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Methane fluxes in an artificial valley reservoir according to field observations and mathematical modeling

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Abstract. A very important problem nowadays is to assess anthropogenic impact on global warming. Increasing greenhouse gas concentrations in the atmosphere is the main reason for the rising global planet temperatures. The most important greenhouse gases (GHGs) are carbon dioxide and methane. The forcing made by carbon dioxide has been studied for a long time, while less attention has been focused on methane in this regard. However, more and more evidence indicates significant methane emissions into the atmosphere from anthropogenic sources. Less studied, but also important sources of methane are artificial reservoirs. In order to more accurately estimate methane emissions from the reservoirs, it is necessary to study the processes occurring with methane in the aquatic ecosystems in detail. For this purpose, in this study a Mozhaysk reservoir located in Moscow region is chosen. Studies of the spatial and temporal variations of methane fluxes have been carried out on this site for 6 years since 2015. In addition, this reservoir is used for validation of a well-known model, LAKE. This model can be used as an instrument for detailed estimation of methane emissions into the atmosphere from the surface of reservoirs.

1. Introduction

Methane is one of the most important greenhouse gases in the Earth's atmosphere. Despite the fact that its concentration in the atmosphere is relatively low (about 1.774 ppb), the global warming potential of methane per molecule is 72 times higher than the potential of carbon dioxide (for a 20-year period) [1]. There are both natural and anthropogenic sources of methane. Large natural sources of methane are wetlands, tropical forest ecosystems, the continental shelf of the World Ocean, etc. Anthropogenic sources are wastelands or garbage areas, rice fields, pastures of cattle, and artificial reservoirs [2]. According to various estimates [2 – 5], reservoirs are responsible for 0.5 to 10% of the total methane emission into the atmosphere.

Methane originates in reservoirs as one of the main anaerobic organic material decomposition products in bottom sediments [6]. It rises to the water surface in the form of a diffusion and bubble

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flux (Figure 1). The diffusive component of the flux is often relatively small and has a much lower intensity than the bubble flux. The value of the diffusive flux primarily depends on the gradient of methane concentration. In addition, dissolved methane molecules are actively oxidized by methanotrophic microorganisms, especially in the presence of oxygen. As a result, about 80% of methane formed in bottom sediments does not reach the atmosphere [7].

The bubble flux cannot be oxidized by methanotrophs. However, methane bubbles can dissolve in water due to rising into water layers less saturated with methane. For this reason, the magnitude of the bubble flux significantly depends on the depth of the reservoir. The bubble component dominates over the diffusive one in the flux structure for the most part of the year due to a high rate of the bubble rise velocity and its “resistance” to oxidation. Also, a significant flux of methane into the atmosphere can be observed at the shallow parts of reservoirs covered with macrophytes plants [2].

In addition to vertical fluxes of methane in the reservoir, it is also necessary to note horizontal ones, such as inflow of methane with river waters flowing into the reservoir, and downstream degasation of methane through dam turbines and with outflow discharge [8] (Figure 1). However, the horizontal fluxes of methane are less significant than the vertical ones.

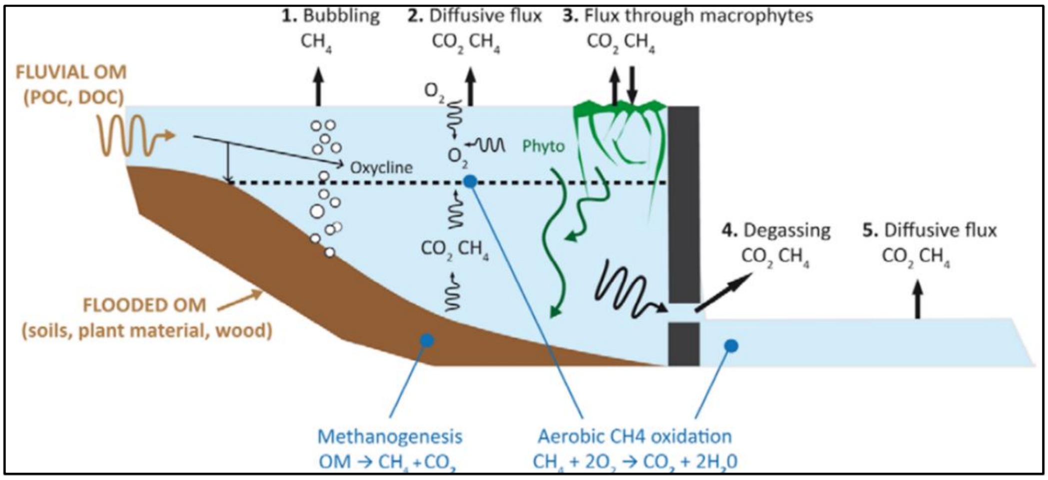

Figure 1. Scheme of distribution of methane fluxes in a reservoir adopted from [2].

There are many studies assessing the total methane emission from artificial reservoirs on the global scale. However, these estimates differ substantially (2 – 122 Tg CH_4*$year^{-1}$ [2 – 5]) because of differences in the calculation methods. In some works, the available field data on various water bodies are used, and the assessment of total methane emission from reservoirs is based on the classification of the world's reservoirs according to climatic zones [3]. In other articles, the assessment is based on basic parameters of the reservoir: average depth, flow rate, eutrophication status, etc. A large group of authors [4] collected most of the available estimates of methane emission in their study and provided a global emission rate of 17,6 Tg CH_4*$year^{-1}$.

A new method for assessing methane emissions from the world's reservoirs is mathematical modeling of the methane processes developing in water bodies. The main input information for these models is meteorological data. Thus, by grouping water bodies by similar characteristics used in a model it is possible to estimate regional and global methane emissions based on the model simulations. This method is more time consuming compared to simple empirical algorithms, however, it is likely to provide more accurate results. An example of such a model is the LAKE model [9 – 11], which has already been successfully applied to various natural water bodies.

2. Object and research methods

The Mozhaysk reservoir was chosen to study spatiotemporal variations of methane fluxes at “sediments – water” and “water – atmosphere” interfaces in an artificial water body. This is a slowflow small reservoir of a valley type located in the upper basin of the Moskva River in Moscow region, Russia. The dam was constructed in 1962. Its main characteristics are shown in Table 1.

Table 1. Morphological characteristics of the Mozhaysk reservoir (all characteristics are given for the NWL) [12].

Length	Max width	Mean width	Max depth	Area	Volume	Annual amplitude of water level variations	Residence time
28 km	2.6 km	1.1 km	22.6 m	30.7 km^2	0.24 km^3	6 m per year	0.56

The measurements are carried out at 5 reference stations (I – V in Figure 2) located along the reservoir above the morphological sections of the flooded bed of the Moskva River. This arrangement of the stations allows one to assess the spatial variability of the processes and the transformation of the water masses along the reservoir length.

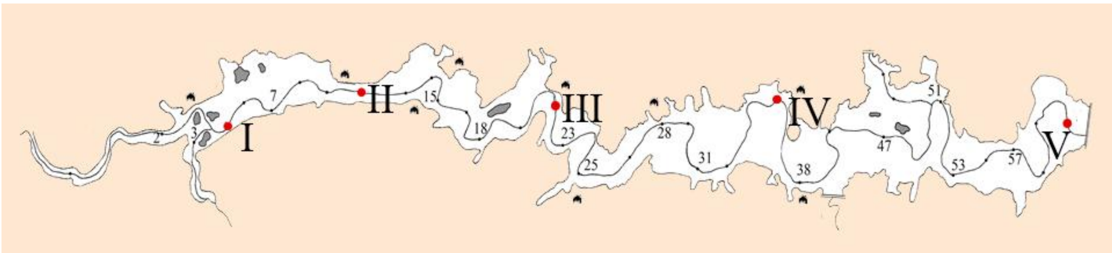

Figure 2. Scheme of Mozhaysk reservoir with stations of observations (I – V).

The study of this reservoir has been carried out for more than 50 years, including many studies of the functioning of various ecosystems. Observations of methane fluxes at the "bottom sediments – water" and "water – atmosphere" interfaces have been carried out for the last 6 years. Most measurements are carried out in the summer period, when the greatest spatiotemporal variability of methane fluxes is observed.

The program of measurements includes taking samples to determine the methane concentration in water by using the “headspace” method [13]. Fluxes of methane into the atmosphere are determined using the “floating chamber” method (total flux), while the diffusive component of the flux is computed using the TBL (thin boundary layer) method [13]. The methane flux from bottom sediments is determined using the Kuznetsov-Romanenko method [14], as well as using bottom chambers. Each series of in situ observations includes measurements of temperature, concentration of dissolved oxygen, and conductivity over the water column. All samples were analyzed on a gas chromatograph to obtain values of methane concentration in water and air and calculate the methane fluxes.

Another direction of the research of methane fluxes in the Mozhaysk reservoir is application of a mathematical model to simulate the processes taking place with methane in the reservoir. A large amount of field data accumulated over a long-term period of methane observations in the Mozhaysk reservoir, and also the well-studied status of this water body, facilitates the use of this reservoir as a reference object for testing the mathematical model. Thus, it is possible to reliably verify the model on the Mozhaysk reservoir, investigate the mechanisms of formation of methane fluxes, create a methodology for estimation of the methane emission basin on modeling results, and subsequently use it for other less studied water bodies.

The relevance of using modeling methods is also due to a low temporal coverage of field data, which is often insufficient for accurate estimates of methane emissions from the reservoir. In addition, sometimes field work is nearly impossible, for example, during the ice melting period, when a large volume of methane accumulated under ice in winter escapes to the atmosphere. These problems can be solved with mathematical models, where a small time step for the calculations can be chosen.

For these purposes, model LAKE was chosen in this study [9 – 11]. This model includes a part for calculating biogeochemical processes, such as the formation of methane in the reservoir sediments, its fluxes to the surface and emission into the atmosphere.

2.1. Description of model LAKE

LAKE is a one-dimensional hydrodynamic model for the calculation of hydrological, thermodynamic, and biogeochemical processes in lakes and reservoirs. It belongs to a class of boundary-layer-type models, and is supplemented by a horizontal pressure gradient parameterization [11], the calculation process can be represented as a numerical grid of the model (Figure 3).

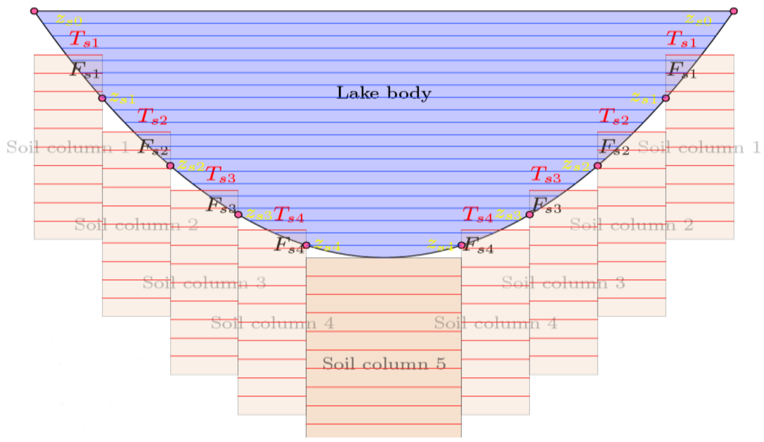

Figure 3. Water body representation in model LAKE [9].

The chain of simulated processes can be described as follows: the formation of methane in soils leads to the gas flux into the lower water layer of the model, corresponding to the respective soil column. The different depth of sediment columns allows one to partially represent the effects of the sloping bottom of the reservoir. Further, the methane oxidation, bubble dissolution etc. are calculated in the water column, and turbulent transport of methane towards the water-air interface. The model reproduces the processes in small water bodies satisfactorily, and demonstrates good correspondence to observational data. This model has been successfully applied to various lakes - Lake Kossenblatter (Germany), Lake Valkea-Kotinen and Kuivajärvi (Finland), Lake Shuchye (Siberia), and Bolshoi Vilyui Lake (the Kamchatka Peninsula).

The model calculates methane within three blocks: sediment methane formation (Sediment Methane Module), diffusion fluxes of dissolved gases (Gas Transport Module), and bubble flux (Bubble Transport Module).

The concentration of methane in bottom sediments is determined by the intensity of four processes: production, oxidation, bubble evasion, and molecular diffusion. Diffusion exchange between layers in the model depends primarily on the difference in methane concentration in these layers. The bubble flux formation in sediments and its change with depth during rise to the water surface is computed for each sediment column separately. The diffusion component of emission into the atmosphere is calculated using the TBL method.

According to the complex specifics of biochemical processes, the model contains more than 20 different empirically determined or calibrated parameters. The first step in the model calibration is defining constants related to the hydrodynamic processes, and the next one involves biochemical constants.

The main input data in the model are meteorological characteristics: air temperature, humidity, atmospheric pressure, wind speed, downward fluxes of short-wave and long-wave solar radiation, and the rate of precipitation. Other input information is the reservoir water level and discharge of main inflow rivers (Moskva and Lusyanka). The water balance of a reservoir in the model is important, because it determines the reservoir depth and, hence, the bubble component of the CH_4 flux.

3. Description of input data

A sufficiently high temporal resolution of the input meteorological data is required to improve the modeling results. The ERA-5 Land reanalysis was chosen as the main source of meteorological information. This source presents hourly data with 0.1° grid resolution in longitude and latitude. However, in order to make sure that these data are of sufficient quality, it is necessary to compare reanalysis data with ground-based observations. The source of ground data was a floating meteorological station located at the station IV "GELLA" (Figure 2). These irregular meteorological observations were carried out in the summer period. The floating station monitors the main meteorological characteristics, excluding solar, atmospheric radiation, and precipitation rate.

For reanalysis validation, the summer-autumn periods of 2017 and 2018 were selected. The temperature, pressure, air humidity, and wind speed averaged over three-hour periods were compared (Figure 4 a – d) (comparison of the wind direction was impossible, since "Gella" is a drifting station and there is a large error in determining the wind direction there). As criteria for the quality of regression models, the Pearson correlation coefficient (R), the Theil coefficient of series divergence (U) [15], and the F-criterion for the ratio of variances according to the regression equation and the residual [16] were used. According to the results of the regression analysis, it was found that temperature, pressure, and air humidity given by reanalysis and measured at the floating meteostation provide good statistically significant linear regression equations that can be used to correct the reanalysis data. The relationship between the wind speed is not linear and, therefore, for this regression not Pearson's correlation coefficient but a multiple correlation coefficient was used. In addition, the relationship for the wind speed is the weakest of all the others, however, it has a high correlation coefficient and a low Theil coefficient (less than 0.2), which means a fairly close relationship [15].

The reanalysis data on the downward shortwave and longwave radiation were compared with measurements carried out at the Zvenigorod station of the A.M. Obukhov Institute of Atmospheric Physics RAS for 2016-2017 (Figure 4 e, f). The latitude correction was not used, since the difference between the selected point of the reanalysis grid and the Zvenigorod station is less than 0.2 degrees.

Similarly to the other characteristics, a regression model was created for radiation fluxes measured at the Zvenigorod observatory, and the reanalysis data. Both regression models (for longwave and shortwave radiation) satisfy statistical criteria for the significance of the regression. For the regression model for shortwave radiation, the coefficient b in the equation was set to 0 to avoid non-zero values at night. It should be noted that the chosen F-test for the significance of the regression loses its relevance with a large number of observations. However, tests for the significance of the regression on smaller selection also showed an adequate p-value (<0.0001).

To assess the possibility of using precipitation reanalysis data, the values obtained with ERA-5 were compared with the data of the nearest meteostation where precipitation is monitored (Figure 4 g), which is Naro-Fominsk. Precipitation in the LAKE model is necessary to calculate the water balance and the thickness of the ice and snow cover in winter. The precipitation values are an extremely irregularly distributed characteristic in space and time, so the monthly precipitation sums were compared according to the reanalysis and Naro-Fominsk data. The regression model satisfies the statistical criteria and shows adequacy of the precipitation reanalysis data.

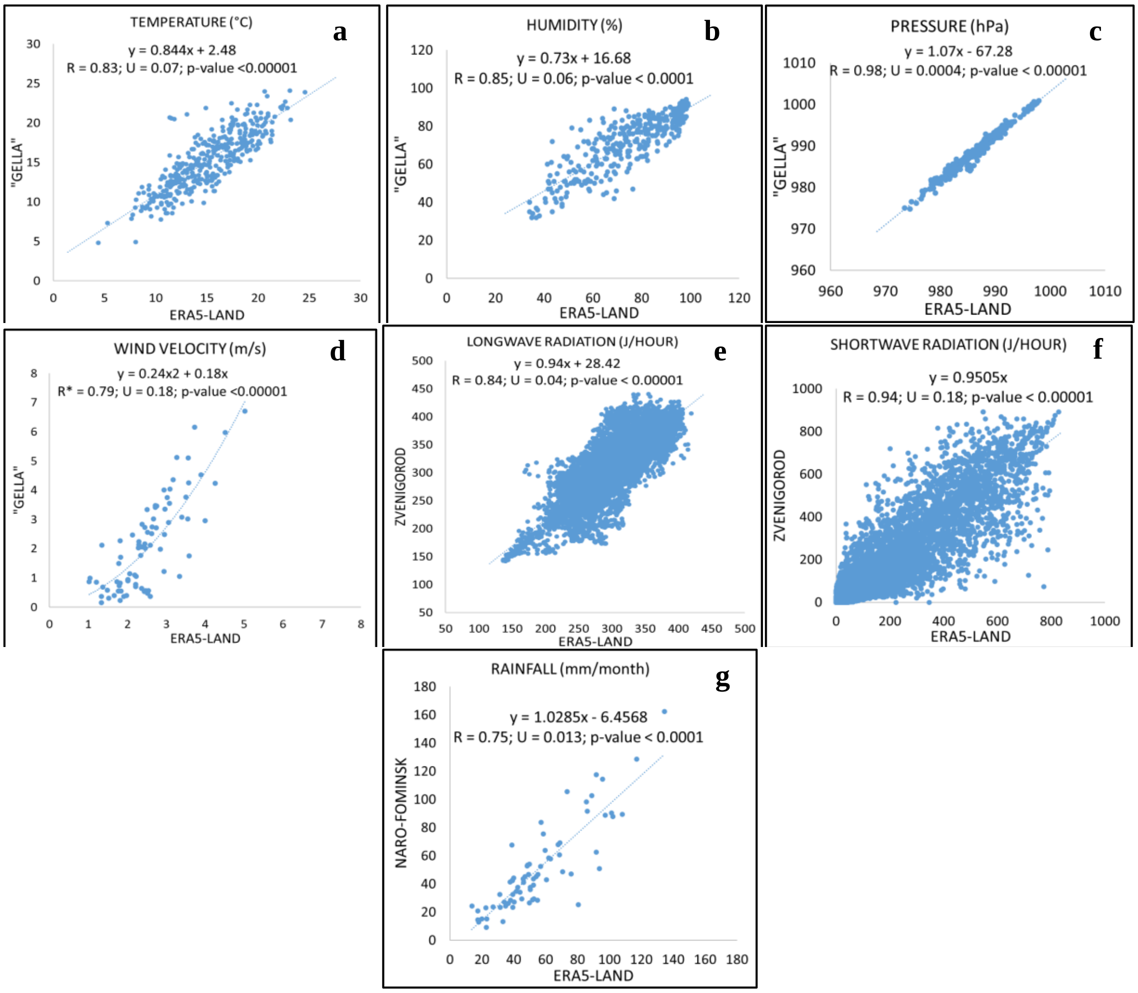

Figure 4. Regression models of meteorological characteristics (a – temperature, b – humidity, c – pressure, d – wind velocity, e – longwave radiation, f – shortwave radiation, g – precipitation) between ERA5_Land data and ground observations.

4. Results

The model was simulated for a 5-year period, from 2015 to 2019. According to the model results, there is a regularity in the annual variability of the methane flux into the atmosphere. During the winter period, a small amount of methane is emitted into the atmosphere due to the ice cover (the total flux does not exceed 5 mg $CH_4*m^{-2}*d^{-1}$). However, methane can accumulate in the ice cover of the reservoir, trapped there in bubble form, and this methane cannot be oxidized. Thus, during the ice melting period in spring, the flux of methane into the atmosphere can be very significant. This fact is confirmed by the modeling data (Figure 5) (there are no field observations during this period due to unsafe ice conditions). Especially significant emissions at the beginning of the spring season took place in 2018 and 2019, according to the model.

Homothermy is observed during spring due to convective mixing of warmed water masses; therefore, the water column is well mixed and saturated with oxygen. The methane flux at this time is not large due to a high content of O2, and also due to low water temperature values and inhibition of the organic processes, as well as the activity of methanogenic microorganisms. As the reservoir warms up, the bubble component of the flux begins to rise. Water temperature increase leads to the growth of plankton organisms; therefore, the amount of detritus entering the bottom horizons gradually increases and the rate of organic decomposition as well as oxygen consumption in the bottom layers rise rapidly. The ebullition flux has a huge heterogeneity in time due to a periodic change in the weather conditions. In warm and calm weather, there is a gradual increase in both the bubble and diffusion components of the methane flux due to the accumulation of methane in hypolimnion, and during the passage of atmospheric fronts and storm mixing of water body, the methane accumulated at the bottom quickly reaches the water surface and evades to the atmosphere, and after that the flux significantly decreases. Most significant values in methane emissions are associated with the passage of atmospheric fronts (Figure 5).

In the summer period, density stratification occurs due to an increase in the air temperature and more intensive heating of the subsurface water horizons. The summer stratification intensifies during calm and warm summer weather, and can be observed during the whole summer period. Stable temperature and density gradient leads to an anoxic zone growth in the bottom horizons. This allows methane to accumulate at the bottom (Figure 5). During the summer period, the greatest spatial and temporary variability of methane fluxes is observed at the boundaries "bottom sediments - water" and "water - atmosphere". In general, in spring methane fluxes start to increase gradually (especially the bubble component), and this continues to grow in summer until the beginning of the autumn mixing stage of the reservoir. Decrease in the air temperatures leads to intense convection that erodes the density stratification. This convective mixing is combined with some storm events. The huge amount of methane accumulated in the bottom layers (when the highest concentrations of methane are observed near the bottom (Figure 5)) can reach the atmosphere. This is the second group of high values in the methane emission distribution during the year (Figure 5).

The diffusion flux is much lower than the bubble flux because of oxidation by methanotrophic microorganisms in the oxygen-saturated horizons. The contribution of the diffusive flux is 5 - 15% of the total flux (which is confirmed by the modeling results). Therefore, the growth of the diffusion component occurs only during the periods of the anoxic zone and the subsequent growth of the concentration of methane in it. The highest value of the diffusion flux, 100.3 mg CH_4*m^{-2}*day^{-1}, was recorded in the autumn mixing stage of 2018. On the same day, the maximum value of the bubble flux was also recorded (223 mg CH_4*m^{-2}*day^{-1}).

After the autumn convection stage and subsequent homothermy, the same as in spring, the methane flux sharply decreases and rare high values are observed at the moments of intensive mixing of the reservoir. There may also be an increase in methane flux at the beginning of winter if heating has destroyed the ice cover formed in late autumn, such as in 2018.

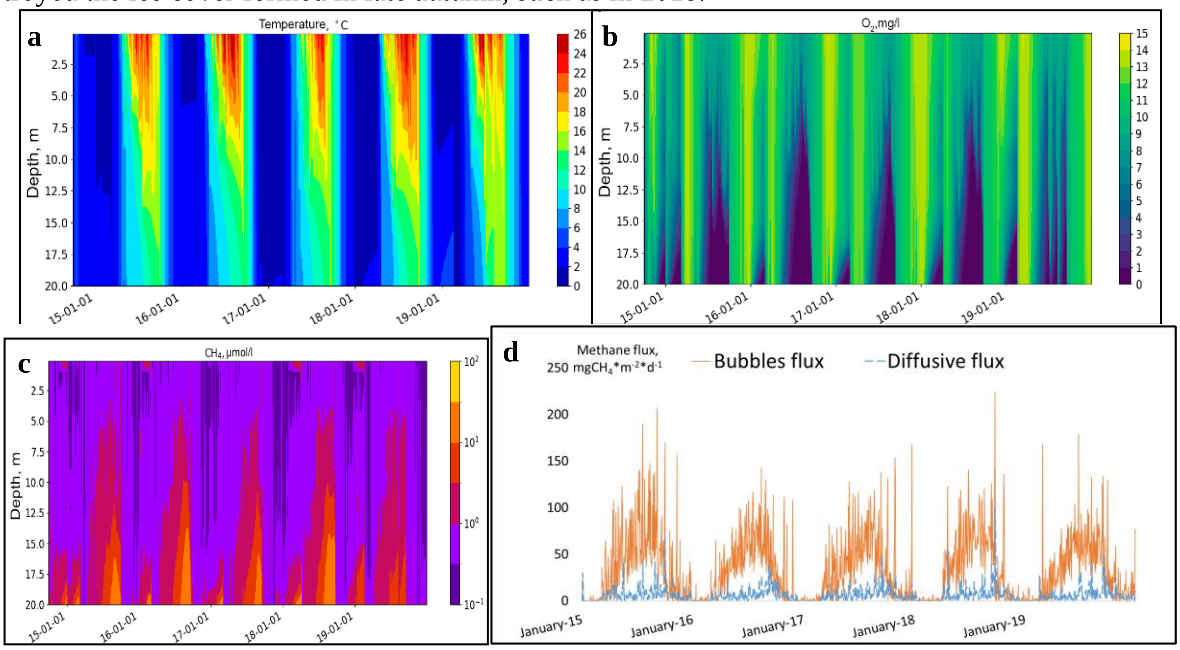

Figure 5. Temperature (a), oxygen (b) and methane (c) concentration distribution in vertical and time series of diffusive and bubble flux (d). Results of LAKE model.

This paper reports preliminary calculations using model LAKE for the Mozhaysk reservoir. In the future, calibration and verification of the model parameters will be done to obtain more accurate results. Nevertheless, model LAKE reproduces the thermodynamic and biochemical processes occurring in the water body adequately already at this stage. Quantitative estimates of methane emissions into the atmosphere were obtained for 2017-2019 based on the results of field observations. Also, these values were estimated based on the modeling results (Table 2).

Table 2. Comparison of estimates of annual methane emissions in carbon equivalent obtained from the model and on the basis of field observations.

Year	Emission by LAKE, tons C-CH_4	Emission by observations, tons C-CH_4
2017	308	425
2018	312	269
2019	305	596

It should be noted that the estimates based on the modeling results will be corrected at later stages of this work. Also, it is necessary to take into account large inaccuracy of the empirical estimates associated with a limited number of observations and the impossibility to carry out observations during periods of atmospheric storms or ice-off. Hence, so far we state similarity of the estimates made by both methods at a qualitative level. The work will be aimed at obtaining more accurate and reliable results in future.

5. Conclusions

The dynamics of methane fluxes from an artificial Mozhaysk reservoir into the atmosphere was successfully simulated using a one-dimensional hydrodynamic model, LAKE. Comparing annual methane emissions estimated from field observations performed since 2015 at the Mozhaysk reservoir, it can be argued that the model adequately reproduces the processes occurring in the reservoir, even without calibration specific for a given water body. However, it is necessary to calibrate the model to obtain more accurate parameter values and reliable results. A statistical analysis of the input meteorological characteristics for the model showed that it is possible to use reanalysis data in the calculation. This shows that such calculations can be carried out for any water body with sufficient accuracy. In the future, this technique will make it possible to obtain reliable estimates of methane emissions from reservoirs in different regions.

Acknowledgments

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