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Methane in the atmosphere and surface water of Lake Baikal

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ABSTRACT

Long-term observation series of the methane content in the near-water atmosphere and in water in the coastal zone of Lake Baikal are analyzed. The measurements were carried out in the Baikal Atmospheric-Limnological Observatory (BALO SB RAS, which is a part of the Resource Sharing Center "Atmosfera"). A steady increase of the methane content in the atmosphere by about 12 ppb per year is observed. The average diurnal methane content in March 2021 (the last measurement cycle) was 2.012 ppm. For the period of ling-term observations, the measured partial methane pressure in the surface water always exceeded that in the atmosphere. This fact indicates that this part of Lake Baikal is a source of methane into the atmosphere. For the coastal zone, the average value of methane flux from water was 380$\mu gCH_4\ m^{-2}\ day^{-1}$. The observed increased methane concentration in the near-bottom layer indicates the predominance of bottom methanogenesis. Our data show that the increment rate of dissolved methane in surface water of Lake Baikal is 0.015 μgCH_4/l per year.

Keywords: Atmospheric methane, dissolved methane, methane trends, methane diurnal pattern, methane seasonal pattern, methanogenesis, Lake Baikal

1. INTRODUCTION

Atmospheric methane plays an important role in formation of the climate. The sources of atmospheric methane are on the ground, while methane destruction proceeds in the atmosphere. A significant part of methane is of biogenic origin, i.e. associated with the vital activity of microorganisms. Methane is usually generated under anaerobic conditions in the earth (and oceanic) crust. The conditions, under which the vital activity of methanogenic microorganisms (archaea) is possible, exist, in particular, in the bottom sediments of Lake Baikal[1]. Despite this, the methane content in the aerobic water column of Lake Baikal decreases from the surface to the bottom, and in the main, deep part of the water column, the methane concentration is lower than that balanced with the atmosphere[2,3]. In the surface water, the pressure of dissolved methane, according to our findings, always exceeds that in the atmosphere[4], although with rare exceptions[5].

This paper presents the data on the methane content in the near-water atmosphere and the surface water as obtained in the Baikal Atmospheric-Limnological Observatory during 25 measurement campaigns since 2012 till the time of this writing in 2021. In our judgments concerning methane dissolved in water, we also refer to the spatial data obtained by us during the Circum-Baikal campaigns[5].

2. MEASUREMENT SITE AND TECHNIQUE

We started stationary methane measurements at Lake Baikal in July 2012. By March 2021, we have 23 series of observations from 10 to 14 days long at the coastal site. The total duration of observations is 4000 hours. The measurements were conducted in the Baikal Atmospheric-Limnological Observatory (BALO SB RAS, a part of the Resource Sharing Center "Atmosfera"). BALO is located at the western coast of the southern part of Lake Baikal (51.90°N and 105.06°E)[6]. A BALO observation post is located on a pier (see figure 1). Air and water are sampled at a distance of about 40 m from the coastline. Water is sampled continuously with vibration pumps installed at a distance of 3-5 m from the pier toward open water (up to 30 m in the spring period, when the pumps are installed from ice). The pumps are set one above another: on the surface (30-50 cm from the water surface) and on the bottom (5 cm from the bottom). The depth here is 3 to 4 m.

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Methane fluxes in the water-atmosphere system are measured with a semi-submersible chamber installed in the immediate vicinity of the water intake pumps. The chamber is ventilated every hourly cycle by lifting above the water surface.

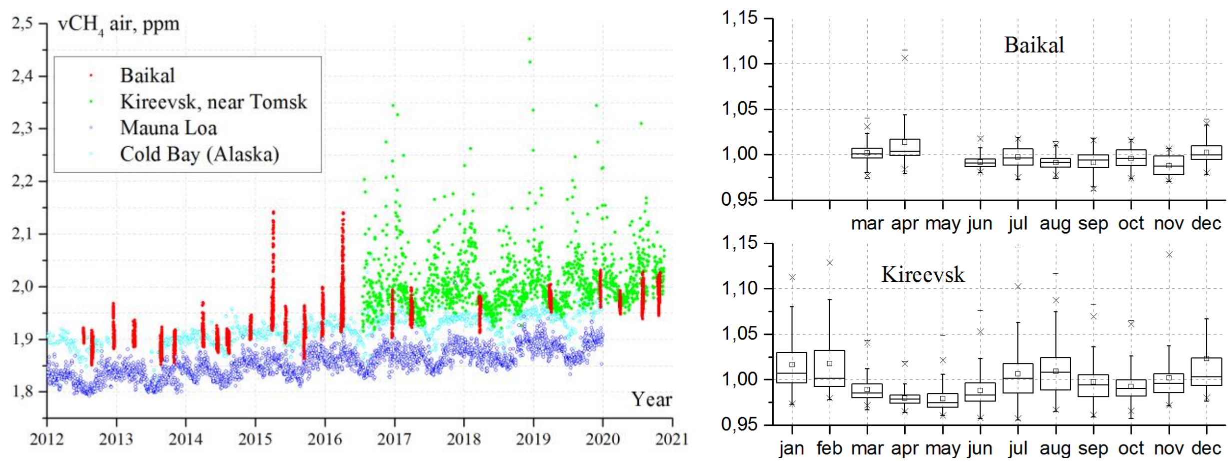
The methane mixing ratio in the atmosphere is measured with the Picarro G2301 gas analyzer (inherent drift in methane data less than 2 ppb a year). The partial pressure of methane dissolved in water is measured by means of flow-through equilibrators by the same gas analyzer (fig. 1, middle photo). The methane concentration in water is calculated from its equilibrium partial pressure (flowing water–air in the equilibrator) with allowance for the solubility at the water temperature at the time of measurement. The detailed description of the BALO instrumentation and operating modes can be found in [5].

Fig. 1. Baikal Atmospheric-Limnological Observatory (BALO) on the pier; BALO interior: equilibrator and gas-analysis equipment; Semi-submersible chamber in the accumulation mode.

3. RESULTS AND DISCUSSION

3.1. Methane in the atmosphere

From the start of our observations in June 2012 to the last measurement cycle in March 2021, the methane content in the atmosphere above Lake Baikal increased, and the average increment rate of the mixture ratio was 12 ppb per year. This trend is characteristic of Western Siberia, where the increment rate varies within 5.7-12.1[7], and is higher than 9 ppb/year according to the data of Cold Bay Observatory, Alaska (located the same latitudes as BALO). The absolute methane content as of October 2020 reached 2 ppm, and its average diurnal values in October 2020 and March 2021 (last measurement cycle) were 1.992 and 2.01 ppm, respectively, which is lower than over the territory of Western Siberia (Kireevsk village, Tomsk region) in the same period (2.026 ppm), but exceeds the data of the observatory in Alaska (see fig. 2a).



a) b)

Fig. 2. (a) Many-year series of atmospheric methane mixture ratio as measured in BALO (red dots), Kireevsk village, Western Siberia (green dots, data provided by the Laboratory for Climatology of Atmospheric Composition of IAO SB RAS, http://lop.iao.ru), volc.

Mauna Loa (Hawaii, blue dots)[8], Cold Bay site (Alaska, cyan dots)[9]; (b) Seasonal profile of the methane content in the atmosphere as measured in BALO and Kireevsk village. Data are normalized to the average value for the corresponding month and pre-detrended. Hereinafter, the upper, central, and lower boxes correspond to 75%, median, and 25%; the square inside a box is for the average value.

The seasonal profile of the methane content in the Baikal atmosphere is not clearly pronounced as judged from the available data (fig. 2b). At the same time, in the West-Siberian region the annual profile is most clearly seen and associated with the anthropogenic impact (winter maximum) and with intensified biogenic generation in the warm season from wetlands, which are the predominant landscape in this region[10]. Thus, we can conclude that despite the predominant western transport, the mentioned sources of the neighboring region do not affect the Baikal atmosphere.

It should be noted that BALO is located at the interface of two ecosystems. Correspondingly, incoming air masses take origin both from land (forests), mainly in the evening and at night, and from the lake. The diurnal profile of atmospheric methane has a slightly pronounced maximum (smaller than 0.5% of the average value), which took place in the daytime (fig. 3). At this time, maritime air masses prevail in the coastal zone. It can be noted that over wetlands, a clearly pronounced maximum of the methane content in the atmosphere is observed at night, which is associated with inversion locking and a decrease in methane emanation from the surface layer[10]. At the Baikal site, the nighttime minimum is indicative of the absence of significant methane sources from the land side.

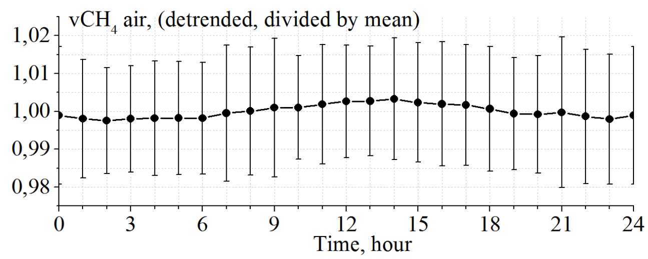


Fig. 3. Diurnal profile of the methane content in the atmosphere (BALO). The data are normalized to the average value for the corresponding hour and pre-detrended.

3.2. Methane in water

As to the content of dissolved methane in the surface water of Lake Baikal, the partial methane pressure in water during our observations both in the coastal zone and in the open water area always exceeded the atmospheric one. Thus, we can conclude that the water area of the lake during the ice-free period is a source of methane into the atmosphere.

In the coastal zone, the supersaturation of dissolved methane relative to the atmosphere varies from 136% to 3500%, and its variability in time is spontaneous. Peaks of methane content are not in any way associated with a specific time of day, but rather are caused by the transport of methane-enriched water masses by circulation currents from the places of its emission from bottom sources.

Dissolved methane, in particular in Lake Baikal, in its origin can be both biogenic, that is, formed as a result of the vital activity of microorganisms in places of accumulation of organic matter under anaerobic conditions[1], and thermogenic, that is, coming from Paleogene sediments[11,12].

In the bottom horizon, the observed methane concentration is higher than that near the surface by an average of 25%. In the littoral zone, the predominant sources of methane inflow into water are located in bottom sediments. At the BALO region, the landscape is so that bottom sources of methane are not uniformly distributed, but localized in zones within a narrow shallow strip along the coastline.

Figure 4 shows the diurnal and seasonal distributions of the dissolved methane concentration in the bottom and surface layers. Despite spontaneous oscillations of the dissolved methane concentration in time, the analysis of long series reveals the diurnal and seasonal profiles, which are most pronounced near the bottom.

The seasonal profile is characterized by the maximum content of dissolved methane in the under-ice and post-ice periods. Before the ice cover melts, the methane accumulation in the water column is not distorted by intense mixing with waters having the low CH_4 content from the deep layers. In surface water, the seasonal and diurnal profiles of the dissolved methane concentration are nearly absent.

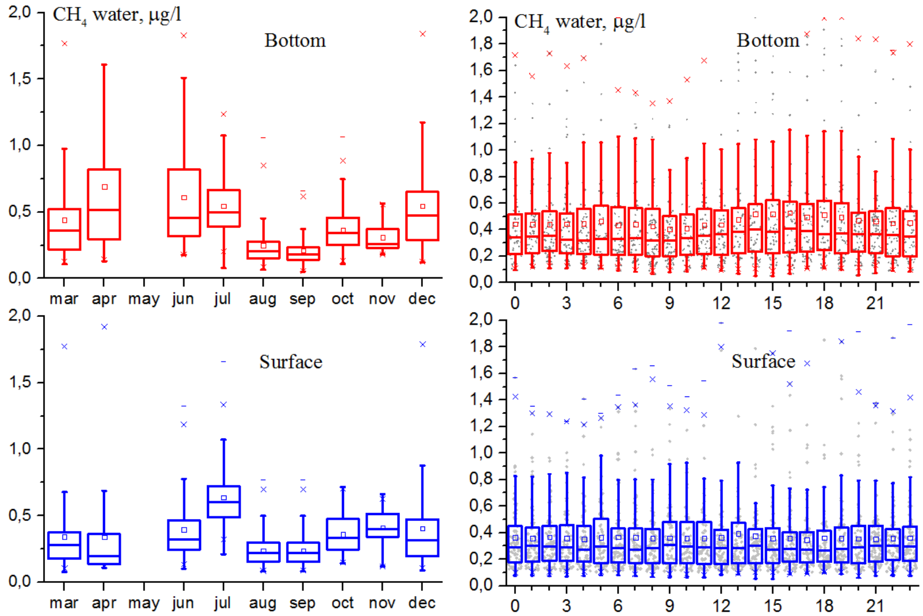


Fig. 4. Seasonal and diurnal profiles of the dissolved methane concentration in the bottom and surface layers in BALO (the depth difference between the bottom and surface water intakes is 2-3 m).

The long-term series of the methane content in water of the coastal zone demonstrates an increase in the methane mean concentration from 0.332 μgCH_4/l (surface) and 0.417 μgCH_4/l (bottom) in the period 2012-2015 to 0.398 μgCH_4/l (surface) and 0.504 (bottom) μgCH_4/l in the period 2016-2020. In this assessment we didn't use data of "March 2021" campaign, when the abnormally high concentrations of dissolved methane were registered.

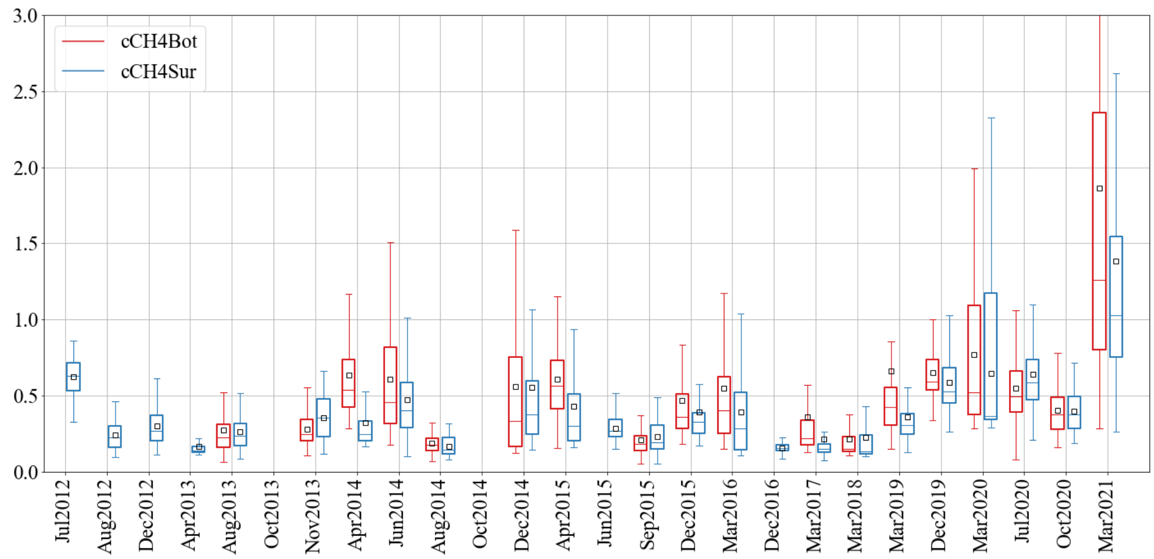


Fig. 5. Temporal profile of the methane content in water: initial data (red and blue dots). Boxes as described in figure 2.

According to the conclusions of the authors[2], for the period from 2002-2004 to 2016-2018 the dissolved methane content increased two to three times in the water column and 1.2–1.7 times in the surface layer of the southern and central basins of Lake Baikal. The data of our observations show that the increment rate of dissolved methane in the surface layer is 0.013 μg/l per year and 0.017 μg/l per year for the near-bottom layer. These values are consistent with those reported in the paper[2]. At the same time, for carbon dioxide, whose content in the atmosphere also grows at a rate of 2 ppm/year, no increase in the content in the Lake Baikal water has been observed over the past 15 years of observations[13].

3.3. Methane fluxes in the water-atmosphere system

The methane flux in the water-atmosphere system as calculated from the rate of change of the methane content in the ventilated chamber is always positive, that is, directed from the water into the atmosphere. The average value of the flux was 380 $\mu gCH_4\ m^{-2}\ day^{-1}$. The methane solubility in water does not exceed the ratio 0.055 L_{CH4}/L_{H2O}*, that is, the emission of even a small amount of methane from water into the atmosphere decreases its partial pressure in the upper layer to a significant extent. Therefore, despite the manifold supersaturation of surface waters with methane, the methane flux from the water rarely exceeds 1.0 $mgCH_4\ m^{-2}\ day^{-1}$. The diurnal profile of the intensity of methane emission into the atmosphere (fig. 6) has maxima nearby 06:00 and 15:00 Local Time. An increase in the rate of methane unloading into the atmosphere is determined by an increase of its water content and by the wind effect in daytime.

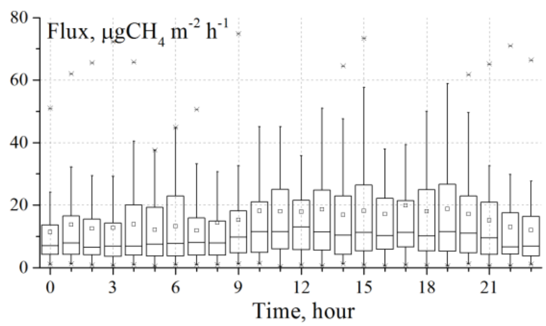


Fig. 6. Diurnal variation of methane fluxes from the water surface (a total of 1380 measurements).

4. CONCLUSIONS

The methane content in the atmosphere of the Baikal region has been shown to increase. In the absolute values and in the increment rate, it corresponds to global values at this latitude. The diurnal profile is slightly pronounced (less than 1%) and likely caused by surges of different air masses to the measurement site with the periodicity of breeze circulation. Maritime air masses bring more methane-enriched air to the coastal zone. The content of methane dissolved in water at the observation site varies widely from 0.06 μg/l to 8.5 μg/l. In all the seasons, the partial pressure of methane in water exceeds that in the atmosphere. In the under-ice water, supersaturation reaches 1300%. This probably leads to powerful methane unloading into the atmosphere in the period of melting of the ice cover. For the period of observation since 2012 till now, the content of dissolved methane increases from 0.32 μg/l (in 2012-2013) to 0.43 μg/l (in 2019-2020), that is, by 0.015 μg/l/year.

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* Maximal methane solubility is achieved at the minimal water temperature, that is, as 0 °C

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