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Numerical Modeling of Methane Emissions from Lakes in the Permafrost Zone

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Abstract—A brief review of published observations of methane fluxes to the atmosphere from bogs and lakes in the permafrost zone is presented. Approaches to modeling the emission of methane from bogs are considered, and their advantages and shortcomings, in particular, from the point of view of their coupling to climate models, are outlined. A one-dimensional model developed by the authors for methane generation, transport, and sink in the ground–water body system and coupled to a hydrothermodynamic model of a water body is described. The approaches used in analogous models for bogs as well as new parametrizations describing lake-specific processes are applied. A parametrization of methane generation in vicinity the lower boundary of the thawed ground zone underneath a water body (talik) is suggested. The results of calibrating this model against available observations of methane emission from the thermokarst Shuchi Lake in northeastern Siberia are discussed.

Keywords: methane, permafrost, lakes, numerical modeling.
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INTRODUCTION

In the climate system, methane is an important greenhouse gas that determines atmospheric chemistry: its direct global warming potential is 39 times the global warming potential of carbon dioxide [1]. The major source of methane input into the atmosphere is its release from the underlying surface (primarily from the land surface). The dynamics of atmospheric methane concentration has already been included in several modern climate models (see, for example, [2, 3]). To simulate the cycle of methane in the atmosphere correctly, however, it is necessary to calculate its geographically distributed emission from the land surface in order to use this information further as boundary conditions in general circulation and atmospheric chemistry models. Moreover, even if reliable estimates of methane emission were to exist now, its prediction for the future could not be done using only observational data. This because the processes resulting in methane production and release into the atmosphere depend highly and nonlinearly on the temperature of environments in which methane is produced (soil) and across which it is transported (soil or a water body), on soil moisture, on the amount and quality of organic matter available for decomposition, etc. This means that the total change in methane emission into the atmosphere in the future cannot be predicted only from a forecast of a global mean or even regional change in air temperature and in other climatic characteristics.

Obtaining an estimate of future methane fluxes from the land surface to the atmosphere requires mathematical models that would relate the physical and biochemical processes producing the methane flux across the surface of soil or a water body at any point of the globe to climatic and biochemical characteristics at this point. The processes occurring in the permafrost zone are of particular importance. It must be noted that models of methane production, transport, and emission from wetland surfaces (including the permafrost zone) have been developed in the last 20 to 30 years, and some of them have been thoroughly calibrated against observed data. At the same time, much less attention has been paid to mathematical modeling methane emission from lakes and, in particular, we are unaware of any parametrizations of this process that are used in climate models. Recent observations, however, suggest the importance of this source in the climate system. In connection with this, the aim of this paper is to develop a one-dimensional water-

column methane transport model coupled to a model of heat and moisture transport and to a biochemical model of the methane cycle (production, oxidation, and transport) in bottom sediments and in the underlying frozen ground, which would be suitable for use in climate models.

This paper is organized as follows. In Section 2, field data on methane fluxes from bogs and lakes in the permafrost zone are briefly reviewed and an analysis of existing methods for calculating the emission of methane is presented. Section 3 describes a hydrothermodynamic model of a water body and a modified model of methane production and transport in the ground and of methane oxidation and transport in the water column. The basis for the latter was the model from [4] as one of the most commonly used and observationally tested models. Results from numerical experiments and calibrating the model against available field data are discussed in Section 4. The main findings of the study are given in the Conclusions.

2. OBSERVATIONS AND METHODS FOR CALCULATING METHANE EMISSIONS FROM BOGS AND LAKES

We have summarized the material of papers [5–10], which, in turn, employed data from 16 publications devoted to assessing global methane sources. Natural sources are tropical swamps and northern-latitude peatbogs, termites, ruminants, volcanoes and hydrothermal sources, and oceans and freshwater lakes (in particular, the recently discovered emission from the bottom of thermokarst lakes in the permafrost zone of eastern Siberia, Canada, and Alaska [11]); in addition, the possibility of methane production by plants under aerobic conditions is being discussed in the literature. According to modern ideas, bogs are the major natural sources of methane (113 Mt CH_4/year out of 530 Mt CH_4/year of the total emission to the atmosphere [5–10]. Although tropical regions account for a significant fraction of the global wetland emission, northern bogs are of particular interest, because the most distinct climate changes and related changes in the area of bogs and in their carbon budget are expected to take place and are already occurring at high latitudes.

Whereas there are numerous field data on methane emissions from bogs (for example, in western Siberia [12–15]), experimental studies of the methane processes in lakes are less satisfactory. Measurements of the methane emission from the surface of wetland lakes have been a focus of relatively few papers [16–23]. In particular, estimates of the methane flux from the open water of 42 lakes in the Yukon–Kuskokwim Delta (Alaska) are presented in [19]. This flux averaged 77 ± 10 mg m^{-2} day^{-1} for small lakes and 3.8 ± 0.8 mg m^{-2} day^{-1} for large lakes. From airborne measurements in the same area, the average value of the flux was 57 ± 6 mg m^{-2} day^{-1} [20]. In [21], mean methane fluxes from three small wetland lakes located in the middle taiga and forest tundra zones of western Siberia ranged from 1.1 to 120 mg m^{-2} day^{-1}, while in [22] the fluxes from wetland lakes in forest tundra, northern taiga, and subtaiga of western Siberia were 17.6, 44.8, and 169.7 mg m^{-2} day^{-1}, respectively. All these values are comparable in order of magnitude to the methane flux from bogs, with methane fluxes from wetland lakes often being maximal among all the elements of a wetland landscape.

There are at least three mechanisms by which CH_4 is transported from soil to the atmosphere: the diffusion of methane in a dissolved form (turbulent diffusion in a water basin), ebullition, and transport through plants [23]. Bubbles may be 6–24% [21] or even 60% [17] methane, which indicates the importance of this transport mechanism. Measurements of CH_4 bubbling from 16 northern lakes of different types (including thermokarst lakes) are given in [24]; the extrapolation of these data to all northern lakes produced a value of about 24.2 ± 10.5 Mt/year, while in an earlier study [9] the uncertainty in CH_4 flux for tundra ranges from 1 to 5 Mt/year without bubbling from point sources (strong and narrow streams of bubbles). In connection with this, it must be noted that floating chambers often used for measuring the methane flux at the lake surface have one fundamental drawback: the probability that a chamber will be able to measure a flux from point sources is low because these sources occupy a very small fraction of the water table of a water body. Therefore, estimates of methane emission from lakes measured with this method may be well underestimated.

The role that lakes play in the formation of the methane flux from the underlying surface to the atmosphere is determined by their being significant sources of CH_4 throughout almost the whole year, unlike bogs. In the cold period, the active layer of bogs freezes up and, hence, methane generation nearly stops there [25]. At the same time, methane generation in a thawed layer (talik) below water basins continues year round (from the data of [11], the freeze-up period accounts for as much as 47% of the annual methane flux). Given this effect and the fact that lakes are common in the permafrost zone, it seems important to consider the contribution of lakes to regional empirical estimates of methane fluxes and to develop specific models of methane emission from lakes, in particular, for their further use in climate models.

At present there is a hierarchy of models developed for calculating methane emission from the soil and bog surface that differ in complexity and details of describing processes depending on the tasks they are designed to solve. To our knowledge, however, analogous models for methane transport in water bodies have hardly been developed at all.

For soil, the simplest models are those that are based on the empirical dependences of methane emission on any soil parameters, for example, the temper-

ature at a certain depth, groundwater level, and/or precipitation (see [26, 27]). Such empirical dependences are derived for particular areas in a particular season and are used to model short-term changes in the methane flux over a given area. It is well known, however, that if vast areas and large timescales are considered, most of the variability of methane flux cannot be described using only these parameters [27]. There appears to be no single regression that would be suitable for use in all geographic regions of the earth or for any season.

A more universal methodology is the one that uses vertically one-dimensional models to calculate the rate of soil methane production and oxidation from a specified groundwater level, vertical temperature profile, and some other parameters characterizing the organic matter of soil [4]. Biological processes in these models are represented by chemical reactions governed by Michaelis–Menten kinetics [28]. In particular, in the model given in [29], the dynamics of soil carbon of different types is described explicitly, but the physical processes of methane transport are not resolved explicitly (methane emission is calculated as the difference between integral production and oxidation). There are, however, models [4, 30] developed for a more general case in which the processes of vertical methane transport are already resolved explicitly. In [30], the limiting influence that the constrained oxygen concentration in soil has on methane oxidation under aerobic conditions is taken into account, but the ebullition transport of methane is omitted. This model was developed to calculate methane emissions from the surface of peatbogs, where, from the point of view of its authors, the transport of methane by plants has a dominant role. In [4], the limiting influence of oxygen is omitted, but, in addition to plant transport, the ebullition and diffusion transports are taken into account. The model in [31] is a combination of model algorithms [4] describing methane production, oxidation, and transport with the algorithms adopted from [32] and describing the limiting influence of some additional factors. All these models cannot be regarded as universal either because they contain a number of parameters in which the information on the chemical composition of soil and plant residues is hidden and whose values are determined by calibrating the model against observed data.

At the other extreme with respect to complexity lie models that thoroughly describe all the kinetics of methane production from original organic matter and the dynamics of microbial populations. They are effectively used in simulating bioreactor processes [33], and efforts to adjust such models to land ecosystems have been undertaken in recent decades [34]. Models of this kind, however, contain a large amount of parameters (for example, 37 in the model described in [34]), some of which characterize biochemical reactions in general and others are calibration parameters and depend on the properties of a given area. All these parameters are known to a limited accuracy, and, in addition, soil properties display a high spatial heterogeneity. This increases the uncertainly in the results and may lead to the loss of advantages of the models of this kind in global climate simulation. From experiments on the sensitivity of their model to the perturbation of its different parameters (in the Florida Everglades), the authors of [34] concluded that the amount of available organic matter, the diffusion rate of oxygen, and methane oxidation rate had the greatest influence on methane emission, while the variability of parameters describing other processes could be neglected. It is therefore possible that a model which is an order of magnitude less complex but describes these most important processes will be able to simulate observed values of methane emission rather accurately.

Thus, the most optimal approach to calculating methane emissions for simulating global climate changes is to develop a model that is not too complicated or detailed and that could hardly be supplied with input parameters accurately, yet not too simple and empirical, in which many physical and biochemical processes are not resolved explicitly and which would be suitable only for a particular area. In connection with this, when constructing a model of methane generation, transport, and emission in water bodies of the permafrost zone, we sought to reach a reasonable compromise between the universality (complexity) of the model and the minimization of the number of undefined (calibration) parameters.

3. MODEL OF A THERMODYNAMIC REGIME AND METHANE CYCLE IN A GROUND–WATER COLUMN SYSTEM

In this study the model of methane production, transport, and sink in a ground–water column system is incorporated into a one-dimensional hydrothermodynamic model of a water body [35, 36]. The thermal state of a water body is described by a one-dimensional heat-flux equation with turbulent heat exchange and solar radiation absorption:

$$\frac{\partial T}{\partial t} = \frac{1}{h^2}\frac{\partial}{\partial \xi} k_T \frac{\partial T}{\partial \xi} + S_\xi(T) - S_1(T) - \frac{1}{c_w \rho_{w0} h}\frac{\partial I}{\partial \xi}, \quad (1)$$

where for any variable x

$$S_\xi(x) = \frac{\xi}{h}\frac{dh}{dt}\frac{\partial x}{\partial \xi}, \quad S_1(x) = \frac{1}{h} B_s \frac{\partial x}{\partial \xi}. \quad (2)$$

The following notations are used: T is temperature; t is time; h is the depth of a water body; $\xi = z/h$, where z is the vertical coordinate directed along the gravity force (downward) and measured from the surface of a water body; k_T is the thermal diffusivity coefficient equal to the sum of molecular and turbulent diffusivities; c_w is the heat capacity of water; $\rho_{w0} = 1000$ kg/m^3 is the mean density of fresh water; I is the solar flux; and B_s is the water budget at the water–air interface (precipi-

tation minus evaporation). In Eq. 1, in comparison with its more traditional form, there are terms S_ξ and S_1, which reflect the transition to the ξ-coordinate system and the movement of the upper boundary of a water body with evaporation and precipitation, respectively. The depth distribution of solar radiation is given by an exponential dependence. The turbulent thermal diffusivity is calculated using a one-dimensional E-ε parameterization [37]. The results of numerical model experiments to simulate free convection in a layer of fluid cooled from above [38] and the penetration of a turbulent layer into stratified fluid [39] have demonstrated the adequacy of the model and consistency with other models [37].

This model calculates a change in the thickness of the ice layer over time and the propagation of heat in the layer according to an equation analogous to (1) but with a molecular thermal diffusivity coefficient. If ice is covered with snow, the depth of the snow cover and the temperature, density, and liquid water content in the snow are calculated using a model developed at the Institute of Numerical Mathematics, Russian Academy of Sciences (INM RAS) [40]. The temperature, moisture, and ice content in the ground beneath a basin are described by a system of heat- and moisture-transfer equations with water phase transitions, which was implemented in the INM RAS climate model [41] (by setting the water-vapor content equal to zero). An adequate description of the heat regime of the ground is important for the accuracy of calculating methane generation, because the latter depends exponentially on temperature. The model considered above participates in the Lake Model Intercomparison Project (LakeMIP) [42].

3.1. Methane Production, Transport, and Sink in Bottom Sediments and in the Ground beneath a Lake

The simulation of methane production, transport, and sink in bottom sediments and in the ground beneath a lake is carried out using the equation for methane concentration C_{CH_4}

$$\frac{\partial C_{CH_4}}{\partial t} = \frac{\partial}{\partial z} k_{CH_4,s} \frac{\partial C_{CH_4}}{\partial z} + P - E, \qquad (3)$$

where P is the methane generation from organic-matter anaerobic decomposition and E is the methane sink due to ebullition. Unlike the analogous equations used in models of methane production, transport, and sink in wetland ecosystems (for example, [43]), the terms in (3) that describe the oxidation of methane and its absorption by plant roots are omitted. Oxidation is neglected because the oxygen content in bottom sediments of the lake with depths of several meters is commonly low. In addition, since vegetation in oligotrophic thermokarst lakes is scarce, its effect can also be ignored to a first approximation. The molecular diffusion coefficient of dissolved methane $k_{CH_4,s}$ is calculated by taking into account the ratio of the liquid and gaseous phase in soil pores and temperature [44]. The methane flux at the lower boundary of the ground layer is assumed to be zero. The condition of coupling is used for methane transport at the interface of bottom sediments and water; that is, methane fluxes and methane concentrations on both sides of the interface are assumed to be equal.

Ebullition occurs when the methane concentration exceeds the critical value $C_{CH_4,cr}$, which is determined by atmospheric pressure p_a, by the water-column hydrostatic pressure dependent on the lake depth h, by nitrogen concentration C_{N_2}, and by porosity Π:

$$E = \max\{0, c_e[C_{CH_4} - \alpha_e C_{CH_4,cr}(p_a, h, C_{N_2}, \Pi)]\}. \qquad (4)$$

Here, $c_e = 2.78 \times 10^{-4}$ s^{-1} is the constant that determines the ebullition rate [44] and α_e is the relative concentration at which ebullition begins (according to [43], it is taken to be equal to 0.4). The critical concentration is the concentration at which the sum of pressures of gases that are in equilibrium with a solution of soil moisture by the Henry law is equal to the environmental pressure, i.e., to the sum of air pressure, the hydrostatic pressure of the water column, and corrections induced by capillary and osmotic forces in the soil [45]. Because the Henry's constants used in the model are measured for a flat surface of water and the surface of bubbles is spherical, the coefficient α_e is designed to take into account the sphericity effect (the equilibrium pressure of gas over a concave surface of a bubble is higher than that over a flat surface of solution), along with the above corrections for pressure. The formula for $C_{CH_4,cr}$ is given in Appendix A.

It is assumed in the model that all the bubbles formed in the ground and bottom sediments reach the lake surface instantaneously without changing their gas composition (this assumption is justified in 3.2). Then the ebullition flux of methane on the open water surface F_a can be written as

$$F_a = \int_0^{h_s} E dz,$$

where h_s is the thickness of a ground layer beneath a lake. In winter, during a freeze-up period, part of the bubbles are trapped by ice and frozen into it as typical clusters. The ice-trapped ebullition flux $F_{a,i}$, is

$$F_{a,i} = k_{tr} \int_0^{h_s} E dz,$$

where k_{tr} is a constant (its value is explained in Section 4). It is assumed that the amount of methane accumulated over winter in ice is instantaneously released into the atmosphere when the ice layer disappears.

The term P responsible for methane generation should reflect the effects of decomposition of two species of organics, first of all the new organic matter that settles onto the bottom of the lake when its ecosystem is functioning. In this case it can be assumed that the corresponding portion of methane generation is proportional to the ecosystem productivity. In addition, the new organic matter erodes into the bottom when shores are abraded by wind-driven waves. Formulas for methane generation due to the decomposition of new organic matter were derived in several studies on wetland ecosystems (for example, [44]), and one such formula is used in our model (see Eq. (6) below). Another kind of organic is old organic matter, which is conserved in permafrost and falls into a region of positive temperatures when the talik deepens [11]. Experimentally, the old organic matter differs from new organic matter in the ratio of carbon isotope ^{13}C to ^{14}C.

In connection with the above, it can be assumed that

$$P = P_{new} + P_{old}, \tag{5}$$

with methane generation due to the decomposition of new organic matter P_{new} being determined by

$$P_{new} = P_{new,0} e^{-\alpha_{new} z_s} q_0^{T/T_0} H(T), \tag{6}$$

where the temperature T is in degrees Celsius; $H(T)$ is the Heaviside function; α_{new} is the parameter that determines the rate of decrease in methane generation with depth (the choice of its value is explained in Section 4); T_0 and q_0 are constants equal to 10°C and 6 units, respectively, in accordance with [44]; $P_{new,0}$ is a calibration parameter (the choice of its value is explained in Section 4); and z_s is the depth measured from the lake bottom.

To derive a formula describing the rate of methane generation due to the decomposition of old organic matter P_{old}, it makes sense to use the following considerations. It is natural to assume that

$$P_{old} = P_{old,0} q_0^{T/T_0} H(T), \tag{7}$$

where the constant $P_{old,0}$ is proportional to ρ_{old}, the density of old organic matter (kg/m^3) available for decomposition. This density decreases as the organic matter decomposes, the decomposition rate being described by the Michaelis–Menten equation

$$\frac{d\rho_{old}}{dt} = -\frac{V\rho_{old}}{\alpha + \rho_{old}}, \tag{8}$$

where two new parameters are introduced, one is V describing the first-order decomposition rate when $\rho_{old} \to \infty$, and another is the half-saturation constant α (parameter values are given in Section 4). Using (7) and (8), we can obtain (Appendix B) the following expression for the rate of methane generation due to the decomposition of old organic matter

$$P_{old} = P^*_{old,0}\rho_{old,0}\left[2 + \lambda_\rho - \sqrt{(1+\lambda_\rho)^2 + 2\gamma_\rho C_t^{-2}(h_t^2 - z_s^2)}\right] q_0^{T/T_0} H(T), \tag{9}$$

where $\lambda_\rho = \rho_{old,0}\alpha^{-1}$, $\gamma_\rho = V\alpha^{-1}$, h_t is the talik thickness; and $\rho_{old,0}$ is the density of old organic matter at the beginning of its decomposition, which is equal to the density of organic matter beneath the talik.

Parameters $P_{new,0}$ and $P^*_{old,0}$ here are calibration parameters, and their values are given in Section 4 in a description of the results of model validation and calibration.

3.2. Transport and Sink of Dissolved Methane in the Water Column

Methane in the water column is transported in bubbles rising from the lake bottom and in a dissolved form. When methane bubbles rise, gas exchange occurs at the interface of the liquid and gaseous phase. Because of gas exchange, the gas composition of bubbles changes so that the ebullition flux of methane on the lake surface differs from that on the bottom. To estimate the magnitude of this effect, we use a model of the gas composition of bubbles proposed in [46] and implemented as the SiBu-GUI software [47]. According to calculations with this model, at water temperatures of 4–10°C and lake depths of 3–10 m (typical temperatures and depths of thermokarst lakes), the bubbles formed on the bottom and consisting only of methane contain no less than 89% of methane when they reach the lake surface. Thus, the change in the gas composition of bubbles can be neglected to a first approximation. The rising bubbles also form a water circulation structurally analogous to thermals, which is called "bubble convection." This circulation induces the additional mixing of the water column. This could be particularly important in winter conditions, when turbulent mixing without bubbles would be negligible. In the present model, however, the bubble convection effect is omitted because it evidently has no significant influence on processes of methane generation.

Methane oxidation occurs when there are methanotrophic bacteria and, to a first approximation, can be described by the following stoichiometric relationship:

$$CH_4 + 2O_2 = 2H_2O + CO_2. \tag{10}$$

The kinetics of methane oxidation is commonly calculated using the Michaelis–Menten equation [48], which in some models of methane processes is written under the assumption of a very high (formally infinite) oxygen concentration. For the lake, it makes sense to reject this assumption because there are often

very low oxygen concentrations in the near-bottom layers (particularly in wintertime). In connection with this, the complete equation for the methane concentration in the water column can be written as

$$\frac{\partial C_{CH_4}}{\partial t} = \frac{1}{h^2}\frac{\partial}{\partial \xi} k_{CH_4} \frac{\partial C_{CH_4}}{\partial \xi} + S_\xi(C_{CH_4}) - S_1(C_{CH_4}) - V_{oxid}(T)\frac{C_{O_2}}{k_{MM,O_2} + C_{O_2}} \frac{C_{CH_4}}{k_{MM,CH_4} + C_{CH_4}}. \quad (11)$$

Here, on the right-hand side, the Michaelis–Menten term appeared with half-saturation constants in the denominator and the velocity $V_{oxid}(T)$, which is dependent on temperature according to the Arrhenius equation. The dissolved oxygen C_{O_2} is considered only in the water column because the concentration near the bottom, in bottom sediments, and in the underlying ground can be taken close to zero.

Different models of oxygen dynamics in lakes have been proposed (see, e.g., [49]) in which turbulent oxygen diffusion, photosynthesis, sink of oxygen for organic-matter decomposition, etc., are taken into account. In this model, the source of oxygen due to photosynthesis is omitted because, according to observations [50], thermokarst lakes are mostly acidic and the plankton responsible for photosynthesis hardly develops there. This simplification allows a number of empirical constants that are commonly included in parameterizations of the photosynthesis process to be excluded from the model. In addition, the oligotrophicity of thermokarst lakes makes it possible to neglect oxygen expenditures on the decomposition of dead organics. With these simplifications, the equation for oxygen concentration becomes

$$\frac{\partial C_{O_2}}{\partial t} = \frac{1}{h^2}\frac{\partial}{\partial \xi} k_{O_2} \frac{\partial C_{O_2}}{\partial \xi} + S_\xi(C_{O_2}) - S_1(C_{O_2}) - 2V_{oxid}(T)\frac{C_{O_2}}{k_{MM,O_2} + C_{O_2}} \frac{C_{CH_4}}{k_{MM,CH_4} + C_{CH_4}}. \quad (12)$$

The equation for the transport of carbon dioxide resembles Eq. (12) to the accuracy of designations, but with a positive sign in the last term on the right-hand side.

For the coefficients of turbulent diffusion of dissolved gases, it is assumed that $k_{CH_4} = k_{O_2} = k_{CO_2} = k_T$. The fluxes of dissolved oxygen and carbon dioxide at the bottom are taken equal to zero. At the water–air interface, the fluxes of gases are specified, which depend on the gradient of their concentration in the near water surface air layer and on the wind speed [51].

4. NUMERICAL EXPERIMENTS AND MODEL CALIBRATION

Measurements of methane fluxes over lakes in the permafrost zone are episodic, which does not allow the model to be calibrated and validated using a large amount of sites representative of different climatic conditions. To the best of our knowledge, Walter et al. [11] produced the longest series of high time resolution at two thaw lakes in northeastern Siberia. The results of measurements of the ebullition flux of methane at one of these lakes (Shuchi Lake) were chosen in our study to calibrate and validate the model. These data span a period from April 28, 2003, through June 30, 2004, with a time interval of 1 h. The data, however, have significant gaps, particularly in wintertime. Measurements were taken in different parts of the lake differing in depth and intensity of the thermokarst process. Because meteorological observations in the vicinity of the lake were not carried out (except for measurements of air pressure), the atmospheric model forcing consisted of time series of basic meteorological elements in the surface layer, which were generated from the ERA-Interim reanalysis data (http://www.ecmwf.int). Fluxes of total solar radiation and atmospheric radiation were calculated from semiempirical formulas [54]. The depth of the lake in the model was 8 m, which can be regarded as an estimate of the average depth (the maximum depth is 11 m).

Evidently, the horizontal variability of methane fluxes within a lake cannot be simulated using a vertically one-dimensional model. Therefore, the aim of model calibration was to make the calculated lake-averaged total annual amount of methane emitted to the atmosphere close to the value estimated from observed data.

Proceeding to the choice of model parameters, we note that it is possible to estimate from measurements at Shuchi Lake that the winter methane flux through unfrozen holes in ice is ~0.1 of the total winter emission. It is therefore appropriate to take the constant k_{tr} (the fraction of bubbles that are formed in the talik underneath the lake mostly from methane and are trapped in wintertime by ice cover) equal to 0.9. The half-saturation constant $\alpha = 0.3$ kg/m^3 from the Michaelis–Menten equation for the decomposition of old organic matter was chosen following [53], and the parameter V was set equal to 2×10^{-3} kg/(m^3 year) in accordance with an estimate from the data of [54]. The organic-matter density beneath the talik $\rho_{old,0}$ was assumed equal to 18 kg/m^3, which is a typical value for the permafrost zone. The value of α_{new} was set equal to 3 m^{-1}, unlike 5 m^{-1} in the model [44]. The smaller value of the rate of decrease in methane generation with depth should take into account the input of organic matter from eroded shores, which, when it accumulates on the bottom, evidently forms a vertically near-homogeneous environment with active methane generation.

$P_{new,0}$ and $P^*_{old,0}$ are undefined (calibration) parameters in the model. The values of $P_{new,0}$ and $P^*_{old,0}$ were

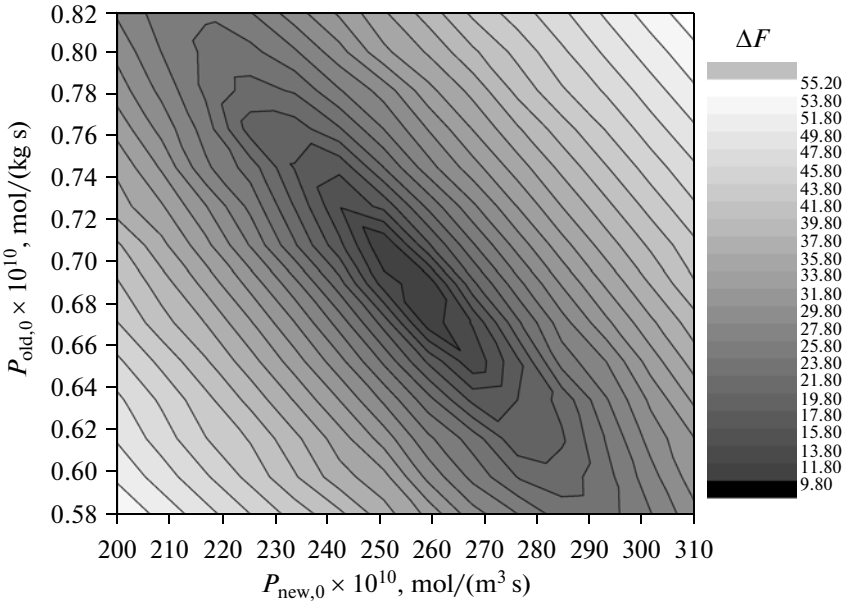

Fig. 1. Model-simulated methane emission error ΔF as a function of parameters $P_{new,0}$ and $P^*_{old,0}$.

chosen so that the function $\Delta F^2 \equiv \left(F_a^w - F_{a,m}^w\right)^2 + \left(F_a^s - F_{a,m}^s\right)^2$ $(\Delta F > 0)$ should have a minimum. The superscripts *w* and *s* here denote the total methane flux over the open-water period and the freeze-up period, respectively, and the subscript *m* denotes measured values. When this function is zero, the model accurately simulates the annual methane flux and its distribution between the warm and cold seasons. The dependence of the error on calibration parameters, $\Delta F\left(P_{new,0},\ P^*_{old,0}\right)$, is shown in Fig. 1. The curve is plotted by the data of 196 numerical experiments (196 pairs of parameter values) using MPI parallel programming technology. As can be seen, the function obtained from calculations has a single minimum, which is supported by numerical experiments over a wider range of parameters than what is shown in the figure.

A comparison of the annual and seasonal methane emissions (from June 1, 2003, through June 1, 2004) at Shuchi Lake from the model results and from measurements for the optimal calibration parameters $P_{new,\,0} = 2.55 \times 10^{-8}$ mol/(m^3 s) and $P^*_{old,0} = 6.9 \times 10^{-11}$ mol/(kg s) is presented is Table 1.

As can be seen from Table 1, calibration provided a very close coincidence between the modeled and measured values.

The results of field measurements of the methane flux at Shuchi Lake from point sources (which are narrow intense streams of bubbles) and of the background ebullition flux are shown in Figs. 2 and 3, respectively. Despite significant gaps in winter observations, it can be seen that there is no distinct annual cycle of the ebullition methane flux from point sources. This is natural because these sources (particularly hotspots) are formed at large depths below the bottom of the lake, where the temperature varies little during the year. At the same time, the background ebullition flux has a maximum in summer (Fig. 3), as does the total ebullition flux [11]. The model reproduces this maximum (Fig. 4), though with a shift in time.

Table 1. Characteristics of the annual methane emission to the atmosphere at Shuchi Lake

Source of data	Annual methane emission, mg/(m^2 year)	Fraction of emission in the open-water period, %	Fraction of emission in the freeze-up period, %
Observations [11][1]	22658[1]	54	46
Model	22588	54	46

Note: [1] Unfortunately, the authors of [11] do not report the accuracy of estimates of methane emission sums by seasons and over the year in general, so the fifth sign in the numbers presented in Table 1 should not be associated with the accuracy of the estimate of these characteristics.

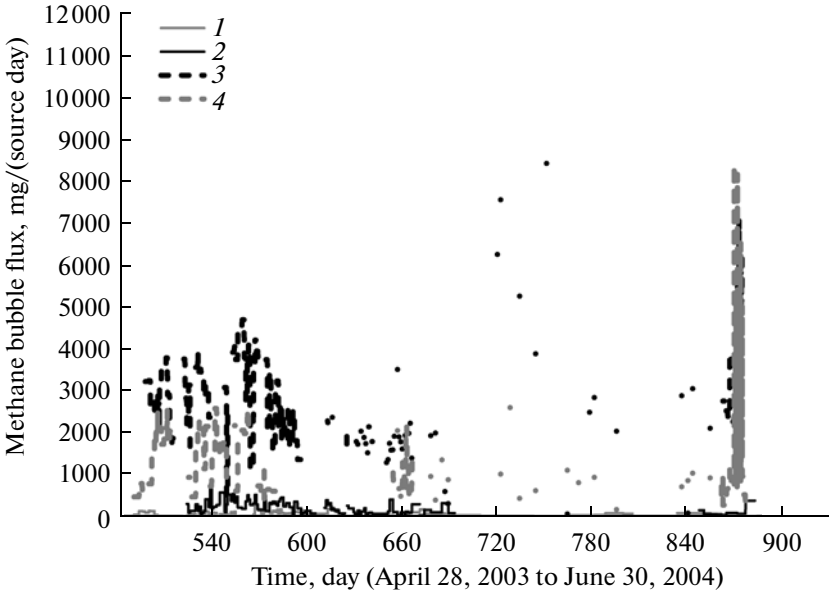

Fig. 2. Point-source and hotspot methane bubble fluxes in four parts of Shuchi Lake (northeastern Siberia) from measurements [11] (April 28, 2003, to June 30, 2004). (*1*) Thermokarst margin in a deep-water part (depth 7.4 m), point source; (*2*) thermokarst margin in a shallow-water part (depth 1.75 m), point source); (*3*) thermokarst (depth 2.25 m), hotspot; and (*4*) thermokarst in deep water (depth 4.75 m), hotspot.

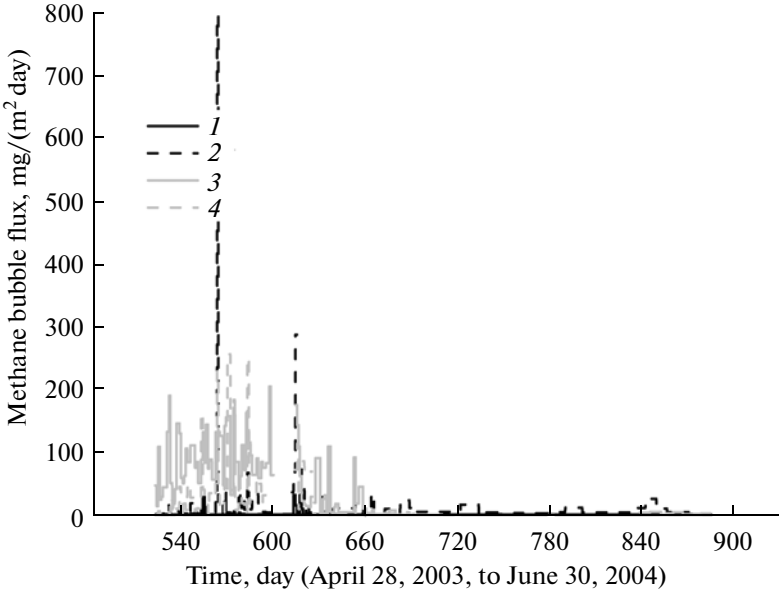

Fig. 3. Background bubble flux in four parts of Shuchi Lake (northeastern Siberia) from measurements [11] (April 28, 2003, to June 30, 2004). (*1*) Center (depth 8 m); (2) thermokarst-free area (depth 1.75 m); (*3*) thermokarst margin (depth 1.5 m); (*4*) margin of the thermokarst-free area (depth 1.4 m).

From the point of view of testing the physical model adequacy, the proportion of the amount of young and old methane in the total methane flux from the lake to the atmosphere is also of interest. Since the model calculates only the integral methane flux, for an estimate of the emission of the two types of this gas it is natural to use the total production of these types of methane $\tilde{P}_{new} = \int_0^T \int_0^{h_s} P_{new} dz dt$ and $\tilde{P}_{old} = \int_0^T \int_0^{h_s} P_{old} dz dt$,

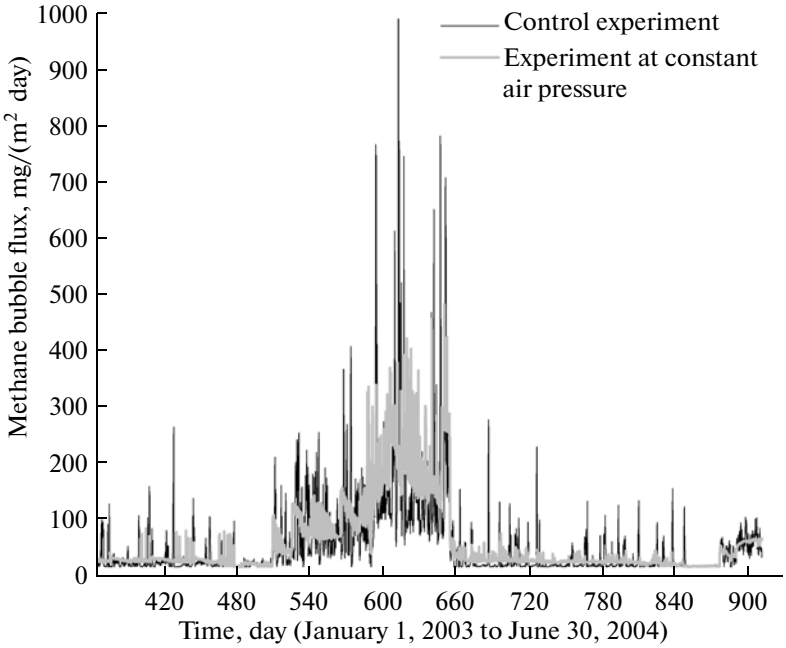

Fig. 4. Area-mean bubble flux at Shuchi Lake from model results (January 1, 2003, to June 30, 2004).

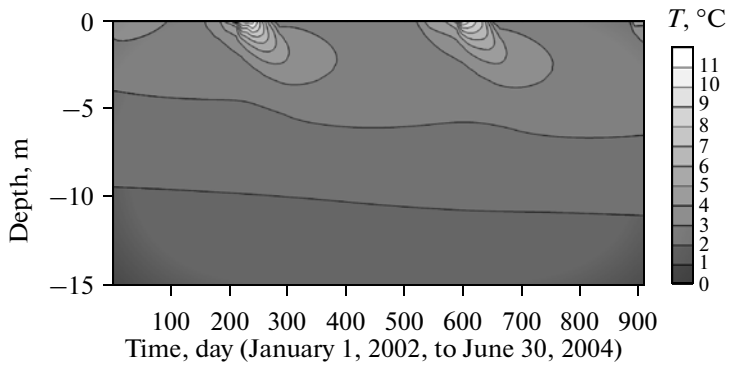

Fig. 5. Thermal isopleths in the talik beneath Shuchi Lake from model results (January 1, 2002, to June 30, 2004).

where T in this case is the open-water or freeze-up period (integral production over such long periods is nearly equal to the integral emission). A comparison of these values and observations is presented in Table 2.

The values in Table 2 demonstrate a qualitative consistency. An abrupt increase in the fraction of young methane in summer emission is explained by the well-known laws of heat transfer in the soil, in our case, in the talik (Fig. 5). As is seen from the figure, the summer temperature maximum is at the bottom of the lake, so intense methanogenesis, which consumes new organic matter that deposits on the bottom when the ecosystem is functioning and shores are abraded, occurs there. In winter, the maximum moves to a depth of a few meters below the lake bottom, where methanogenesis is due to the decomposition of the older organic matter.

Table 2. Fraction of young and old methane in the methane flux at Shuchi Lake from model results and observations

Source of data	Open water	Freeze-up
Fraction of young methane in ($^{14}CH_4$) emission, % (from observations [11])	47	6
Fraction of young methane in the total methane generation $\tilde{P}_{new}/(\tilde{P}_{new} + \tilde{P}_{old})$, %	61	32

It is known [55] that significant fraction of the time variability of methane generation in peatlands is caused by fluctuations of air pressure. It is important that the model simulates this effect, too. The numerical model experiment at a specified air pressure of 1000 hPa produced the result shown in Fig. 4. The

variability of methane flux decreases significantly at constant pressure. Evidently, fluctuations of air pressure induce fluctuations of the critical methane concentration in accordance with Eq. 4, leading, in turn, to variations in the ebullition emission of methane.

In conclusion, there are several comments on the results of model calibration. It is evident that the model's error measure ΔF depends on many parameters, which can be divided into three groups:

(1) Calibration parameters ($P_{new,0}$ and $P^*_{old,0}$);

(2) Physical parameters whose values for a particular lake are unknown or largely uncertain. Among them are the average depth of the lake and the extinction coefficient of solar radiation in water, on which the temperature at the lake bottom and below it and, hence, the rate of methane generation depend heavily;

(3) Boundary conditions for the model, namely, the time series of meteorological elements, which are not measured near the lake, as was noted above.

At the same time, ΔF is formally minimized as a function of only calibration parameters ($P_{new,0}$ and $P^*_{old,0}$). Therefore, if the physical parameters and boundary conditions were known with a better accuracy, the optimal $P_{new,0}$ and $P^*_{old,0}$ would have taken other values. Hence, the values of $P_{new,0}$ and $P^*_{old,0}$ obtained here may be regarded as approximate, and they should be further checked at other lakes, possibly again at Shuchi Lake if the model is better supplied with input data.

5. CONCLUSIONS

A numerical one-dimensional model of methane generation, transport, and sink in a ground–lake system is presented. Based on the diffusion equation with sources, this model simulates the generation of methane in the ground, its molecular diffusion, and its emission due to ebullition. The main feature of this model which differentiates it from analogous models for wetlands is that methane generation is represented as the sum of generation due to decomposition of new and old organic matter. The input of new organic matter is determined by lake productivity and by the abrasion of shores, while old organic matter falls into the zone of a positive temperature (thus becoming available for decomposition) when the talik deepens. The transport of dissolved methane in the water column is described by the equation of turbulent diffusion with a sink responsible for methane oxidation. This model is included in the one-dimensional hydrothermodynamic model of a water body [35].

A hydrothermodynamic model with a scheme of methane generation, transport, and sink has been validated and calibrated against the ERA-Interim reanalysis data and observations [11] of methane flux at Shuchi Lake (2003–2004). After the parameters were calibrated, good consistency has been found between the modeling results and field data in terms of the annual methane emission into the atmosphere, the distribution of emission between the open-water and freeze-up periods, and the ratio of the amount of methane formed by the decomposition of new and old organic matter. In addition, this model has qualitatively well reproduced the well-known effect of pressure fluctuations on the time variability of methane emission [55]. We expect that, in the future, as reliable meteorological data are made available, a comparison will be made between field and model-simulated time series of methane emission. It is also planned to incorporate this model as one of the components of the land surface scheme of a climate model.

APPENDIX A

CRITICAL CONCENTRATION OF SOIL METHANE

The critical concentration of methane in the ground can be calculated in the following way. Let bubbles formed in the ground consist only of methane and nitrogen, so that the balance of pressures inside a bubble and in the environment is satisfied

$$p_{\mathrm{CH}_4} + p_{\mathrm{N}_2} = p_a + p_l + p_*,$$

where p_a is air pressure; $p_l = \rho_{w0} g h$ is the hydrostatic pressure of the water column; p_{CH_4} and p_{N_2} are partial pressures of methane and nitrogen, respectively; and p_* is the term describing other effects, in particular, the soil pressure distribution due to capillary and osmotic forces. The last term in the model is set equal to zero, which is compensated in part by introducing the coefficient α_e in (4). Assuming that partial pressures of gases are related to their concentrations in solution by the Henry law, it is possible to write the following relationship:

$$\begin{aligned} C_{\mathrm{CH}_4,cr}(p_a, h, C_{N_2}, \Pi) &= \Pi K_{\mathrm{H,CH}_4}(T) \\ &\times \left[p_a + \rho_{w0} g h - C_{\mathrm{N}_2} / K_{\mathrm{H,N}_2}(T)\right]. \end{aligned} \quad \text{(A. 1)}$$

Here, Π is the porosity of the grand, C_{N_2} is the concentration of nitrogen in pores, and $K_{\mathrm{H,CH}_4}$ and $K_{\mathrm{H,N}_2}$ are temperature-dependent Henry constants [56]. The nitrogen concentration in the model is given by the exponential dependence on depth in the ground [57].

APPENDIX B

RATE OF METHANE GENERATION IN A TALIK DUE TO THE DECOMPOSITION OF OLD ORGANIC MATTER ACCORDING TO MICHAELIS–MENTEN KINETICS

By integrating Eq. (8) under the initial condition

$$\rho_{old} = \rho_{old,0} \text{ at } t = t_0,$$

we can obtain the following time dependence for a change in the density of old organic matter

$$\alpha \ln \frac{\rho_{old}}{\rho_{old,0}} + \rho_{old} - \rho_{old,0} = -V(t - t_0). \quad \text{(B. 1)}$$

Using the first two terms of the Taylor expansion of the logarithm

$$\ln x \approx (x-1) - \frac{1}{2}(x-1)^2, \quad x = \frac{\rho_{old}}{\rho_{old,0}}$$

and substituting this formula into (B. 1), we obtain a quadratic equation in ρ_{old}. For the two real solutions to it, the following root corresponds to decrease of the density of old organic matter with time:

$$\rho_{old} = \rho_{old,0}\left[2 + \lambda_\rho - \sqrt{(1+\lambda_\rho)^2 + 2\gamma_\rho C_t^{-2}(h_t^2 - z_s^2)}\right], \quad \text{(B. 2)}$$

where $\lambda_\rho = \rho_{old,0}\alpha^{-1}$ and $\gamma_\rho = V\alpha^{-1}$. Relationship (B. 2) is approximate, but the numerical solution of transcendental equation (B. 1) by a chord method demonstrated a high degree of its accuracy when the values of the coefficients were consistent with real conditions of the anaerobic decomposition of organic matter.

In deriving formula (B. 2), the results were taken from [58] in which, on the basis of a numerical solution to the equation of heat conduction in the ground beneath a lake, it was shown that the deepening of the talik rather accurately matches the classical formula [59]

$$h_t = C_t\sqrt{t},$$

with $z_s = C_t\sqrt{t_0}$ (the initial organic-matter decomposition time t_0 is the time when the depth of the talik was z_s). From [58], it follows that $C_t \approx 0.5$ m/year$^{1/2}$. After substituting (B. 2) into (7), we have the following expression for the rate of methane generation by the decomposition of old organic matter:

$$P_{old} = P^*_{old,0}\rho_{old,0}\left[2 + \lambda_\rho - \sqrt{(1+\lambda_\rho)^2 + 2\gamma_\rho C_t^{-2}(h_t^2 - z_s^2)}\right] q_0^{T/T_0} H(T). \quad \text{(B. 3)}$$

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