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RADIATION AND BIOSPHERE

Spatial Distribution of Methane Concentration in Baikal Surface Water in the Spring Period

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Abstract—The results of describing the spatial distribution of methane concentration in the surface water of Lake Baikal in the spring are presented. The basis was the measurements of CH_4 content which were first carried out in the round-the-clock continuous mode along the entire route of the passage of research vessel in the complex expeditions of Limnological Institute, Siberian Branch, Russian Academy of Sciences, in the spring seasons of 2013, 2016, 2017, 2018, 2021, and 2022. Based on the results of six expeditions, a merged data array was compiled; it included 12 100 segments (with a step of 0.005° in latitude and 0.01° in longitude) which covered the total area 4466.7 km^2, or 14% of the surface of Lake Baikal. For a more detailed description of the spatial distribution of methane concentrations in surface water throughout the Baikal water area, the statistical characteristics were calculated in four zones: between 0 and 100 m, 100 and 200 m, 200 and 400 m, and over 400 m isobaths. The comparison of the methane concentrations in the analyzed array with the data of other researchers obtained in different years in nearby regions of the water area made it possible to conclude that the results presented in the work adequately reflect the most stable features of the spatial distribution of methane concentration in surface water of Lake Baikal in spring seasons.

Keywords: Lake Baikal, greenhouse effect, climate, concentration, methane, surface water, oxygen, biogenic elements, partial pressure, equilibrator, isobaths

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INTRODUCTION

Methane is an integral component of carbon cycle in the Earth's natural system [1–3]. It is a greenhouse gas which absorbs intensely the Earth's thermal radiation in the infrared region of the spectrum [4–6]. In recent years, in selecting priority measures of preventing possible negative consequences of climate change, marked attention has been devoted to methane due to the idea that anthropogenic emissions of "short-lived factors of climate change" are easier to regulate and reduce [7, 8]. The sources of methane entering the atmosphere are many but the largest part of methane is of biogenic origin [1]. The poor representativeness of data of field measurements has the consequence that model estimates of the natural methane sources are characterized by large dispersion even on a global scale [3, 4]; large uncertainties are moreover noted at the regional level [6, 9].

Considering the uniqueness of the aqueous ecosystem of Baikal, which is over 31.5 thousand km^2 in area, and large drainage territory (of the order of 570 thousand km^2) [10], it can be speculated that the lake is one of the significant natural methane sources in the region. In 1990s, in Baikal the studies began of the processes of methane formation in bottom sediments [11] and of the distribution of methane concentration in water and bottom sediments [12], and gas hydrates at the lake sediment depth were detected [13, 14]. To date, specialists have a large amount of information on the spatial distribution of methane content in the surface water [15], CH_4 sources and sinks [16–18], processes of formation of hydrocarbon gases by microbial community in bottom sediments [19–21], and on the vertical profile of methane concentration in the water column [22]. It should be noted that all data were obtained from analysis of water samples, collected during stops of research vessels at individual stations.

The purpose of this work is to analyze the spatial distribution of methane concentration, using continuous measurements along the route of passage of a research vessel in the Baikal basin in the spring periods of 2013, 2016, 2017, 2018, 2021, and 2022.

MATERIALS AND METHODS OF STUDY

Measurements

In 2013, we began for the first time the continuous round-the-clock measurements of the CO_2 and CH_4

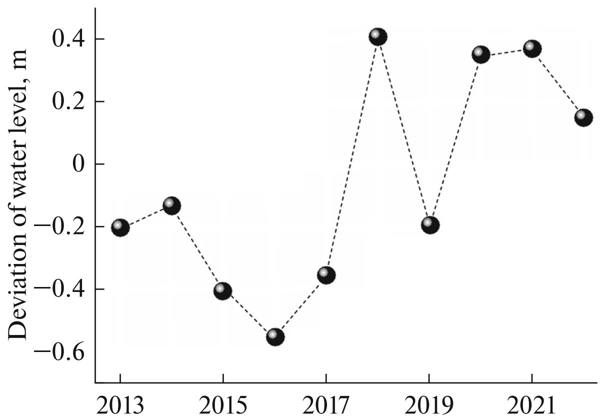

Fig. 1. Deviation of Baikal water level from average (456.2 m) over the expedition period from 2013 to 2022 (https://allrivers.info/gauge/baykal).

contents in the near-surface atmosphere and surface water throughout the Baikal water basin [23]. The spatial distribution of methane was studied during complex circum-Baikal expeditions onboard research vessels (RVs) of Limnological Institute, Siberian Branch, Russian Academy of Sciences, in spring season (May 29–June 7, 2013; May 25–June 6, 2016; May 26–June 5, 2017; May 25–June 6, 2018; June 1–June 13, 2021; and June 4–June 14, 2022). In each cruise, water samples were collected from the surface to the near-bottom region at the same 20 central stations [24, 25] and across a few transects [23]; and the concentrations of dissolved gases and biogenic elements were operationally determined in the ship's laboratory.

The period of measurements was selected based on the following considerations. It is just the spring season, when the lake is gradually warmed from south to north, and the ice cover is removed [23], is characterized by the strongest spatiotemporal variations in nearly all hydrological and biological processes [26]. At this time, the intrinsic interannual and seasonal rhythms of aquatic biota are seriously affected by the temperature regime of the region, which determines the rates of lake ice removal and river ice breakup, which, in turn, depend not only on spring weather, but also on the temperature and duration of the preceding cold period of the year. Also, many organic and inorganic compounds, accumulated during the winter season on a large drainage area and on the Baikal ice cover, enter the surface waters of the lake in the ice melting process [27]. This complex set of processes, manifesting most strongly in the coastal zone, also affects the entire lake basin.

It should be noted that the above-described series of expeditions was conducted in the period when large-amplitude interannual variations in the water level were observed on Lake Baikal. A long relatively stable period ended in 2013. After the advent of extremely low-water years of 2014–2017, the water level started decreasing and reached a minimal mark in the spring–summer seasons of 2016–2017. Then, in the warm season of 2018, the water level started growing [28] until approaching maximal values by 2021–2022 (https://allrivers.info/gauge/baykal) (Fig. 1).

Instrumentation

The instruments used and the main methodic aspects were described in [23]. Here, we recall briefly the components of the mobile complex, insuring recording of partial pressures of carbon dioxide and methane in the near-water atmosphere and surface water. It comprises the Picarro G2301-f CO_2/CH_4 gas analyzer; a selector for five sampling channels (three equilibrator channels + two atmospheric channels); an equilibrator equipped with temperature and water consumption sensors; a meteorological station; a navigator; and a system for continuous supply of surface water. The complex is included in the instrumentation in the Center for Collective Use "Atmosphere." The CH_4 concentration in water was calculated from measurements of methane partial pressure in the equilibrator chamber and methane solubility at a given temperature. The equilibrium methane partial pressure is calculated, taking into account the controllable consumption rate of pumped water and the rate of air supply to gas analyzer.

An important methodic aspect is that the 2013 measurements were carried out using a large-volume equilibrator, in which the time for reaching the equilibrium CH_4 pressure in the instrument chamber was 16 min [23]. The measurement period at hydrological stations is much longer (1–2 h), which ensures high-quality determination of partial pressure. However, when the ship is in motion, time required to reach the equilibrium partial CH_4 pressure affects appreciably the determination of the spatial resolutions of the concentration distributions. Analysis in work [23] showed that the coordinate referencing of the CH_4 concentration was uncertain by about 2 km when the ship speed was 15 km/h.

In subsequent cruises, we applied a small-volume multistage equilibrator. As compared to the preceding version, its construction has undergone dramatic changes, which made it possible to greatly reduce the time for reaching equilibrium partial pressures of the studied gases directly in the instrument chambers (less than 3 min, for methane). Also, the system of sensors, controlling the consumption and temperature of water flow, was modernized, enabling refinement of the data processing software. In this construction of the equilibrator, the methane partial pressure, measured using controllable parameters, is uncertain by no more than 10%, and the spatial resolution is reduced down to 200 m at the ship speed of 15 km/h. Evidently, if the RV passes fast across a local source of methane release, when the rate of change in gas content in water exceeds the equilibrator relaxation rate, the errors markedly increase.

RESULTS AND DISCUSSION

To estimate possible manifestation of interannual variations in the composition of surface water in the lake pelagial as a whole, in each springtime cycle we calculated the statistical characteristics of the concentrations of oxygen and biogenic elements over array of measurements at 20 central stations during spring circum-Baikal expeditions from 2010 to 2022 (Fig. 2).

No special interannual variations in the oxygen concentration are observed from 2010 to 2022 (Fig. 2a). Since 2013, a reliable tendency of decreasing concentrations of biogenic elements PO_4^{3-} and Si can be discerned in the pelagial surface water (Figs. 2b and 2c). A detailed study of the set of hydrochemical and biologic processes in this period is a multifaceted multidisciplinary problem, the solution of which requires enormous efforts of many specialists. But, already first publications [29] show that strong variations in the lake level lead to changes in the content of biogenic elements which are thought by the authors to be due to climate warming in the region of Lake Baikal. As applied to the purposes of this work, we can admit that the data, analyzed here, were affected, to one degree or another, by these processes and, in particular, by interannual rhythms of variations in river discharge, caused by the alternation between low- and high-water periods.

Figure 3 presents the results of recording the methane concentration along the route of the vessel passage in springtime expeditions in different years across the Baikal basin.

Evidently, the spatial CH_4 distribution in surface water is heavily determined by regional and local processes and, primarily, by the onset and duration of the gradual removal of the lake ice cover from south to north and, correspondingly, by the dynamics of water warming; and by wind regime, wave-driven mixing, etc., in different segments of the route. Omitting the detailed analysis of the spatial distribution of methane content in each expedition, because it requires a separate consideration and is beyond the scope of this work, we only note that: despite the strong interannual variations in water level, in the concentrations of certain biogenic elements, and in weather conditions in six expeditions, the main features of the spatial pattern of the methane content distribution in different years on large areas of the lake basin are similar in many respects. As was shown in previous works [16, 25], the largest CH_4 concentrations were recorded in the impact zones of river discharges (Fig. 3). With increasing distance from the coast, the methane content in surface water usually strongly decreased, and varied mainly in the range from 70 to 300 ng/L in the region of large depths.

Figure 4 presents the statistical characteristics of the CH_4 concentration in the surface water of Baikal at 20 central stations, calculated over the dataset from six

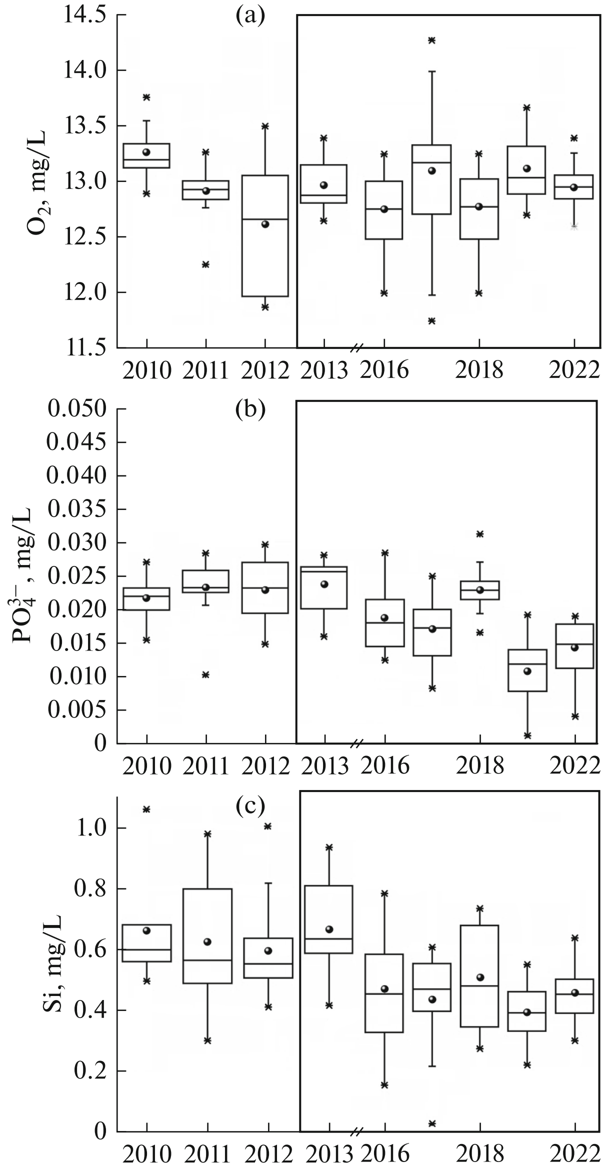

Fig. 2. Statistical characteristics of the (a) O_2, (b) PO_4^{3-}, and (c) Si concentrations in surface water in the pelagial of Lake Baikal in spring seasons of different years (averaging over 20 central stations): average values (circles), 25th and 75th percentiles (boxes); median (central line), 1 and 99% of values of the considered characteristic (crosses).

cruises. As can be seen, the methane concentration in the surface water varies in quite a narrow range of values from the southern tip of Baikal (station no. 1 is 15 km from the coast near Kultuk settlement) to station no. 19 (the center of the transect Tyya River – Cape Nemnyanko).

The exceptions are only the results obtained at station no. 6 (the center of the transect Krasnyi Yar – Kharauz), where the lake is about 260 m deep; and the water composition is usually strongly affected by the Selenga River discharges, leading not only to an increase in the CH_4 content, but also to its larger vari-

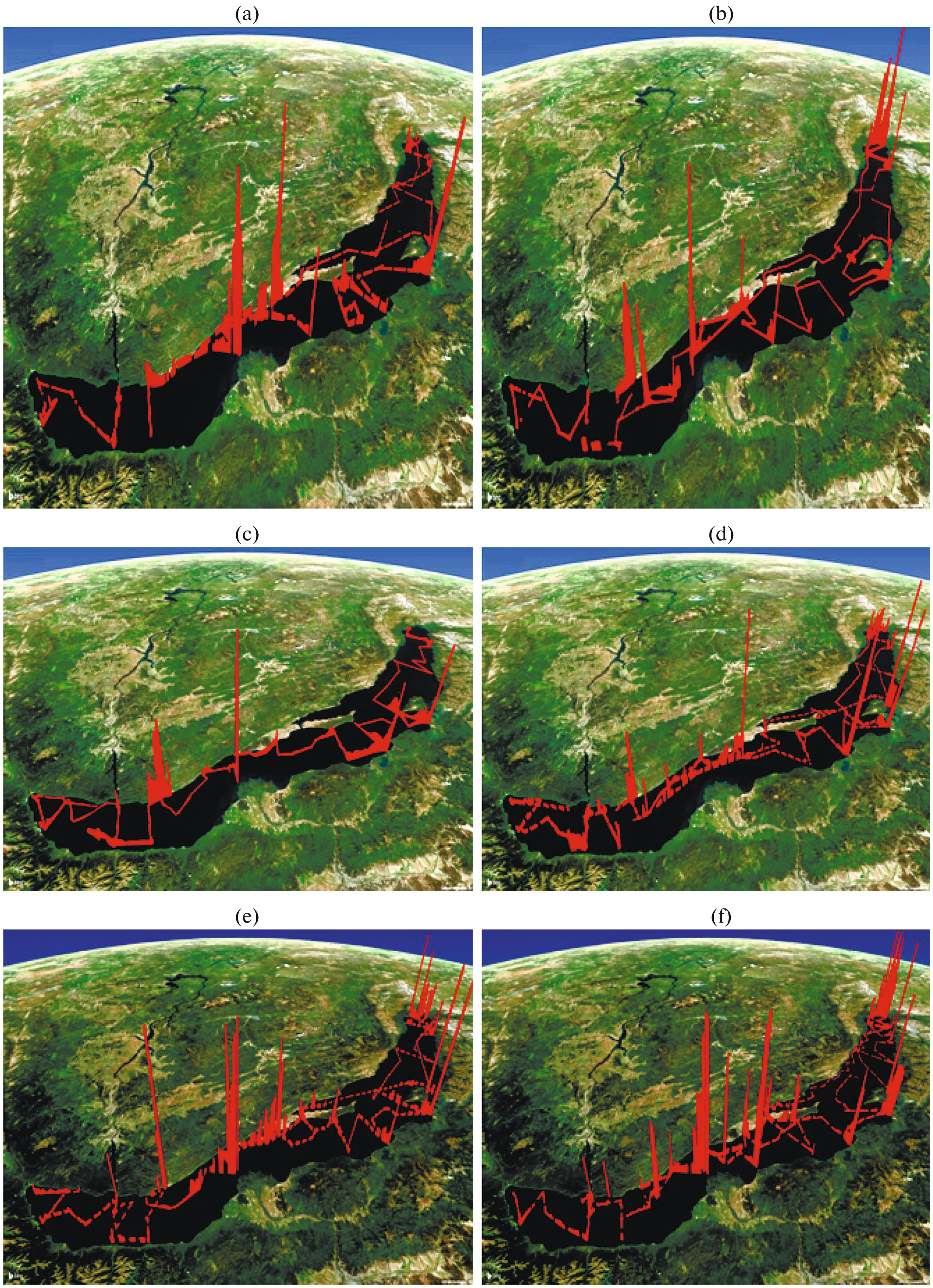

Fig. 3. Spatial distribution of the methane concentration in surface water in Lake Baikal during spring cruises: (a) 2013; (b) 2016; (c) 2017; (d) 2018; (e) 2021; and (f) 2022.

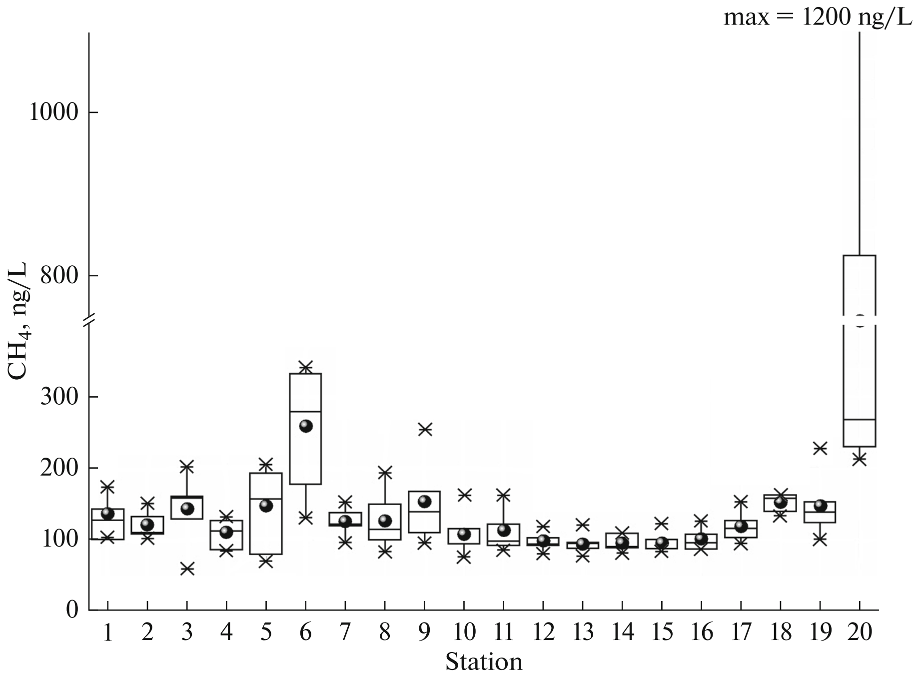

Fig. 4. Statistical characteristics of the CH_4 concentration in surface water in Lake Baikal at central stations. Notations are same as in Fig. 2.

ations. When the ship stopped at this station for 1–1.5 h, the methane concentration sometimes rapidly grew and the water surface temperature increased from 2–3 to 10°C, clearly indicating the arrival of water discharged by river. Station no. 20, where the CH_4 concentrations were maximal and frequently much exceeded 1000 ng/L, is located 7 km from Nizhneangarsk; and here, along the northern coast, the water composition is heavily determined by the constant strong impact of Rivers Kichera and Upper Angara.

SPATIAL DISTRIBUTION OF METHANE CONCENTRATION IN BAIKAL SURFACE WATER

The main purpose of this paper is to identify the most characteristic, stable features of the spatial distribution of methane concentrations in surface water of Lake Baikal in the spring period. But it is evident that the statistical representativeness of the dataset on different lake areas in each complex expedition depended on the route of the ship passage, which was determined by the scientific program and weather conditions (in total, there are 196 500 records in database from six cruises). Therefore, the CH_4 records in each geographic site strongly differed in number.

To reduce the contribution of variations in CH_4 concentrations, measured in a specific cruise, which were determined heavily by specific weather processes on different segments of the route, and to equilibrate artificially the statistical weights of results, obtained in continuous round-the-clock measurements in all cruises, we carried out the following procedure. The Baikal basin in the system of geographic coordinates was divided into segments (cells) with steps 0.005° in latitude and 0.01° in longitude. Their sizes were 556 × 7006 m (0.389 km^2) in the southern part of the lake and 5566 × 6216 m (0.345 km^2) in the northern part. Then, for each cruise the measured CH_4 concentrations were distributed over cells in accordance with their coordinates, and the average methane concentration was calculated in each cell. Next, average concentration over six cruises was calculated in each segment. Thus compiled resulting array contains the average values of water-dissolved CH_4 in 12 100 segments, the total area of which is 4466.7 km^2, or 14% of the total Baikal water surface.

This method of equilibrating the statistical weights of results for each segment of the Baikal basin envisages that only one value of the CH_4 concentration is available in a specific cell. Recall that at the central stations (see Fig. 4) the signal is recorded for 1–1.5 h; and only one average value is entered in the combined array. Only the average CH_4 content is also present in segments in which the methane concentration was measured repeatedly in different expeditions.

We think that the application of this artificial, "rough" method for equating the statistical weights for each segment in a combined array makes it possible to identify precisely the most stable features of the spatial pattern of distribution of the methane concentration in surface water over the Baikal basin in the spring period (Fig. 5).

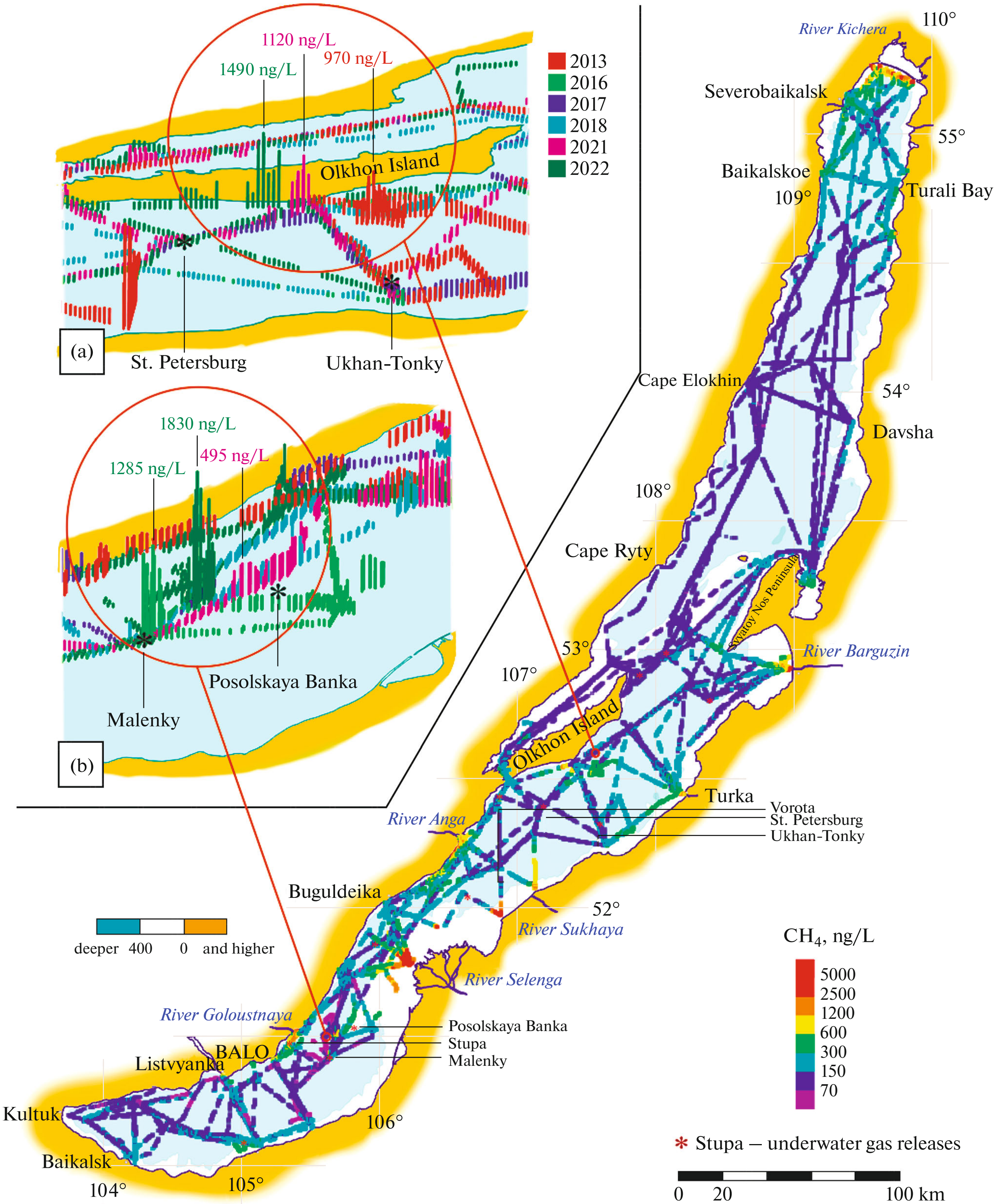

Fig. 5. Spatial distribution of the methane concentration in Baikal surface water in resulting data array from 2013, 2016, 2017, 2018, 2021, and 2022 expeditions (blue background in the map indicates the region with lake depth larger than 400 m); (a, b) fragments where anomalously high CH_4 concentrations were observed in the region of large depths in three out of six cruises in different years.

As we can see from Fig. 5, on the large part of the lake basin most of the CH_4 values are in the interval from 80–100 to 200–250 ng/L and observed within the 400-m isobath, and the concentrations are larger (300–500 ng/L) in the coastal zone. The concentrations of the order of 1000–11 000 ng/L were regularly recorded near outlets of large rivers: Selenga, Kichera, Upper Angara, Barguzin, Turka, Anga, and Sukhaya [23–25].

Against the background of this quite typical pattern, which in general traits was observed in each expedition, in certain cruises we recorded anomalously high CH_4 concentrations (Figs. 5a, 5b). We first detected a strong methane release, up to 970 ng/L, in 2013 at the distance of 9 km from the coast of Olkhon Island, where the depth is more than 1 km (Fig. 5a) [25]. The methane concentrations in this region varied in the range of 120–220 ng/L in the 2016–2018 expeditions; but in 2021 and 2022, the anomalously high concentrations up to 1120 and 1490 ng/L had again been manifested (Fig. 5a). We so far failed to propose any likely hypothesis on the possible methane source in this region; however, we hope our data will be useful for clarifying this issue and setting up special experiments.

Figure 5b presents a fragment of the distribution of methane concentration near the Malenky volcano (51.9200° N; 105.6400° E, depth 1305–1345 m) [30]. In this region, CH_4 content increased to 1285 ng/L in 2016 directly near the source, and from 495 to 1830 ng/L in 2021 and 2022 at the distance of 5–12 km from the source. Evidently, these methane concentrations on the surface were not due to supply in the form of gas bubbles (the depths are larger than 1 km in this zone), but rather to a rapid ascent of a large mass of water, enriched by methane due to the "eruption" of the volcano, the sporadic emissions of which were noted in work [30].

For a more detailed analysis, we compiled four datasets, obtained in the areas between isobaths: 0 and 100 m (array 1), 100 and 200 m (array 2), 200 and 400 m (array 3), and deeper than 400 m (array 4). This division is based on publications of other researchers [21, 22, 30]. From the coastline to the 100-m isobath the methane concentration in surface water is heavily determined by its supply from bottom sediments and discharges of river water on the separate lake sections [22]. In the region of the depths of 100–200 m, there is a weaker effect of small rivers, but methane is still supplied from near-bottom region and, in particular, by bubble gas releases [30]. Between the 200- and 400-m isobaths, CH_4 releases from the lake bottom still make a certain contribution to the methane content; and the maximal methane concentrations along the route of the ship passage are due to discharge of the River Selenga in the southern hollow and to discharges of Rivers Upper Angara and Kichera in the northern hollow [21]. As shown in work [30], in the Baikal basin up to the depths shallower than 380 m we recorded more than 120 shallow-water near-bottom sources of gas, 90% of which are in the delta of River Selenga. As the distance to the 400-m isobath decreases, their effect vanishes because bubbles are almost totally dissolved at the depth of 250–280 m [30]. The region of the lake within the 400-m isobath (74% of the total Baikal area) can conventionally be considered as background territory.

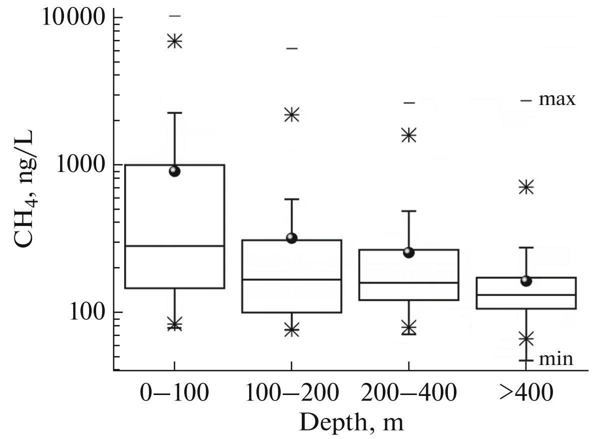

Fig. 6. Statistical characteristics of methane concentration in surface water of Lake Baikal between the corresponding isobaths. Notations are the same as in Fig. 2.

Figure 6 presents the statistical characteristics of methane concentration, calculated from measurements in these four arrays. It can be seen that maximal, average, and median values of the methane concentration decrease when moving from coastline to the region of large depths. This tendency is distorted just by 1% of the maximal concentrations in array 4, which are caused by separate anomalous methane releases (Figs. 5a, 5b). We note that the minimal concentrations are almost identical in the first three arrays, but they are reliably smaller in array 4. This is because precisely in the region of large depths low CH_4 concentrations in the range of 50–70 ng/L were observed in all cruises on separate segments of the route. And, as shown in [31], this was determined by the supply of methane-poor waters to the surface layer in the process of vertical exchange with weak winds and by the development of springtime temperature convection (homothermy) [31, 32]. In Fig. 6, the average and median values markedly differ in each array; therefore, no further analysis of statistical characteristics under the assumption of normal distribution is expedient.

At the next stage of the data processing, the statistical characteristics were calculated for the logarithms of the CH_4 concentrations. As expected, the distribution of the dataset in array 1 could not be described by any function. On the segments near rivers, the methane content in surface water depends not only on the average volume of the river discharge, but also on many local processes near the coast and, in particular,

Table 1. Average methane concentrations and confidence interval at the 99% confidence level

Data array	Average concentration, ng/L	Confidence interval at 99% confidence level, ng/L	Array in percent of total lake area, %	Number of cells in array (their areas, km^2; in percent of area in a corresponding array, %)
0–100 m* (75% of data in total array)	215*	200–230*	9	560 (207; 9)
100–200 m	200	182–219	5	527 (194; 12)
200–400 m	190	181–198	9	1337 (491; 16)
>400 m	141	140–143	74	9490 (3505; 15)

* Calculations disregarded concentrations larger than 1000 ng/L.

on the wind direction and speed. In array 1, 25% of the data were obtained when the ship cruised in the river impact zone, where the concentrations were extreme and varied from 1000 to 10 160 ng/L (the coefficient of variation in the total dataset was about 65%). In arrays 2, 3, and 4, the distribution of results of the calculations corresponded well to a lognormal law. The table presents the estimates of the average methane concentrations and of confidence interval at the 99% confidence level.

As can be seen from our measurements, the average methane concentration in surface water varies in quite a narrow range from 140 to 220 ng/L in the region of depths larger than 100 m in the Baikal basin (about 90% of the total lake area). This is quite consistent with results of other researchers (e.g., [15–17, 20–22, 30]).

CONCLUSIONS

In this work we analyze the spatial distribution of the methane concentration over the entire Baikal basin in the spring period, using multiyear continuous measurements in six expeditions of 2013, 2016, 2017, 2018, 2021, and 2022. The statistical characteristics were determined using combined dataset in 12 100 segments (steps of 0.005° in latitude and 0.01° in longitude), the total area of which had been 4466.7 km^2, or 14% of the mirror of Lake Baikal. Owing to the artificial method of equating the statistical weights for each segment in the combined array, in each cell there is only one CH_4 concentration. In our opinion, this approach makes it possible to identify more clearly the most stable features of the spatial distribution of methane concentration. To describe the distribution of the methane concentration in surface water throughout the water basin in more detail, we calculated the statistical average values and the confidence interval at the 99% confidence level in four zones: between isobaths 0 and 100 m; 100 and 200 m; 200 and 400 m, and deeper than 400 m. The good agreement between methane concentrations in the array analyzed with the data of other researchers (e.g., [15–17, 20–22, 30]), obtained in different years on nearby areas of the water basin, makes it possible to conclude that the results, presented in this work, reflect adequately the most stable features of the spatial methane distribution in surface water of Lake Baikal in spring seasons.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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