 32 nmol/L. In samples collected at different horizons at the station CTD5 in the central part of the Chukchi Sea shelf, abnormal methane concentrations of approximately 20 nmol/L were observed at 25 and 40 m horizons. When comparing the maxima of chlorophyll-a and colored dissolved organic matter contents given in [21] with the maxima of methane content, no significant agreements were found. The agreement for all three parameters is observed in the near-bottom layer at the station CTD4 and at the 25 m horizon below the layer of changes in thermohaline parameters at the station CTD5; dissolved methane and colored dissolved organic matter concentrations agree in the surface layer at the station CTD3, and at the station CTD6 they agree above the layer of changes in thermohaline parameters at the 6 m horizon, while dissolved methane and chlorophyll-a concentrations agree in the near-bottom layer. No clear pattern confirming the methane production during activity and destruction of hydrobionts was found, probably due to the small amount of data.

Table 3. Correlation coefficients between methane fluxes values and related parameters

	F	C	t	S	C^*	ΔC	N	U
F	1.0							
C	0.6	1.0						
t	–0.6	–0.6	1.0					
S	0.3	0.2	–0.3	1.0				
C^*	0.3	0.4	–0.8	0.1	1.0			
ΔC	0.6	1.0	–0.6	0.2	0.3	1.0		
N	0.6	1.0	–0.6	0.2	0.3	1.0	1.0	
U	0.8	0.2	–0.3	0.2	0.1	0.2	0.2	1.0

Since the waters at the stations in the Russian sector of the Eastern Arctic during the period of studies were characterized by a two-layer structure (with a water layer of reduced salinity in the surface layer, see Fig. 2), the transfer of dissolved methane from the water to the surface water layer and further to the atmosphere was limited [21]. At the profile along the inner shelf, the dissolved methane concentrations in the surface layer were two times higher than the equilibrium values with the atmosphere both for four hydrological stations and for the majority of the shelf stations. The exception was three stations in the East Siberian Sea, at which the dissolved methane concentrations were nine and four times higher than the values in equilibrium with the atmosphere (see Table 1).

The maximum dissolved methane concentration in the surface water layer was 27.1 nmol/L, and the maximum methane supersaturation of waters was 694 % at the station near Cape Billings (see Table 1). The sediment sampling station LV45-10 is located nearby and marks the beginning of the profile carried out in 2008. The verti-

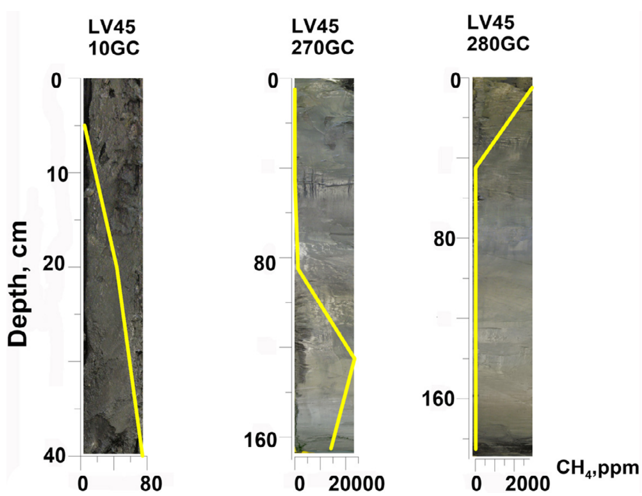

Fig. 3. Distribution of methane concentrations in sediment cores at stations LV45-10, LV45-270, LV45-280, R/V *Akademik M.A. Lavrentyev* (cruise No. 45).

cal methane distribution in bottom sediment cores for the station LV45-10 and two stations in the central part of the profile with abnormally high methane content, LV45-270 and LV45-280, is given in Fig. 3.

The high regional hydrocarbon background and hurricane values of methane content in sediment cores as a result of long-term natural gas seepage may have led to dissolved methane concentrations exceeding equilibrium values with the atmosphere.

Thus, the observed increased concentrations of dissolved methane in the near-bottom and surface water layers and abnormally high values of methane content in bottom sediment cores probably indicate the presence of geological sources from which methane migrates to the upper layer of marine sediments and then enters the water column and the atmosphere.

This conclusion is consistent with the results of gas-geochemical studies of bottom sediments and geological structures of the East Siberian Sea. The studies [22, 23] show that hydrocarbon gases of the southeastern part of the East Siberian Sea are represented by biogenic gases of modern sediments and peat bogs; metamorphogenic gases of coal-bearing formations, gas deposits, solid bitumen, presumed gas hydrate, gas condensate, oil and gas deposits; and gases of magmatic formations.

Conclusions

Studies of methane exchange rates at the water–atmosphere boundary revealed that the water area of the inner shelf of the Russian sector of the Eastern Arctic in the summer of 2013 was a source of methane input into the atmosphere. The flux from the sea surface varied from an equilibrium of 0.6 mol/(km^2·day) in the Chukchi Sea to an abnormal-intensity emission of 61.6 mol/(km^2·day) in the East Siberian Sea.

Correlation analysis of the data showed that wind speeds, dissolved methane concentrations, methane supersaturation of surface waters, and temperature have a major influence on the flux (correlation coefficients Q = 0.8; Q = 0.6; Q = 0.6; Q = –0.6, respectively). The inhomogeneous methane distribution in surface seawater and the strong dependence of the gas transfer coefficient on wind speed cause changes in the magnitude of methane fluxes at the water–atmosphere boundary.

The vertical methane distribution in seawater is inhomogeneous, and abnormal methane concentrations can be observed in the lower horizons of the water column, possibly entering the water column from marine sediments, including underwater geological sources.

Dissolved methane concentrations in the surface seawater layer in 50 % of cases were two times higher than the equilibrium values with the atmosphere. Dissolved methane concentrations in the surface layer were lower in the Chukchi Sea than in the East Siberian Sea. The average value of methane supersaturation of waters and its variation ranges were also lower in the Chukchi Sea (78±39 %) than in the East Siberian Sea (346±247 %).

The average value of methane fluxes to the atmosphere from the sea surface in the East Siberian Sea with higher methane concentrations and higher wind speeds was greater than in the Chukchi Sea (32±24 and 4±4 mol/(km^2·day), respectively).

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