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## NUMERICAL SIMULATION OF METHANE EMISSION FROM SUBARCTIC LAKE IN KOMI REPUBLIC (RUSSIA)

**ABSTRACT.** During last decades, a special attention has been paid to methane emission from lakes [Bastviken et al., 2004; Wik et al., 2016 and etc.] as one of the significant sources of this important greenhouse gas to the atmosphere. However, attempts to simulate methane production and efflux at the air-water interface are scarce [Stepanenko et al., 2011; Tan et al., 2015a; Tan et al., 2015b] and models proposed so far need further validation using observation datasets. In this study, we use the 1D + numerical model LAKE [Stepanenko et al., 2011; Stepanenko et al., 2016]. The LAKE model was applied to a small subarctic lake in the Seida study site (Komi Republic, Russia) for identification of the key factors influencing the surface CH<sub>4</sub> flux and its concentration in the lake. We carried out a calibration of biogeochemical constants involving qualitative considerations of the character of biogeochemical and physical processes occurring in the lake and aiming at a satisfactory agreement with observations, performed by the University of Eastern Finland (UEF) [Lind et al., 2009; Marushchak et al., 2016]. Comparing our model calibration results to earlier studies suggest that the crucial parameter of the model – methane production rate constant ( $P_{new, 0}$ ) – has similar values for lakes of different types in high latitudes.

**KEY WORDS:** methane, methane production rate, methane oxidation, lakes, numerical simulation.

## INTRODUCTION

Since the second half of the 20<sup>th</sup> century, atmospheric methane concentration has increased by about 1.5 times [IPCC, 2013]. According to the International Panel on Climate Change (IPCC), methane is the second greenhouse gas in its contribution to modern global warming. Though lakes occupy only 1.8–4 % of a terrestrial surface [Downing et al., 2006; Doganovsky, 2012; Choulga et al., 2014], methane emissions from lakes (by different estimates) contribute to about 6–16 % of the total flux of natural biogenic sources [Tranvik et al., 2009; Bastviken et al., 2011; Ortiz-Llorente & Alvarez-Cobelas, 2012]. Therefore, it is important to consider the contribution of lakes to the regional estimates of methane fluxes, as well as to develop models of methane emissions from lakes, in particular, for their subsequent incorporation into climate models.

Currently, about four known mathematical models of methane emission from lakes have been developed: one comparatively simple model describing a majority of the chemical processes of methane and carbon dioxide emission from lakes, but with schematic representation of physical processes [Makhov & Bazhin, 1999; Bazhin, 2001] and more complex, sophisticated models [Stepanenko et al., 2011; Stepanenko et al., 2016], [Kessler et al., 2012], [Tan et al., 2015a; Tan et al., 2015b]. Model of [Kessler et al., 2012] is a 3-D numerical model of permafrost dynamics below lakes with a simulation of methane release, taking into consideration topography of the landscape. The model by [Tan et al., 2015a; Tan et al., 2015b] is 1-D and contains a mathematical description of dynamics of the following gases: CO<sub>2</sub>, CH<sub>4</sub>, O<sub>2</sub>, and N<sub>2</sub>. Meanwhile, the sources and sinks of carbon dioxide and oxygen do not include such important processes as photosynthesis and respiration in the water column. All of them were calibrated on lakes commonly located in Siberia and Alaska. Additionally, observation datasets collected on Lake Kuivajärvi, Finland, have also been used with the model simulations [Stepanenko et al., 2016].

In our study we use the lake model LAKE [Stepanenko et al., 2011; Stepanenko et al., 2016]. The LAKE model includes key physical and biogeochemical processes occurring in a lake that control methane fluxes to the atmosphere. In previous studies [Stepanenko et al., 2011; Stepanenko et al., 2016] the model was verified using observation data collected on two lakes: Shuchi Lake (North Eastern Siberia, Russia) and Lake Kuivajärvi (Finland). In both cases, LAKE demonstrated good agreement with the observations in thermal state and vertical transfer of greenhouse gases.

Here, we applied this model to a small subarctic lake in the Seida study site, located in the North European Russia. The objective was to obtain a detailed understanding of the processes regulating the CH<sub>4</sub> budget of this lake, such as an aerobic methane oxidation and a sedimentary oxygen demand, etc. We made a calibration of constants keeping in mind physical considerations and basing on existent empirical data on these constants from literature. The aim was to achieve satisfactory agreement with observations. Calibration of constants is inevitable in biogeochemical modules as the mathematical formulation there so far cannot be derived from first principles.

## MATERIALS AND METHODS

### *Area of study*

This study was conducted for a lake in a tundra area in North East European Russia (67°03'N, 62°56'E, 103 m a.s.l.) in Seida study site, Komi Republic. The climate of this region is subarctic with both Arctic and Atlantic influence. In general, it can be characterized as severe, with a short and cool summer and a long frosty winter. The climate is formed under the conditions of limited solar radiation (the net radiation is about 700 MJm<sup>-2</sup>yr<sup>-1</sup>), under the influence of the Atlantic seas and the intensive westerlies. An advection of a marine warm air, occurring with the Atlantic cyclones, passing through the area of study,

and the frequent outbreaks of the cold Arctic air from the Arctic Ocean leads to instability in weather conditions throughout the year. The area of study belongs to the discontinuous permafrost zone. Here, the effects of climate warming are assumed to be significant since it is close to the transition zone between taiga and tundra and also because the long-term mean temperature of permafrost is close to zero [Lind, 2009].

According to the long-term weather data from the nearest meteorological station, Vorkuta (67°48'N, 64°01' E, 172 m a.s.l.; 70 km from the study site), the mean annual air temperature in the region is  $-5.6\text{ }^{\circ}\text{C}$  and the total precipitation is 501 mm (long-term averages for 1977–2006; Komi Republican Center for Hydrometeorological and Environmental Monitoring). The duration of the growing season is approximately 80 days. In long-term, the warmest month of the growing season is July ( $+13.0\text{ }^{\circ}\text{C}$ ). The landscape consists of shrub tundra, peat plateau complex with thermokarst lakes and wet fens located along to the border of peat plateau.

According to [Lind, 2009] the fen and thermokarst lakes are important sources for atmospheric  $\text{CH}_4$ . It is found in the study that lakes produce high  $\text{CH}_4$  emissions ( $48\text{--}112\text{ mg CH}_4\text{m}^{-2}\text{ d}^{-1}$ ).

The object of our study is one of the three thermokarst lakes studied by [Lind et al., 2009] – a small, shallow lake with an area of approximately 0.9 ha (Fig. 1). The depth in the lake varies from 1.1 to 2.6 m. It has intermediate surface area and depth compared to the two other lakes located nearby and studied by UEF as well. Our choice of the lake was determined by that the measurement data from this lake were most apt to be compared to our lake model results: gas collectors were in the middle of the lake and there was small vegetation coverage. The lake model being used is 1D and does not explicitly take into account gas transfer by vegetation. The subsurface gas collectors in one of these two lakes were deployed near the shore with high peat walls and other lake had too small open water area because of the vegetation [Lind, 2009].

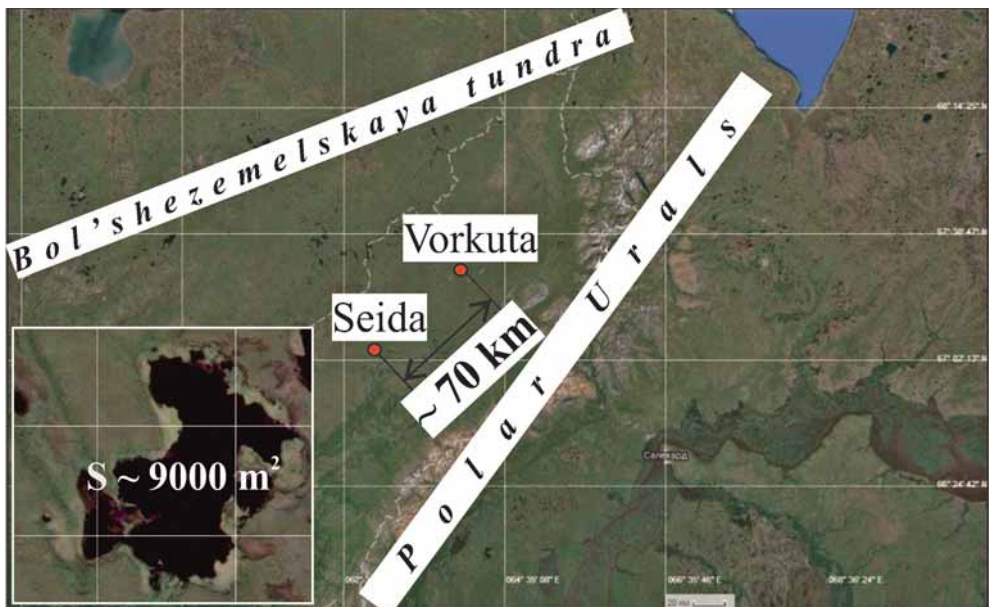


Fig. 1. Location of a lake under study.

### Observation Data and Measurement Site

All observation data used in this research were provided by University of Eastern Finland, Kuopio [Lind et al., 2009; Marushchak et al., 2016]. These data consist of two types of measurements: lake surface water samplings and atmospheric measurements. The lake surface water measurements were conducted between mid-July and the end of August 2007. Among the main observed characteristics were: water temperature at the depth of 10 cm ( $T_{10cm}$ ), surface-water methane and carbon dioxide concentration ( $X_{CH_4}$ ,  $S_{CO_2}$ ), diffusion flux ( $F_d$ ) and ebullition flux ( $F_e$ ) of methane to the atmosphere (Table 1). The last two variables represent two methane emission pathways to the atmosphere: *diffusion* from the surface water to the atmosphere following Fick's law and *ebullition* occurring due to methane concentration reaching a critical value in the lake sediments. The water samples were taken in the middle of the lake at the same times as the floating chamber measurements were done.

The site weather data form the driving variables for a top boundary condition of the model. Meteorological variables were recorded in the Seida study site using a Hobo Micro Station H21 data logger (Onset, USA), including: wind speed (S-WSA-M003, Onset, USA), air temperature, relative

humidity (S-THAM0006, Onset, USA), air pressure (S-BPA-CM10, Onset, USA), precipitation (7852 Rain Collector, Davis, USA), photosynthetically active radiation (PAR; S-LIA-M003, Onset, USA), cloudiness [Lind et al., 2009; Marushchak et al., 2016].

### Overview of the LAKE model

The numerical model LAKE [Stepanenko et al., 2011; Stepanenko et al., 2016] is a model of a closed water body with biogeochemical module for simulation of processes causing the development of lake's methane, oxygen and carbon dioxide vertical concentration profiles. It is a one-dimensional, hydrothermodynamic model. One of the important improvements recently introduced in the model are the multiple soil columns (Fig. 2) that allow to

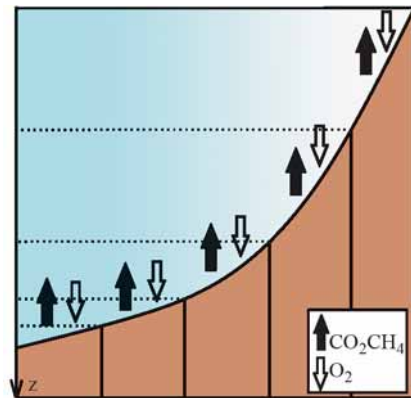


Fig. 2. A scheme of the soil columns in LAKE model.

Table 1. Observation data on Seida lakes [Lind et al., 2009; Marushchak et al., 2016]

Observed value	Dates of measurements	Total number of measurements (1 time per day)	Measurement techniques
$T_{10cm}$	22.07.2007, 28.07.2007, 04.08.2007, 11.08.2007, 18.08.2007, 27.08.2007	6 times	Tenma 72-2060 with temperature probe (Fluke 80PK-22)
$C_{CH_4}$ , $C_{CO_2}$ ( $mol \cdot m^{-3}$ )	22.07.2007, 28.07.2007, 04.08.2007, 11.08.2007, 18.08.2007, 27.08.2007	6 times	head-space method [Huttunen et al., 2002]
$F_d$ ( $mg CH_4 \cdot m^{-2} \cdot d^{-1}$ )	04.08.2007, 11.08.2007, 18.08.2007, 27.08.2007	4 times (between 9:00–20:00)	floating chamber [Lambert & Fréchet, 2005]
mean $F_e$ ( $mg CH_4 \cdot m^{-2} \cdot d^{-1}$ )	15.7.2007, 22.7.2007, 24.7.2007, 28.7.2007, 4.8.2007, 11.8.2007, 18.8.2007, 27.8.2007	8 times	subsurface bubble gas collectors [Huttunen et al., 2001]

account for bottom temperature and gas fluxes horizontal distribution, according to the morphometry of a lake (justifying to add "+" into the dimensionality abbreviation of the model, i.e. "1D + model").

A structure of the LAKE model in essence consists of three numerical modules jointly solving a coupled system of one-dimensional equations for different environments (snow, ice, water, soil):

1. hydrothermodynamic module – describes thermal and hydrodynamic processes in a water body;
2. thermodynamic module – includes thermodynamics of snow, ice at the top of lake and soil at the bottom;
3. biogeochemical module – includes biogeochemical processes, acting on concentrations of methane, carbon dioxide and oxygen in the lake, all linked to each other.

*Hydrothermodynamics of the model.* The basic equation for the first numerical component of the LAKE model is a 1D heat transfer equation including vertical turbulent heat transport, solar radiation absorption and heat exchange to sediments being the main terms:

$$c_w \rho_w \frac{\partial \bar{T}}{\partial t} = -\frac{1}{A} \int_{\Gamma^{A(\xi)}} T(u_n \cdot n) dl + \frac{1}{Ah^2} \frac{\partial}{\partial \xi} \left( Ak_T \frac{\partial \bar{T}}{\partial \xi} \right) - \frac{1}{Ah} \frac{\partial A \bar{S}}{\partial \xi} + \frac{1}{Ah} \frac{\partial A}{\partial \xi} [S_b(\xi) + F_{iz,b}(\xi)] + M(\xi, t) \frac{\partial \bar{T}}{\partial \xi}, \quad (1)$$

where  $\bar{T}$  – horizontally averaged water temperature,  $t$  – time;  $h$  – the maximum depth of a water body;  $\xi = z/h$ ;  $z$  is a vertical coordinate, directed along the gravity force (downward) and measured from the surface of a water body;  $k_T$  – the thermal diffusivity coefficient equal to the sum of molecular and turbulent diffusivities;  $S_b(\xi)$  –

shortwave radiation flux;  $F_{iz,b}(\xi)$  – soil heat flux at the level  $z$ ;  $c_w$  is the heat capacity of water;  $\rho_w$  – the mean density of fresh water;

$M(\xi, t) = \left( \frac{\xi}{h} \frac{dh}{dt} = \frac{B_s}{h} \right)$  being a metric term arising from usage of the normalized vertical coordinate;  $B_s$  – the water budget at the water–air interface (precipitation minus evaporation,  $r-E$ ).

*Gas transfer in the water column.* Biogeochemistry module of LAKE considers the dynamics of three gases in the water column: *methane, carbon dioxide and oxygen*. Their concentrations in the water column follow, respectively  $(C_{CH_4}, C_{CO_2}, C_{O_2})$ , the following governing equations:

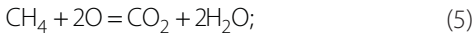
$$\frac{\partial C_{CH_4}}{\partial t} = M(\xi, t) \frac{\partial C_{CH_4}}{\partial \xi} + \frac{1}{Ah^2} \frac{\partial}{\partial \xi} \times \left( Ak_s \frac{\partial C_{CH_4}}{\partial \xi} \right) + B_{CH_4} - O_{CH_4}; \quad (2)$$

$$\frac{\partial C_{CO_2}}{\partial t} = M(\xi, t) \frac{\partial C_{CO_2}}{\partial \xi} + \frac{1}{Ah^2} \frac{\partial}{\partial \xi} \times \left( Ak_s \frac{\partial C_{CO_2}}{\partial \xi} \right) + B_{CO_2} - P_{CO_2} + R_{CO_2} + D_{CO_2} + S_{CO_2} + O_{CO_2}; \quad (3)$$

$$\frac{\partial C_{O_2}}{\partial t} = M(\xi, t) \frac{\partial C_{O_2}}{\partial \xi} + \frac{1}{Ah^2} \frac{\partial}{\partial \xi} \times \left( Ak_s \frac{\partial C_{O_2}}{\partial \xi} \right) + B_{O_2} - P_{O_2} + R_{O_2} + D_{O_2} + S_{O_2} + O_{O_2}, \quad (4)$$

where the right-hand sides of these equations represent diffusion (assuming  $k_s$  to be the same eddy diffusivity for all species), sources and sinks due to the following processes: dissolution/exsolution of gases at the bubble-water interface  $(B_{CH_4}, B_{CO_2}, B_{O_2})$ , photosynthesis  $(P_{CO_2}, P_{O_2})$ , respiration  $(R_{CO_2}, R_{O_2})$ , biogeochemical oxygen demand in the water column  $(D_{CO_2}, D_{O_2})$ , sedimentary oxygen demand – SOD  $(S_{CO_2}, S_{O_2})$ , methane aerobic oxidation in the water column  $(O_{CH_4}, O_{CO_2}, O_{O_2})$ .

*Methane aerobic oxidation.* The stoichiometry of methane oxidation (MO) in water and soil follows equation (Eq. 5) below and its rate is described by Michaelis-Menten kinetics (Eq. 6).



$$O_{\text{CH}_4} = V_{\text{max}} \frac{C_{\text{CH}_4}}{K_{hs,\text{CH}_4}} \frac{C_{\text{O}_2}}{K_{hs,\text{O}_2} + C_{\text{O}_2}}. \quad (6)$$

In the last expression there are three parameters: a potential MO rate  $V_{\text{max}}$  and half-saturation constants  $K_{hs,\text{O}_2}$  and  $K_{hs,\text{CH}_4}$ , that influence the rate of oxidation.

*Sedimentary oxygen demand (SOD).* The sedimentary oxygen demand appears as a sink in Eq. 4 ( $S_{\text{O}_2}$ ) representing a downward  $\text{O}_2$  flux at the lake's margins, and at the same time it is one of the main sources of  $\text{CO}_2$  in Eq. 3. In the LAKE model, this process follows the simplified stoichiometry (Eq. 7 below) [Stefan & Fang, 1994] and kinetics [Walker & Snodgrass, 1986] (Eq. 8 below). The Walker and Snodgrass formulation contains a Michaelis-Menten kinetics term (similar to Eq. 6) that is called a "biological" part, and the other term is the "chemical" part, quantified as a diffusion at the bottom of a lake.



$$S_{\text{O}_2} = \mu \frac{C_{\text{O}_2}}{J_{hs,\text{O}_2} + C_{\text{O}_2}} + k_{c_0} C_{\text{O}_2}, \quad (8)$$

where  $k_{c_0}$  is the oxygen diffusion constant, that is in general case dependent on the turbulent mixing in near-bottom flows in a lake, temperature and soil porosity.

*Methane production.* The methane production, transport and sink in the bottom sediments beneath a lake are included in the following equation for methane concentration,  $C_{\text{CH}_4,\text{sed}}$  [Walter & Heimann, 2000]:

$$\frac{\partial C_{\text{CH}_4,\text{sed}}}{\partial t} = \frac{\partial}{\partial z} k_{\text{CH}_4,m} \frac{\partial C_{\text{CH}_4,\text{sed}}}{\partial z} + P - E - F. \quad (9)$$

This mathematical expression represents the three main methane transport mechanisms: diffusion of dissolved methane  $\left( \frac{\partial}{\partial z} C_{\text{CH}_4,m} \frac{\partial C_{\text{CH}_4,\text{sed}}}{\partial z} \right)$ , its ebullition ( $E$ ), and the transport by plants ( $F$ ). The main source of methane is the methane production ( $P$ ) from anaerobic decomposition of organic matter.

In the current study we don't take into account methane transport by plants, as this flux has not been measured on the lake, and up-to-date approaches to simulate this pathway are not comprehensive and need specific validation. Also, plants grow only on the shoreline of the lake, which accounts for a relatively small area. The anaerobic decomposition of organics occurs due to the *Archaea* activity in the bottom sediments and is represented in the LAKE model by two types-production from old ( $P_{\text{old}}$ ) [Stepanenko et al., 2011] and new ( $P_{\text{new}}$ ) [Walter & Heimann, 2000] organic material:

$$P = P_{\text{new}} + P_{\text{old}} \quad (10)$$

In the current study we neglect the second term in (Eq. 10), as there is no exact information, whether permafrost exists below the lake, and  $P_{\text{old}}$  is significant in vicinity of the talik-permafrost boundary only. The methane production due to the new organics decomposition is calculated as follows:

$$P_{\text{new}} P_{\text{new},0} e^{-\alpha_{\text{new}} z} q_0^{T/T_0} H(T), \quad (11)$$

where  $P_{\text{new},0}$  is a calibration parameter,  $\alpha_{\text{new}}$  is the parameter that determines the rate of decrease in methane generation with depth of sediments,  $3 \text{ m}^{-1}$ ,  $z_s$  is the depth measured from the lake bottom,  $q_0$  and  $T_0$  are constants equal to 2 units [Stepanenko et al., 2011] and  $10^\circ \text{C}$  [Walter & Heimann, 2000], respectively,  $H(T)$  is the Heaviside function, temperature,  $T$ , is given in degrees Celsius.

As a result, in general, the process of methane production depends on the temperature, the depth and the calibration constant  $-P_{\text{new},0}$ .

Table 2. Main parameters of a baseline experiment

Parameter	Value	Parameter	Value
Lake's area, m <sup>2</sup>	9000	Coefficient of solar radiation extinction in the water body, m <sup>-1</sup>	5
Maximum depth, m	2.6	Constant of temperature dependence $q_0$ of methane production (Eq. 11)	2
Number of computational layers in water	10	Methane production rate constant $P_{new,0}$ , mol · m <sup>-3</sup> · s <sup>-1</sup> (Eq. 11)	4.0 · 10 <sup>-8</sup>
Number of computational layers in soil	10	Half-saturation constant $K_{hs,CH_4}$ , mol · m <sup>-3</sup> (Eq. 6)	6.88 · 10 <sup>-3</sup>
Number of soil columns	5	Half-saturation constant $K_{hs,O_2}$ , mol · m <sup>-3</sup> (Eq. 6)	2.1 · 10 <sup>-2</sup>
Time step of integration, s	10	Methane oxidation potential $V_{max}$ , mol · m <sup>-3</sup> · d <sup>-1</sup> (Eq. 6)	1 · 10 <sup>-1</sup>
The duration of integration, days	92	Oxygen diffusion constant $k_{O_2}$ , m · s <sup>-1</sup> (Eq. 8)	0.045
Start of simulation	01.07.2007		

*Boundary conditions.* The boundary conditions for methane at the sediments-water body interface are:

$$\frac{-k_s}{h} \frac{\partial C_{CH_4}}{\partial \xi} \Big|_{\xi=1} = -k_{s,s} \frac{\partial C_{CH_4, sed}}{\partial z_s} \Big|_{z_s=0}; \quad (12)$$

$$C_{CH_4} \Big|_{\xi=1} = \left( \frac{C_{CH_4, sed}}{\rho} \right) \Big|_{z_s=0}, \quad (13)$$

where  $\rho$  – soil porosity.

### Model experimental setup

The parameters of a baseline experiment with LAKE are demonstrated in Table 2. The values of biogeochemical constants were tuned in previous LAKE model applications to other water bodies.

The surface water temperature from baseline experiment reasonably well corresponds to the observed one, with root-mean-square deviation (RMSD) 1.38 °C [Stepanenko et al., 2014].

## RESULTS AND DISCUSSION

### Calibration of methane production rate constant in the baseline experiment

We calibrated the methane production rate constant ( $P_{new,0}$ ) for the adequate simulation

of the observed value of methane ebullition flux ( $F_e$ ). Calculated and observed ebullition flux time series are shown on the Fig. 3 (red dashed line and green points, respectively).

The values of modelled and observed ebullition fluxes agreed well with  $P_{new,0} = 4 \cdot 10^{-8}$  mol · m<sup>-3</sup> · s<sup>-1</sup> (RMSD – 20 mg CH<sub>4</sub> · m<sup>-2</sup> · d<sup>-1</sup>, mean observed  $F_e$  – 34 mg CH<sub>4</sub> · m<sup>-2</sup> · d<sup>-1</sup>, mean modeled  $F_e$  – 32 mg · CH<sub>4</sub> · m<sup>-2</sup> · d<sup>-1</sup>). Fig. 3 suggests that temporal variation of ebullition flux is likely to be significantly higher than that measured by infrequent sampling. In the model, oscillations of  $F_e$  are partially a consequence of atmospheric pressure fluctuations caused by synoptic weather variability. Modeled time series of methane ebullition flux are smoother, if atmospheric pressure is set constant (see Fig. 3, blue dashed line) (RMSD – 26 mg · CH<sub>4</sub> · m<sup>-2</sup> · d<sup>-1</sup>, mean modeled  $F_e$  – 29 mg · CH<sub>4</sub> · m<sup>-2</sup> · d<sup>-1</sup>).

It is important to note, that the  $P_{new,0}$  value obtained here is close to those estimated in the previous studies both for lakes and wetlands at high latitudes (Table 3). This suggests, that this constant can be of the same order of magnitude for all subarctic lakes, however, this hypothesis requires further validation. We regard this experiment as a baseline for comparison with other

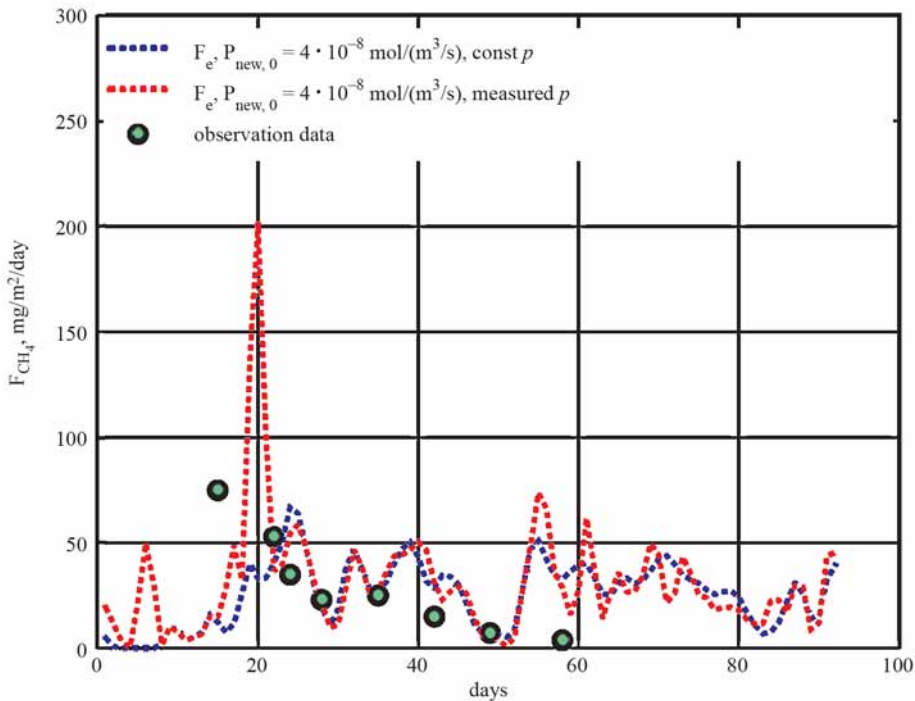


Fig. 3. Daily-averaged model ebullition flux of methane ( $F_e$ ) at the lake surface (July–September 2007); “0” on horizontal axis corresponds to 01.07.2007 (blue dashed line represent model experiment with constant atmospheric pressure,  $p$ , red dashed line – with measured  $p$ ).

Table 3. Methane production rate constant  $P_{new,0}$  in other studies

$P_{new,0}$ ( $\text{mol} \cdot \text{m}^{-3} \cdot \text{s}^{-1}$ )	Source
$3.0 \cdot 10^{-8}$	Lake Kuivajärvi, Finland [Stepanenko et al., 2016]
$2.55 \cdot 10^{-8}$	Shuchi Lake, North Eastern Siberia, Russia [Stepanenko et al., 2011]
$8.3 \cdot 10^{-8} - 1.6 \cdot 10^{-7}$	High latitude wetlands [Walter & Heimann, 2000]
$4.0 \cdot 10^{-8}$	Lake at the Seida site, current study

model runs, described in the following sections.

### Calibration of other biogeochemical constants

*Methane aerobic oxidation.* Results of a baseline experiment demonstrate order of magnitude lower values of surface methane concentration and diffusion flux at the water-air interface in comparison with the observation data (green line in Fig. 4). Therefore, calibrating the methane

production constant only is not sufficient to match observations in terms of these variables.

Once methane is produced in sediments and transported upwards by turbulence, the main process reducing the value of methane concentration and its diffusion flux at the lake surface is the methane aerobic oxidation (see Eq. 5, Eq. 6). It is likely that the model overestimated methane oxidation in the baseline run.



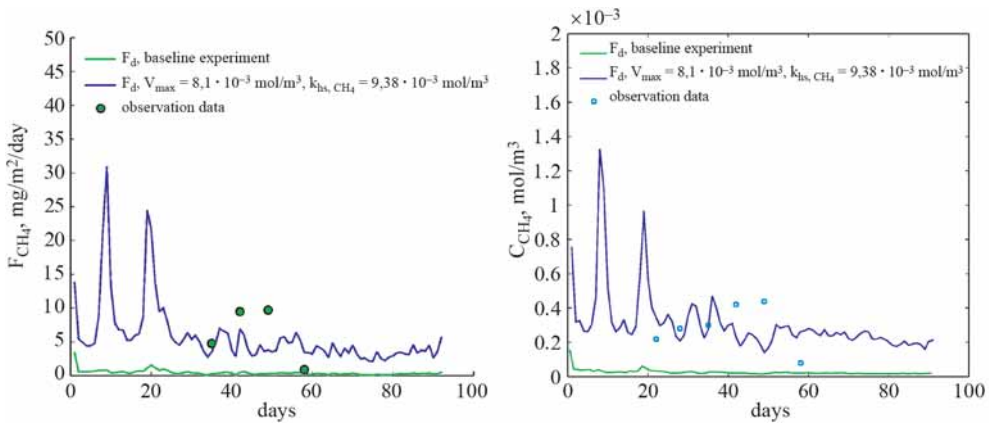


Fig. 4. (A) – daily-averaged diffusion flux of methane to the atmosphere; (B) – daily-averaged concentration of methane at the lake surface (July–September 2007); “0” on horizontal axis corresponds to 01.07.2007 (RMSD for  $F_d$  – 0.45  $mg\ CH_4 \cdot m^{-2} \cdot d^{-1}$ ; RMSD for  $C_{CH_4}$  –  $7.9 \cdot 10^{-5} mol \cdot m^{-3}$ ).

Table 4. Constants of methane aerobic oxidation reaction rate

Parameter	Value in the baseline experiment	Mean value $\pm$ SD	New calibrated value
$V_{max}$ ( $mol \cdot m^{-3} \cdot d^{-1}$ )	0,1	$1.8 \cdot 10^{-2} \pm 0.01$ [Martinez-Cruz et al., 2015]	0.0081
$K_{hs, CH_4}$ ( $mol \cdot m^{-3}$ )	$6.88 \cdot 10^{-3}$	$6.8 \cdot 10^{-2} \pm 0.003$ [Liikanen et. al., 2002; Lofton et. al., 2014] max: $4.4 \cdot 10^{-2}$ [Martinez-Cruz et al., 2015]	$9.38 \cdot 10^{-3}$

In a baseline model experiment we used the values of methane oxidation constants, MO potential rate  $V_{max}$  and half-saturation constant  $K_{hs, CH_4}$  from [Liikanen et. al., 2002], who investigated an interval of values for single lake Kevätön. The recent research [Martinez-Cruz et al., 2015] presents a wider range of values for these constants, estimated for several lakes in non-yedoma permafrost region (Table 4). Seida Lake belongs to the same type.

We calibrated MO potential rate and a half-saturation constant for methane in the range known from [Martinez-Cruz et al., 2015] so as to reduce the methane oxidation rate in water, and thus to increase the methane concentration at 1.5–2 m depth (Fig. 5) and at the air-water interface. As a result, we obtained an agreement between calculated data and measurements in typical averaged levels

of methane diffusion flux ( $F_d$ ), and surface methane concentration ( $C_{CH_4}$ ) (Fig. 4). As for the half-saturation constant for oxygen,  $K_{hs, O_2}$ , its empirical range is much narrower [Martinez-Cruz et al., 2015], and we found its variation to cause insignificant effect on surface water methane content.

*Sedimentary oxygen demand (SOD)*. Since the measurement data of carbon dioxide concentration in the mixed layer of the lake were available, it was reasonable to check how the model would represent it. The baseline experiment underestimated values of carbon dioxide concentration at the lake surface, about three times lower than in the observation data (Fig. 6). This was likely due to the erroneous calculation of sedimentary oxygen demand (or *SOD*, see Eq. 8), as this is the main source of  $CO_2$  in many lakes.

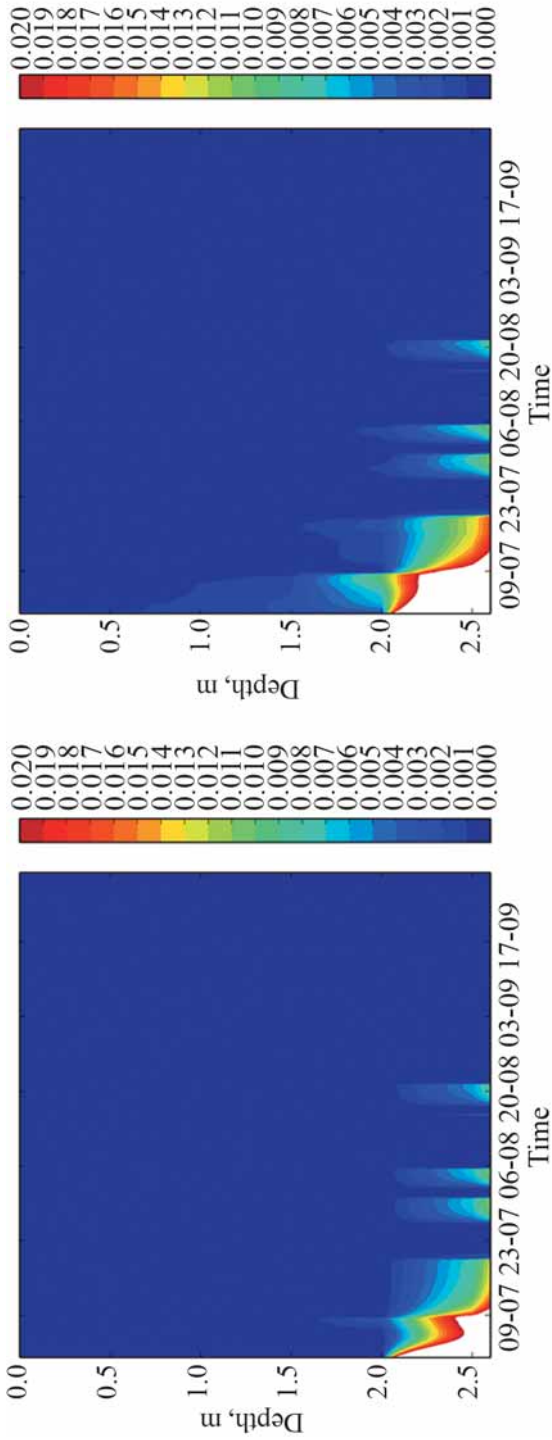


Fig. 5. The time-depth distribution of  $\text{CH}_4$  concentration ( $\text{mol}\cdot\text{m}^{-3}$ ) (July–September 2007): (A) – a baseline experiment; (B) – an experiment with calibrated  $V_{\max}$  and  $K_{fs, \text{CH}_4}$ .

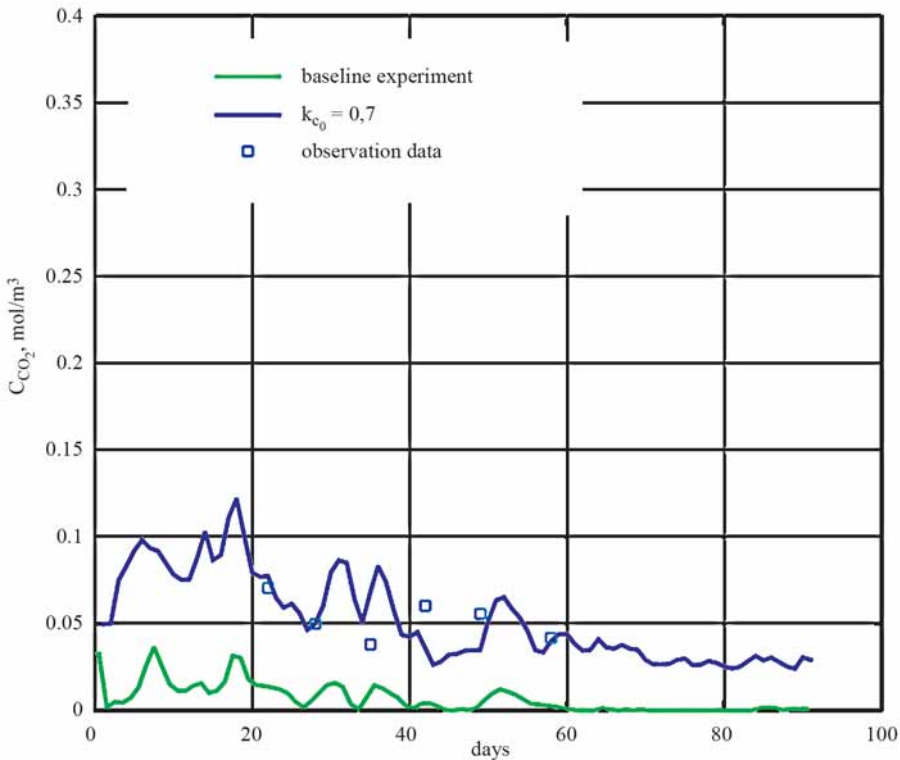


Fig. 6. Daily-averaged concentration of  $\text{CO}_2$  at the lake surface (July – September 2007); “0” on horizontal axis corresponds to 01.07.2007 (RMSD for  $C_{\text{CO}_2} - 1.6 \cdot 10^{-2} \text{ mol} \cdot \text{m}^{-3}$ ).

As mentioned above, the Seida region is characterized by strong winds, thus there should be the seiche-like bottom flows in lakes. These flows increase the turbulent mixing near the bottom thereby enhancing an oxygen diffusion into the sediments and corresponding  $\text{CO}_2$  production. The coefficient  $k_{c_0}$  in Walker and Snodgrass model is responsible for oxygen diffusion, however, to the best of our knowledge, no dependence on near-bottom turbulence has been proposed for it in the literature.

In view of this, we increased  $k_{c_0}$  from the baseline value  $k_{c_0} = 0.045 \text{ m} \cdot \text{s}^{-1}$  to  $k_{c_0} = 0.7 \text{ m} \cdot \text{s}^{-1}$ , delivering much better agreement between simulated  $\text{CO}_2$  surface concentration and that observed (Fig. 6).

This change of  $k_{c_0}$  led to higher production of carbon dioxide (according

to Eq. 7) in sediments, and its penetration into upper layers of the lake. This effect is demonstrated in Fig. 7. Physical justification of this 15-fold increase of  $k_{c_0}$  would involve more sophisticated SOD models incorporating bottom boundary layer characteristics.

## CONCLUSION

Basing on the experience gained from this simulation exercise, we conclude the following.

1. The value of methane production rate constant maybe of the same order of magnitude for all northern lakes.
2. LAKE model adequately simulates average surface water concentration of methane and carbon dioxide, diffusion and ebullition fluxes of methane into the atmosphere.

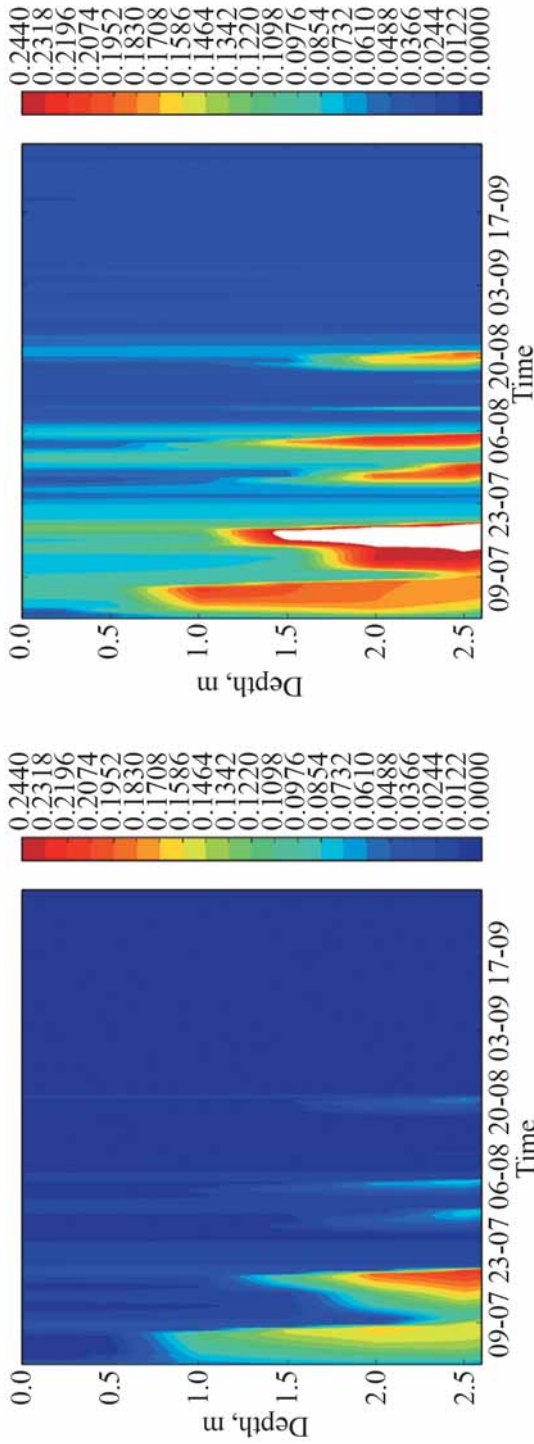


Fig. 7. The time-depth distribution of CO<sub>2</sub> concentration (mol·m<sup>-3</sup>) (July–September 2007):  
 (A) – a baseline experiment; (B) – an experiment with calibration of  $k_{c0}$  to 0.7.

3. LAKE model demonstrates significant sensitivity to variation of biogeochemical parameters controlling methane-related processes and those of oxidation of organic matter in sediments.

The model LAKE requires further calibration on the other types of lakes in the different geographical regions.

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#### REFERENCES

1. Bastviken, D., Cole, J., Pace, M., Tranvik, L. (2004) Methane emissions from lakes: dependence of lake characteristics, two regional assessments, and a global estimate. *Glob. Biogeochem. Cy.*, N 18, GB4009.
2. Bastviken, D., Tranvik, L.J., Downing, J.A., Crill, P.M., Enrich-Prast, A. (2011) Freshwater methane emissions offset the continental carbon sink. *Science*, N 331, pp. 50.
3. Bazhin, N.M. (2001) Gas Transport in a Residual Layer of a Water Basin. *Chemosphere–Global Change*, N 3, pp. 33–40.
4. Choulga, M., Kourzeneva, E., Zakharova, E., and Doganovsky, A. (2014) Estimation of the mean depth of boreal lakes for use in numerical weather prediction and climate modeling. *Tellus A*, N 66, pp. 17.
5. Doganovsky, A.M. (2012) Land surface hydrology. A general course. St. Petersburg, RGGMU, 524 p. (In Russian).
6. Downing, J.A., Prairie, Y.T., Cole, J.J., Duarte, C.M., Tranvik, L.J., Striegl, R.G., McDowell, W.H., Kortelainen, P., Caraco, N.F., Melack, J.M., Middelburg, J.J. (2006) The global abundance and size distribution of lakes, ponds, and impoundments, *Limnology and Oceanography*, N 51, pp. 2388–2397.
7. Huttunen, J.T., Lappalainen, K.M., Saarijärvi, E., Väisälä, T. and Martikainen, P.J. (2001) A novel sediment gas sampler and a subsurface gas collector used for measurement of the ebullition of methane and carbon dioxide from a eutrophied lake. *The Science of the Total Environment*, N 266, pp. 153–158.
8. Huttunen, J.T., Väisänen, T.S., Heikkinen, M., Hellsten, S., Nykänen, H., Nenonen, O., Martikainen, P.J. (2002) Exchange of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O between the atmosphere and two northern boreal ponds with catchments dominated by peatlands or forests. *Plant and Soil*, N 242, pp. 137–146.
9. IPCC, 2013: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United

Kingdom and New York, NY, USA, pp. 1535.

10. Kessler, M.A., Plug, L., Walter Anthony, K.M. (2012) Simulating the decadal to millennial scale dynamics of morphology and sequestered carbon mobilization of two thermokarst lakes in N.W. Alaska, *J. Geophys. Res.*, pp. 117.
11. Lambert, M., Fréchet, J.-L. (2005) Analytic techniques for measuring fluxes of CO<sub>2</sub> and CH<sub>4</sub> from hydroelectric reservoirs and natural water bodies. In: *Greenhouse Gas Emissions – Fluxes and Processes: Hydroelectric Reservoirs and Natural Environments.* (eds. Trembley A., Varfalvy L., Roehm C. and Garneau M.). Springer Press, pp. 37–60.
12. Lind, S. (2009), *Methane fluxes during a snow-free season in Russian tundra*, Master's Thesis, Environmental Science, University of Kuopio, Department of Environmental Science, pp. 56.
13. Liikanen, A., Huttunen, J.T., Valli, K., Martikainen, P.J. (2002) Methane cycling in the sediment and water column of mid-boreal hyper-eutrophic Lake Kevaton, Finland. *Arch. Hydrobiol.*, N 154, pp. 585–603.
14. Lofton, D.D., Whalen, S.C., and Hershey, A.E. (2014) Effect of temperature on methane dynamics and evaluation of methane oxidation kinetics in shallow Arctic Alaskan lakes, *Hydrobiologia*, pp. 209–222.
15. Makhov, G.A., Bazhin, N.M. (1999) Methane emission from lakes. *Chemosphere*, N 38, pp. 1453–1459.
16. Martinez-Cruz, K., Sepulveda-Jauregui, A., Anthony, K.W., Thalasso, F. (2015) Geographic and seasonal variation of dissolved methane and aerobic methane oxidation in Alaskan lakes. *Biogeosciences*, N 12, pp. 4213–4243.
17. Marushchak, M.E., Friborg, T., Biasi, C., Herbst, M., Johansson, T., Kiepe, I., Liimatainen, M., Lind, S.E., Martikainen, P.J., Virtanen, T., Soegaard, H., and Shurpali, N.J. (2016) Methane dynamics in the subarctic tundra: combining stable isotope analyses, plot- and ecosystem-scale flux measurements. *Biogeosciences*, N 13, pp. 597–608, doi:10.5194/bg-13-597-2016.
18. Ortiz-Llorente, M.J., Alvarez-Cobelas, M. (2012) Comparison of biogenic methane emissions from unmanaged estuaries, lakes, oceans, rivers and wetlands. *Atmospheric Environment*, N 59, pp. 328–337.
19. Stefan, H.G., Fang, X. (1994) Dissolved oxygen model for regional lake analysis, *Ecological Modelling*, N 71, pp. 37–68.
20. Stepanenko, V.M., Glazunov, A.V., Guseva, S.P., Lykosov, V.N., Shurpali, N., Biasi, C., Martikainen, P. (2014) Numerical simulation of greenhouse gases transport in a system "lake – atmospheric boundary layer", in *International conference and early career scientists*

- school on environmental observations, modeling and information systems (ENVIROMIS), SCERT, Tomsk, pp. 56–57.
21. Stepanenko, V., Machul'skaya, E.E., Glagolev, M.V., Lykossov, V.N. (2011) Numerical modeling of methane emissions from lakes in the permafrost zone. *Izvestiya, Atmospheric and Oceanic Physics*, N 2, pp. 252–264.
  22. Stepanenko, V., Mammarella, I., Ojala, A., Miettinen, H., Lykosov, V., Vesala, T., Glazunov, A., (in review, 2016) A model for methane, carbon dioxide and oxygen dynamics in a fresh-water lake. Accepted to *Geosci. Model Dev.*, doi:10.5194/gmd-2015-261.
  23. Tan, Z., Zhuang, Q., and Anthony, K.W. (2015) Modeling methane emissions from arctic lakes: Model development and site-level study, *J. Adv. Model. Earth Syst.*, N 7, pp. 459–483.
  24. Tan, Z., Zhuang, Q. (2015) Methane emissions from pan-Arctic lakes during the 21st century: An analysis with process-based models of lake evolution and biogeochemistry, *J. Geophys. Res. Biogeosci.*, N 120.
  25. Tranvik, L.J., Downing, J.A., Cotner, J.B., and Loiselle, S.A. (2009) Lakes and reservoirs as regulators of carbon cycling and climate. *Limnol. Oceanogr.*, N 6, pp. 2298–2314.
  26. Walker, R.R., Snodgrass, W.J. (1986) Model for sediment oxygen demand in lakes. *Journal of Environmental Engineering*, N 112 (1), pp. 25–43.
  27. Walter, B.P. and Heimann, M. (2000) A process-based, climate-sensitive model to derive methane emissions from natural wetlands: Application to five wetland sites, sensitivity to model parameters, and climate, *Global Biogeochem. Cy.*, N 14, pp. 745–765.
  28. Wik, M., Varner, Ruth K., Anthony, K.W., MacIntyre, S., Bastviken, D., Climate-sensitive northern lakes and ponds are critical components of methane release [online]. *Nature Geoscience*, advance online publication available from URL: <http://dx.doi.org/10.1038/ngeo2578> [Jan, 2016].

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