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### Estimation of "water-atmosphere" carbon balance in the coastal zone of Lake Baikal based on the CO<sub>2</sub>/CH<sub>4</sub> fluxes

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#### ABSTRACT

Long-term observation series of carbon dioxide and methane fluxes in the water-atmosphere system in the coastal zone of Lake Baikal are analyzed. It is shown that the total sink of carbon dioxide from the atmosphere to the water surface is 5.9 gCO2 m<sup>-2</sup> year<sup>-1</sup>, while the methane emission from water is estimated as 80 mgCH4 m<sup>-2</sup> year<sup>-1</sup>. For comparative estimation of the radiative forcing, it is taken that the contribution of the atmospheric methane molecule to global warming is 27 times greater than that of the CO<sub>2</sub> molecule. Then, the coastal area of Lake Baikal contributes to global warming due to the methane emission 8-20% more than to the cooling due to absorption of carbon dioxide by the water.

Keywords: carbon dioxide, methane, air-sea gas exchange, diurnal and seasonal pattern, global warming potential, littoral, Lake Baikal

#### 1. INTRODUCTION

The carbon cycle of aquatic ecosystems is associated primarily with the vital activity of organisms inhabiting the water column. The unique Baikal aquatic biota, which provides the safety of clean water, significantly depends on chemical composition of water coming with river runoff [1] and the stability of gas exchange in the "atmosphere-water surface" system. Biochemical processes occurring in a reservoir obviously affect the composition of the near-water atmosphere through the gas exchange between the water surface and the atmosphere [2, 3].

Carbonaceous gases, such as carbon dioxide and methane, are directly involved in this cycle. As atmospheric constituents, these gases have a significant impact on the climate, contributing approximately 66% (CO<sub>2</sub>) and 17% (CH<sub>4</sub>) of the radiative forcing by long-lived greenhouse gases (LLGHGs) [4].

Taking into account the current climate challenges, a detailed study of the carbon cycle is still one of the key problems needed for assessing the environmental state of the Baikal ecosystem and its contribution to the global climate picture.

In our paper, to estimate the total  $CO_2$  and  $CH_4$  fluxes at the water-atmosphere interface in Lake Baikal, we use the data of *in situ* measurements of the fluxes of these gases obtained by the floating chamber technique and the partial pressures of these gases in the surface water and the near-water atmosphere [3]

It is obvious that to estimate the total budget of exchange of carbonaceous gases with the atmosphere, all-season measurements of fluxes in various aquatic ecosystems of the lake (pelagial, littoral, confluence areas of large rivers, sor zones, etc.) are required. The reported results of long-term all-season measurements of  $CO_2$  and  $CH_4$  fluxes were obtained in the Baikal Atmospheric-Limnological Observatory (BALO SB RAS) located in the coastal zone of the western coast of the south Baikal. This zone is characterized by some features, among which the following should be highlighted: the coastal zone stands out for its maximal biodiversity [5]; the proximity of the bottom has a significant impact on the content of dissolved carbonaceous gases in the surface water layer due to the bioproductivity of benthic communities. In particular, the bottom is the main source of methane [6], which freely reaches the upper horizons. For almost the entire western coast, as well as the BALO area, the bottom topography is characterized by a sharp increase in depth, due to which water masses

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from deep horizons are transported to the surface by seiches or storm events, even under conditions of stable stratification. However, due to the particular composition of deep waters, such situations can be quite easily differentiated.

Based on the aforesaid, it can be noted that the reported data are true for coastal water areas of the west and east Baikal except for the areas subject to anthropogenic influence [1, 7], opposite the outflow of large rivers and bays.

#### 2. MEASUREMENT SITE AND METHODS

The results reported in the paper were obtained in the Baikal Atmospheric-Limnological Observatory (BALO SB RAS, a part of the "Atmosfera" Resource Sharing Center) of the Limnological Institute (LIN) SB RAS and V.E. Zuev Institute of Atmospheric Optics (IAO) SB RAS. The observatory is located at the scientific field station of the Limnological Institute on the western coast of the South Baikal (51.90°N, 105.06° E) (Fig. 1). We started the measurements of carbon dioxide fluxes at Lake Baikal in September 2004. Before 2012, the measuring system employed LiCOR-840 NDIR  $CO_2/H_2O$  gas analyzer (LICOR Inc, USA). In July 2012, it was complemented with the  $CO_2/CH_4$  gas analyzer (Picarro G2103, USA). Since that time, the observation series include data on methane fluxes as well.

Gas fluxes in the water-atmosphere system are measured with a semi-submersible chamber floating freely on the water surface at a distance of about 40 m from the coast (Fig. 1). To ventilate the chamber in an hourly measuring cycle, it is detached away from the water and held for 5 min for free ventilation with the atmosphere. A particular advantage of this approach is that the complete detachment of the chamber allows the top layer of water, which is held under the chamber during the exposure, to be replaced. The exposure time is 20 or 50 min depending on the flux intensity. The flux is calculated from the increment/decrement in the content of the test gas during the exposure [3].



Fig. 1. Arrangement of the measuring equipment of the BALO coastal part with the coordinates of the measurement site (top); chamber installation on the water surface (bottom).

At the same time, the content of dissolved carbon dioxide and methane in the surface water layer is recorded with a multistage equilibrator [8]. The comparison of data on the flux and the difference in partial pressures of gases in the wateratmosphere system is used to estimate fluxes in the Baikal pelagic zone, where continuous flux measurement by the chamber method is difficult.

To date, the number of simultaneous measurements of  $CO_2$  and  $CH_4$  fluxes in BALO is 24 series from 10 to 14 days long. The measurements span all seasons except for the periods when the lake is covered by ice. The reported results are based on 7330 readings of the  $CO_2$  flux and 1600 readings of the  $CH_4$  flux. The composition and operating mode of the BALO measuring system are described in detail in [8].

#### 3. **RESULTS AND DISCUSSION**

#### 3.1 Diurnal and seasonal patterns of CO<sub>2</sub> fluxes in the water-atmosphere system

It is well-known that the carbon dioxide flux through the water-atmosphere interface is regulated primarily by biochemical processes, such as photosynthesis and respiration of aquatic biota. Correspondingly, the value and direction of the  $CO_2$  flux have the pronounced seasonal and diurnal profile. Based on our experience of previous studies [9, 10] we can note that the variability of the  $CO_2$  content in the near-water air layer manifests itself most clearly in the diurnal cycle. The amplitude of diurnal variations of the  $CO_2$  content in the atmosphere of the studied region does not exceed 5% (standard deviation from the average value) even at the peak of photosynthetic activity (July-August), whereas the variability range of the  $CO_2$  concentrations in the surface water is about 50% [11].



Fig. 2. Diurnal profile of carbon dioxide flux for every month (a total of 7330 measurements since 2004 till 2021) and methane flux (a total of 1590 measurements since 2012 till 2021). Each reading on the plot corresponds to the average value and standard deviation over the data array for every hour of a day of a particular month. Negative values correspond to the sink of the gas to the water surface.

Figure 2 (top) shows the long-term monthly average diurnal profile of carbon dioxide fluxes in the littoral zone of Lake Baikal from May to December. As can be seen, the diurnal variation is clearly pronounced for every month of the open-water period. Even in December (pre-freeze period), when the flux is directed, on average during 24 hours, to the atmosphere, its value decreases in the afternoon. In other months, the emission of carbon dioxide from the water surface into the atmosphere is observed in nighttime, and the sink is maximal at 16:00 Local Time.

Table 1 presents the diurnal average fluxes for every month in the open-water period. Based on these data, we can conclude that 1 sq. m of this water area absorbs 5.9 gCO<sub>2</sub> per year (-135 mmolCO2 m<sup>-2</sup> year<sup>-1</sup>).

#### 3.2 Diurnal and seasonal patterns of CH<sub>4</sub> fluxes in the water-atmosphere system

First of all, it should be noted that for the entire period of observations in the coastal zone, we have not observed a single case of methane sink to the water surface, it is always directed from the water to the atmosphere. The partial pressure of the dissolved methane always exceeds the partial pressure in the near-water atmosphere. Methane fluxes demonstrate no pronounced diurnal and seasonal cyclicity at the measurement site (Fig. 2). Sporadic bursts of the methane partial pressure in water many times exceeding the background level are observed sometimes. In this analysis, the background level is taken to be equal to minimal values ( $\sim$ 5 µatm). Looking ahead, we can note that at a distance of 3-5 km from the coastline, situations that the methane partial pressure in the surface water exceeds 5 µatm are very rare.

The average value of the methane flux from water to the atmosphere in the coastal zone is 400  $\mu$ gCH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup> or 80 mgCH<sub>4</sub> m<sup>-2</sup> for the entire open-water period (5.4 mmolCH4 m<sup>-2</sup> year<sup>-1</sup>).

	May	June	July	August	September	October	November	December
CO <sub>2</sub> flux	-22.5±34	-26.4±127	-47.3±135	-146.5±122	-82.4±114	-13.4 ±244	14.5±47	$125.9 \pm 64$
CH <sub>4</sub> flux	_	0.27	0.54	0.30	0.59	0.6	0.11	0.45

Table 1. Diurnal average gas fluxes for every month, mg m<sup>-2</sup> day<sup>-1</sup>

#### **3.3 Global Warming Effect**

The exchange of carbonaceous gases of the underlying surface with the atmosphere is one of the important regulators of the planetary radiative balance. In terms of carbon, each square meter of the lake surface absorbs 1.6  $g_C$  in the form of carbon dioxide and releases 0.08  $g_C$  in the form of methane during the period of open water. Thus, we can conclude that the water of the Baikal coastal zone receives 20 times more carbon due to the sink of carbon dioxide than releases into the atmosphere with methane fluxes.

For comparison of the global warming impacts of different gases, the Global Warming Potential (GWP) was developed. GWP of carbon dioxide is taken equal to 1 [12]. For an approximate assessment of the radiative balance provided by the coastal waters of Lake Baikal, we take that the annual sink of  $CO_2$  is 135 mmol  $CO_2$  m<sup>-2</sup> year<sup>-1</sup> and the methane emission is 5.4 mmolCH<sub>4</sub> m<sup>-2</sup> year<sup>-1</sup>. The  $CO_2/CH_4$  molecular balance ratio is about 25.

The recent studies show that the Global Warming Potential (GWP) of methane is estimated as 27-30. That is, the contribution of the methane molecule to the global warming is more than 27 times greater as compared to the CO<sub>2</sub> molecule. Hence it follows that the contribution of Lake Baikal coastal waters to the global warming due to the methane emission into the atmosphere is 8-20% greater than their contribution to the cooling due to the absorption of carbon dioxide. It should be noted, however, that in the pelagic zone of Lake Baikal, which occupies the largest area, the methane emission is noticeably (about two orders of magnitude) lower than in the coastal zone. As to the sink of CO<sub>2</sub> to the water in the open water area of the lake, it remains approximately at the same level as in the littoral. The carbon balance of the open water area of Lake Baikal is considered in our next publications.

#### 4. CONCLUSIONS

From the data of long-term measurements, we have determined that the carbon sink from the atmosphere due to the carbon dioxide flux is an order of magnitude higher than its emission into the atmosphere due to the methane emission. Taking into account the obtained approximate estimates, we can conclude that the gas exchange of carbon dioxide and methane in

the "water surface-atmosphere" system in the Baikal coastal zone contributes the global warming due to the methane emission into the atmosphere.

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