Effect of dissolved oxygen on methane production from bottom sediment in a eutrophic stratified lake

Maruya, Yasuyuki
Graduate School of Engineering, Kyushu University

Nakayama, Keisuke
Graduate School of Engineering, Kobe University

Sasaki, Masafumi
Department of Engineering, Kitami Institute of Technology

Komai, Katsuaki
Department of Engineering, Kitami Institute of Technology

出版情報：Journal of Environmental Sciences. 125, pp.61-72, 2023-03. Elsevier
バージョン：
権利関係：

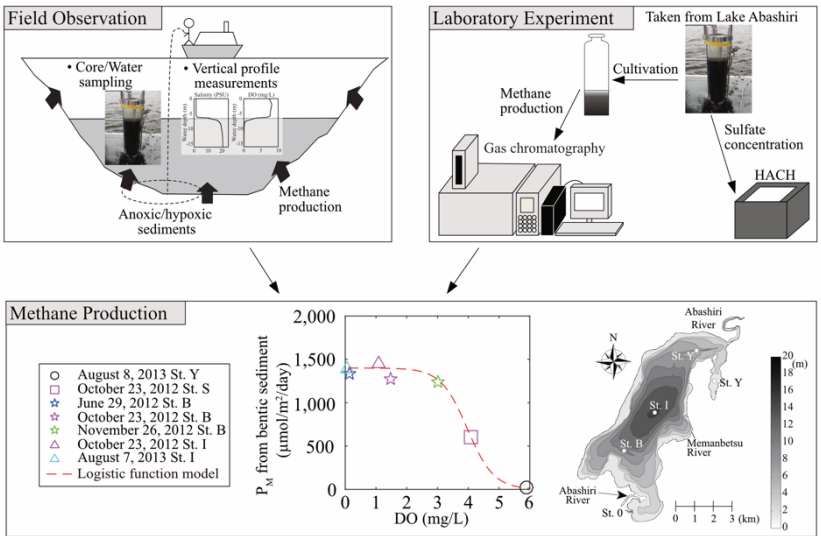
Effect of dissolved oxygen on methane production from bottom sediment in a eutrophic stratified lake

Yasuyuki Maruya[1,*], Keisuke Nakayama[2], Masafumi Sasaki[3], Katsuaki Komai[3]

1. Graduate School of Engineering, Kyushu University, 744 Motooka, Nishi-Ku, Fukuoka 819-0395, Japan
2. Graduate School of Engineering, Kobe University, Rokkodai-Cho 1-1, Nada-Ku, Kobe 657-8501, Japan
3. Department of Engineering, Kitami Institute of Technology, Kohen-Cho 165, Kitami 090-8507, Japan

*Corresponding Author: Yasuyuki Maruya, Graduate School of Engineering, Kyushu University, 744 Motooka, Nishi-Ku, Fukuoka 819-0395, Japan. E-mail: ysyk.mry@gmail.com

Received October 26, 2021
Revised January 11, 2022
Accepted


Field Observation
• Core/Water sampling
• Vertical profile measurements
Anoxic/hypoxic sediments
Methane production
Laboratory Experiment
Taken from Lake Abashiri
Cultivation
Methane production
Gas chromatography
Sulfate concentration
HACH
Methane Production
August 8, 2013 St. Y
October 23, 2012 St. S
June 29, 2012 St. B
October 23, 2012 St. B
November 26, 2012 St. B
October 23, 2012 St. I
August 7, 2013 St. I
Logistic function model
P_M from bentic sediment (μmol/m²/day)
DO (mg/L)
Abashiri River
St. Y
St. I
St. B
St. 0
Memanbetsu River
(m)
(km)

21

Abstract: Clarifying the role of sulfate and dissolved oxygen (DO) in methane production may allow for precise and accurate modeling of methane emissions in eutrophic lakes. We conducted field observations of sulfate, methane, and DO concentrations in Lake Abashiri, a typical brackish and eutrophic lake in a cold region, to develop a DO-based method for quantitively estimating methane production in a eutrophic lake and analyzed the results. We found that sulfate concentrations decreased rapidly from 900.0 mg/L in water overlying the sediments to nearly 0.0 mg/L in the bottom sediment. Methane production was almost uniform across sediment depths of 0.05 to 0.25 m, ranging from 1,400 to 1,800 μmol/m^2/day. Also, methane production was found to be a function of DO concentrations in water overlying the bottom and could be modeled by a logistic function: constant production at 1,400 μmol/m^2/day for DO concentrations of 0.0 to 3.0 mg/L, rapidly decreasing to 0 μmol/m^2/day for DO concentrations of 3.0 to 6.0 mg/L. This methane model was verified using a simple one-dimensional numerical model that showed good agreement with field observations. Our results thus suggest that the proposed methane model reduces uncertainty in estimating methane production in a eutrophic lake.

Comment E-2

Comment E-2

Keywords:
Dissolved Oxygen (DO)
Stratification
Methane
Anoxia
Numerical simulation

---

* Corresponding author. E-mail: ysyk.mry@gmail.com (Yasuyuki Maruya)

Introduction

The Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report (AR5) has stated that global sea levels and air temperatures have increased and will continue to increase due to climate change (IPCC, 2013). The incidence of natural disasters such as extensive flooding appears to be increasing, which may be due to changes in precipitation patterns due to climate change (e.g., Komori et al., 2012).

Greenhouse gases, such as carbon dioxide (CO_2) and methane (CH_4), play an essential role in climate change. The lifetime of CO_2 is 5–200 years, whereas the lifetime of CH_4 is shorter, about 10 years, as CH_4 is likely to react with labile molecules and OH radicals. However, since the greenhouse effect of CH_4 is about 25 times greater than that of CO_2, the increase in CH_4 may enhance global warming to a greater extent (IPCC, 2007).

The IPCC (2007) estimated that global CH_4 emissions amount to 582 Tg CH_4/year, of which natural CH_4 emissions from wetlands, tidal flats, and lakes account for 110–240 Tg CH_4/year (IPCC, 2007; Reeburgh, 2003; Bastviken et al., 2011). CH_4 emissions from lakes have been estimated to be 8–48 Tg CH_4/year (Bastviken et al., 2004). Several studies have also investigated CH_4 ebullition and diffusive CH_4 emissions from lakes in cold regions (Walter et al., 2006; Sasaki et al., 2009; Sasaki et al., 2010). Collectively, these studies suggest that current and future increases in CH_4 emissions will intensify climate change. Therefore, it is necessary to clarify the specific processes related to CH_4 production and emissions from lakes to predict changes in the global environment.

Comment E-2

Among the processes contributing to CH_4 emission from lakes, i.e., ebullition, diffusive emission, water column storage, and plant-mediated emission, ebullition accounts for 50%–60% of total emissions, and diffusive emission and water column storage together account for 40%–50% of total emissions (Bastviken et al., 2004). Bastviken et al. (2008) conducted field observations in three different lakes, and confirmed that ebullition accounted for the largest portion of emissions (52%–74%) with a significant contribution from diffusive emissions (26%–48%). CH_4 concentrations increase in the bottom layer during summer stratification because CH_4 is produced in hypoxic layers by methanogenic archaea (Cicerone and Oremland, 1988; Kiene, 1991; Bastviken et al., 2002). The release of diffusive CH_4 flux is significantly greater during autumn and spring than in winter or summer, since overturn collapses stratification (Rudd and Hamilton, 1978; Striegl and Michmerhuizen, 1998; Schubert et al., 2012; Fernández et al., 2014).

CH_4 is produced in sediments under anaerobic conditions due to methanogenic archaea decomposing organic matter. The terrestrial water inflow and oceanic water intrusion in brackish lakes connected to the ocean enhance a two-layer stratification system. Therefore, the exchange time between the upper and lower layers tends to be longer than in freshwater lakes, leading to enhanced organic matter accumulation and CH_4 production from the sediment. CH_4 emissions from freshwater lakes are different from those of brackish lakes, even if the lake sizes are the same, because CH_4 production is associated with water quality parameters such as DO (Roulet et al., 1997). Additionally, Capone and Kiene (1988) and Minami et al. (2012) showed

that CH_4 is produced in sediments under anaerobic conditions where sulfate-reducing bacteria (SRB) are less active. Hantush (2007) and Di Tero et al. (1990) developed a CH_4 and ammonia oxidation model in bottom sediment based on sediment oxygen demand, which affects the balance of oxygen levels in rivers, estuaries, and lakes. In their model, CH_4 production in the anaerobic bottom layer was estimated based on the following chemical formula:

$$2CH_2O \rightarrow CO_2 + CH_4 \quad (1)$$

CH_4 production is expected to be associated with DO and sulfate concentrations in sediments because it is produced in sediments where SRB is less active. However, Eq. (1) can be applied only to the freshwater environment, because DO and sulfate concentrations affect CH_4 production in brackish and eutrophic lakes, where sulfate-reducing bacteria are active. Few reports are available on CH_4 production in brackish and eutrophic lakes, although CH_4 production in freshwater lakes has been investigated in previous studies (e.g., Martinez and Anderson, 2013; Yan et al., 2019). Moreover, few attempts have been made to quantify CH_4 production from bottom sediment due to "diffusion" using the DO concentration in water overlying "brackish" and eutrophic lake sediment under strong stratification.

Stratification enhances the occurrence of hypoxia in the lower layer of a lake, resulting in the release of large amounts of nutrients from the bottom sediment, which in turn causes eutrophication. As a result, the eutrophic lakes release significantly more diffusive emissions than oligotrophic lakes (e.g., Utsumi et al., 1998; Takii et al., 1997; Taoka et al., 2020; Bastviken et al., 2008). Therefore, we hypothesize that a hypoxic condition enhances methane production from the bottom sediment, but not an oxidized condition. Methane emission from stratified lakes has been investigated previously, as mentioned above. However, the relationship between the methane production from the bottom sediment and the DO concentration remains unclear. Thus, establishing a method for estimating CH_4 production based on the DO concentration would contribute to the prediction of CH_4 emissions in stratified lakes. Therefore, in this study, we sought to develop a method for quantitively estimating CH_4 production under anaerobic and aerobic conditions based on changes in the DO concentration in a eutrophic lake.

1 Materials and methods

1.1 Study site and water quality field observations

Freshwater input to Lake Abashiri from upstream reaches is underlain by seawater intrusion through the seaward end of Abashiri River, resulting in a thin pycnocline. Sasaki and Endo (2014) demonstrated that the methane emission from lakes on the island of Hokkaido (excluding Lake Abashiri) into the atmosphere is almost the same as the methane emission from

123 lakes in Sweden and North America (Bastviken et al., 2004) (**Fig. 1a** and **Fig. 2a**) (see section
124 1.3 for details). Although the surface area of Lake Abashiri accounts for only 4% of the total
125 surface areas of the lakes on the island of Hokkaido, the methane emission to the atmosphere
126 from Lake Abashiri is 11% of the total methane emission from Hokkaido. In addition, the DM
127 concentration of Lake Abashiri is five times as large as that estimated by Eq. (3) (Sasaki and
128 Endo, 2014). Excluding Lake Abashiri, the TN concentrations of the lakes on Hokkaido are
129 0.08–0.52 mg/L, but the TN concentration of Lake Abashiri is about 1.4 mg/L. Therefore, we
130 chose Lake Abashiri, a typical cold-region eutrophic lake connected to the Okhotsk Sea through
131 the Abashiri River (river length 7.2 km), as the target lake in this study (**Fig. 1b**).

Comment E-2

132 We conducted field observations on June 29, 2012, October 23, 2012, November 26, 2012,
133 August 7, 2013, and August 8, 2013 to investigate the vertical profiles of salinity and DO in
134 Lake Abashiri using a water quality profiler (AAQ1183; JFE Advantech, Japan). Salinity and
135 DO were measured at vertical intervals of 0.1 m. Because DO from the saturated upper layer
136 cannot cross the pycnocline, the lower layer becomes anoxic due to the consumption of DO by
137 the bottom sediment. In addition, snowmelt flooding during the spring and regular flooding
138 during the summer transport organic matter accumulated in the lake sediments (Maruya et al.,
139 2010; Maruya, 2014). According to Capone and Kiene (1988) and Minami et al. (2012), since
140 CH_4 is produced in sediments under anaerobic conditions, CH_4 production in the lower layer of
141 Lake Abashiri is likely more extensive than that in the other freshwater lakes (Sasaki and Endo,
142 2014).

143 An essential characteristic of Lake Abashiri is that a large amount of CH_4 is released from
144 the water surface once a year or once every two years. Maruya et al. (2010) revealed that the
145 lower layer water with high DM concentration reaches the water surface due to upwelling when
146 wind speeds exceed 10 m/sec for more than 5 hr, causing CH_4 gas to be released from Lake
147 Abashiri. Maruya et al. (2010) also suggested that the upwelling is associated with the stability
148 of the pycnocline, and the CH_4 emissions due to the upwelling can be modeled using the
149 Effective Wedderburn number (Shintani et al., 2010) and Lake number (Imberger and Patterson,
150 1990). Therefore, the vertical salinity profile was measured every 1 hr by Japan's Ministry of
151 Land, Infrastructure, and Transport using a rise and fall system. Wind vectors were measured
152 every 1 hr in 2012 and 2013, and the river discharge was estimated based on water depth by
153 applying the relationship between water depth and river discharge.

Comment E-2

154 Capone and Kiene (1988) and Minami et al. (2012) showed that CH_4 is produced in
155 sediments under anaerobic conditions. However, Watson and Nedwell (1998) showed that
156 sulfate-reducing bacteria decrease the activity of methanogenic bacteria under anaerobic

conditions. It is also known that CH_4 is oxidized under anaerobic conditions by sulfate-dependent anaerobic CH_4 oxidation (Caldwell et al., 2008). We therefore conducted cultivation experiments to estimate the CH_4 production and measured sulfate concentrations (SO_4^{2-}) using extracted pore water from sediments obtained at Station (St.) I on August 7, 2013. Since CH_4 can be easily transported from sediments into the water column, it is necessary to sample sediments intact, without disturbance. We thus used a non-disturbing core sampler (HR-type Core Sampler; RIGO, Japan) to sample anaerobic sediments (Appendix A **Fig. S1**). We sliced the core sample into four layers under oxic conditions in a laboratory: 0–0.05 m, 0.05–0.10 m, 0.10–0.15 m, and 0.15–0.20 m. After obtaining the sliced sediment, pore water was extracted using a centrifuge. The DO of pore water was anoxic (about 0.0 mg/L). The overlying water was sampled using a Van Dorn water sampler (RIGO). Sulfate concentrations (SO_4^{2-}) of the pore water and the overlying water were measured using a DR/2400 portable spectrophotometer (Hach, USA) and Hach kits following the SulfaVer4 method, which has been used widely in previous studies (e.g., Mwangangi et al., 2007; Liu et al., 2010). The sulfate concentration over the maximum limit of determination (70 mg/L) was measured by diluting pure water. Because the activity of methanogenic bacteria changes as a function of water temperature, it is necessary to measure DM concentrations under constant water temperature. Therefore, one-fifth of a 100 mL vial container (φ 0.004 m × 0.13 m) was filled with the sampled sediment, the remainder was filled with the pore water, and the sample was cultivated under a water temperature of 20°C using a thermostatic shaking incubator (AT12R; Thomas Kagaku, Japan). DM concentrations were measured by a headspace technique using a gas chromatograph coupled to a flame ionization detector (Sasaki and Endo, 2014) (GC-FID: GC-8A; Shimadzu, Japan).

Comment E-3

Comment E-2

Comment E-2

1.2 Methane observation

To evaluate the CH_4 production from sediments and investigate the effect of the DO concentration on CH_4 production, seven sediment samples were taken from four stations, B, I, S, and Y, from 2012 to 2013 using an undisturbed core sampler (**Fig. 1b** and **Table 1**). Vertical water temperature profiles, salinity, and DO concentration were also measured using a water quality profiler (AAQ1183; JFE Advantech, Japan). We used the first layer (0–0.05 m) of sediment mainly to evaluate the CH_4 production, as this layer is considered to play a more significant role in CH_4 release than the second to fourth layers (as discussed below in section 2.2). In the cultivation experiments, the DO concentrations in the pore water were set at 0.0 mg/L to mimic actual conditions occurring in the sediments of Lake Abashiri. The CH_4 production was evaluated using Eq. (2):

Comment E-2

$$P_M = \frac{A_S h(M_t - M_{initial})}{A_S t_e} \quad (2)$$

where P_M is the CH_4 production from sediment (μmol/m^2/day), A_S is the horizontal cross-area of sediment (m^2), h is the water depth in the vial container (= 0.10 m, with a vial container size of φ 0.004 m × 0.13 m and a volume of 100 mL), M_t is the DM concentration in the cultivation experiment (μmol/L), $M_{initial}$ is the initial DM concentration (μmol/L), and t_e is the duration (day).

Comment E-2

Comment E-2

Unlike in the lower layer, DO concentrations in the upper layer are saturated due to wind forcing, and thus oxidation plays a significant role in determining DM concentrations in the upper layer. Roulet et al. (1997) demonstrated that CH_4 emissions were negatively correlated with DO concentrations in a beaver pond in Canada, suggesting that microbes oxidize CH_4 under aerobic conditions and that CH_4 emissions decrease with increasing DO concentrations. We therefore investigated the oxidation effect on CH_4 production using cultivation experiments under aerobic conditions (Baba et al., 2011). Cultivation experiments were conducted under incubator temperatures of 15℃ and 30℃ using the samples from St. I. The vial bottle was sealed to prevent oxygen exchange with the air. Two different water temperatures were used in the laboratory experiments, 15℃ and 30℃. At 15℃, the DO concentrations were 9.7, 11.0, 10.6 and 9.5 (mg/L) on days 0, 11, 22, and 30, respectively. At 30℃, the DO concentrations were 9.7, 9.4, 9.2, and 8.8 (mg/L), respectively, on the same days.

Comment E-2

1.3 One-dimensional methane model

In a previous study, Bastviken et al. (2004) revealed that dissolved methane (DM) concentrations were associated with the surface areas of lakes in Sweden and North America. The smaller the surface area of a lake, the larger the DM concentration. They expressed this relation as

$$M = 262.42 A_H^{-0.227} \quad (3)$$

where M is the DM concentration (μmol/L) and A_H is the surface area of a lake (m^2).

Comment E-2

Sasaki and Endo (2014) conducted field observations at 12 lakes in Hokkaido and measured DM concentrations adjacent to the water surface (**Fig. 1a** and **Fig. 2a**). Their results revealed a similar trend in DM concentration in Hokkaido (Eq. (4)):

$$M = 381.63 A_H^{-0.498} \quad (4)$$

The absolute value of the power function coefficient in Eq. (4) is larger than that in Eq. (3) (**Fig. 2a**). One possible reason for this difference would be the difference in locations studied. Although both field observations were carried out in cold regions, Bastviken et al. (2004)

224 performed their observations in Sweden and North America, and Sasaki and Endo studied lakes
225 in northern Japan. However, the annual mean air temperature is about 6.6℃ in Sweden and 8.4℃
226 in North America, while that in Hokkaido is 6.1℃. These similar values suggest that air
227 temperature may not have been responsible for the difference between Eqs. (3) and (4).
228 Therefore, water quality was considered another external condition that might have contributed
229 to the difference in the power function coefficient. Bastviken et al. (2004) carried out field
230 observations in lakes with low nutrient levels, while Sasaki and Endo (2014) conducted field
231 observations in high-nutrient lakes. Although there may have been other factors that affect DM
232 concentrations, eutrophication probably increases DM concentrations in nutrified lakes
233 compared with lakes with low nutrient levels, which may explain the differences in the
234 coefficients of Eqs. (3) and (4). Sasaki and Endo (2014) suggested the importance of clarifying
235 CH_4 production in a brackish and eutrophic lake with strong stratification because of the higher
236 CH_4 flux. They showed that anaerobic CH_4 oxidation with sulfate reduction did not significantly
237 impact CH_4 production from the bottom sediment and that hypoxia had the predominant impact
238 on CH_4 production.
239 To elucidate the mechanism of CH_4 production, the reproducibility of the vertical CH_4 profile
240 was evaluated using a one-dimensional model (methane model), which was developed based
241 on the CH_4 production estimated by Eq. (5) as follows:

242 $$\frac{1}{A_z}\frac{\partial}{\partial t}\left(\int MdA\right) = \frac{\nu}{A_z}\frac{\partial^2}{\partial z^2}\left(\int MdA\right) + \frac{P_M}{A_z}\frac{\partial A}{\partial z} \quad (5)$$

243 where A_z is the area in each depth (m^2), M is the DM concentration (μmol/L), ν is the diffusion
244 coefficient (m^2/sec), and P_M is the CH_4 production (1,400 μmol/m^2/day).

Comment E-2

245 The methane model is a simple diffusion model with the release of CH_4 from bottom
246 sediment, including the effect of the horizontal cross-sectional area on CH_4 diffusion. Okada
247 and Nakayama (2007) demonstrated the high applicability of a one-dimensional vertical DO
248 model to examine long-term changes in DO concentrations in an enclosed bay; their results
249 showed good agreement with the field observations. However, in their model, the diffusion
250 coefficient must be decided by trial and error. Since we previously carried out three-
251 dimensional numerical simulations in Lake Abashiri using Fantom modeling (Nakayama et al.,
252 2012; Nakayama et al., 2014; Nakayama et al., 2016; Nakayama et al., 2019; Nakayama et al.,
253 2020), we know that this method can be used to appropriately estimate the vertical distribution
254 of the diffusion coefficient (Maruya et al., 2010).
255 We decided to neglect ebullition in our model for the following reasons. Bastviken et al.
256 (2004) revealed the importance of ebullition and the water column storage effect. Tokida et al.

(2005) demonstrated that air pressure change is significant in CH_4 ebullition in a shallow lake. CH_4 is produced mainly at a water depth of 10 m to 15 m in Lake Abashiri, meaning that the impact of air pressure on hydrostatic pressure is minor compared to that in a shallow lake. Moreover, the previous studies in Lake Abashiri also showed that diffusion is predominantly relative to ebullition (Kato et al., 2006; Kato et al., 2007; Jin et al., 2008; Wakamatsu et al., 2009; Wakamatsu et al., 2010; Baba et al., 2012; Miura et al., 2013). Ebullition is almost negligible in Lake Abashiri. Therefore, we applied only the diffusion of DM in our methane model. The vertical mesh size and computational time step were set to 0.5 m and 0.1 days, respectively, for Eq. (5).

To verify the accuracy of the methane model, we compared it with the vertical profile of DM concentrations of field observation on June 11, 2008, July 22, 2008, and September 29, 2008, at St. I by Miura et al. (2013). DM concentrations were measured by GC-FID and cultivation experiments (see section 1.2 for details).

2 Results

2.1 Stratification and DM vertical profile in Lake Abashiri

Lake Abashiri is a typical brackish lake, and has strong stratification (**Fig. 1b**). The pycnocline thickness ranges from about 0.5 m in summer to 1.0 m in winter. Bottom sediments consume dissolved oxygen in the hypolimnion, and the strong stratification suppresses the DO flux between the hypolimnion and epilimnion stratification. Therefore, the DO concentration tends to become zero, generating anoxia (**Fig. 2b**). Anoxic water always exists in the lower layer, and mass transport between the upper and lower layers is suppressed due to stratification, resulting in high DM concentrations in the lower layers (**Fig. 3**). Because the vertical profiles of DM concentrations are always the same in Lake Abashiri under calm conditions, the CH_4 production from the sediment is expected to be balanced by CH_4 production from the bottom sediment, DM flux from the lower to upper layers, oxidation in the upper layer and CH_4 emission from the water surface. Therefore, to evaluate CH_4 emission from the water surface in a eutrophic lake, the total CH_4 production from anaerobic sediments, accompanied by CH_4 oxidation under aerobic conditions, must be clarified. Since Lake Abashiri is a typical brackish and eutrophic lake with an aerobic/anaerobic state, it is considered one of the most suitable lakes for investigating CH_4 production in anaerobic layers and aerobic layers. Regarding the essential characteristics of Lake Abashiri, the pH, total nitrogen (TN), total organic carbon (TOC), and potential oxidation-reduction data in the upper (above pycnocline: about 5 m) and lower (under

pycnocline: about 5 m) layer in 2008 were 8.0–9.2 and 7.2–9.4, 0.35–1.0 and 14–15 (mg/L), 3.1–4.4 and 3.7–4.3 (mg/L), and 140–220 and –380 (mV), respectively.

Comment E-2

Wind vectors at St. I, river discharge at St. 0, and the vertical profile of water temperature and salinity at St. I in 2012 and 2013 are shown in **Fig. 4**. When strong south-southeast wind occurred at the beginning of September in 2012, the upper and lower layers were well mixed. The pycnocline level, which corresponds to a large gradient in salinity, decreased by a few meters (**Fig. 4d**). Maruya et al. (2010) found that upwelling occurred at the beginning of September in 2012 using a three-dimensional numerical model, Fantom. The vertical level of the pycnocline decreased by mixing due to upwelling. Upwelling and decrease in the pycnocline level were also confirmed at the beginning of August in 2013 (**Fig. 4h**). After the upwelling phenomena, strong stratification was rapidly generated as denser oceanic water intruded into the lower layer from the ocean in Lake Abashiri.

2.2 Methane production from bottom sediment

As Bastviken et al. (2008) suggested, one factor in reducing DM concentrations is microbial CH_4 decomposition, which converts CH_4 to CO_2 due to oxidation under aerobic conditions (Reeburgh, 2003; Kiene, 1991; Galchenko et al., 1989). Also, the methanogenic bacteria may be inactivated under non-anoxic conditions. Capone and Kiene (1988) and Minami et al. (2012) showed that CH_4 is produced under anaerobic conditions with low sulfate-reducing bacteria.

The CH_4 production rate has a strong relation to the bottom sediment characteristics. If sediment characteristics are different even in the same lake, the CH_4 production rate is expected to vary spatially. Therefore, we determined the geochemical composition of the bottom sediment at Sts. B, F, H, I, K, and M in Lake Abashiri on June 25, 2008, using X-ray fluorescence analysis (Supermini, Rigaku, Japan) to clarify the characteristics of the distribution of bottom sediment (**Fig. 5** and **Table 2**). It was found that the bottom sediments in Lake Abashiri were uniformly accumulated. Therefore, the organic loading impacts of the bottom sediment distribution on CH_4 production could be minor in the lake. In addition, TOC from organic matter would impact CH_4 production. Sampei et al. (1997) demonstrated that the TOC concentration at the bottom sediment ranges from 3%–4% in Lake Abashiri, meaning there are no significant differences in TOC. Therefore, since the distribution of TOC is almost uniform, the impact of TOC on CH_4 production would be negligible in Lake Abashiri.

Oceanic water from the Okhotsk Sea has a high concentration of sulfate, about 2,700 mg/L. Since the salinity of the lower layer in Lake Abashiri reflects oceanic water intrusion from the Okhotsk Sea through the Abashiri River, sulfate concentrations in the overlying water were

around 820–1,020 mg/L (mean value ~900 mg/L). On the other hand, sulfate concentrations in pore water from the first to the fourth layer (from 0 to 0.20 m) were almost 0.0 mg/L (**Fig. 6**). The long-term field observations revealed that DM and the other water quality components did not vary spatially and temporally in Lake Abashiri from 2006 to 2012, and thus it is reasonable to assume that the methane production from the bottom sediment also did not change substantially (Kato et al., 2006; Kato et al., 2007; Jin et al., 2008; Wakamatsu et al., 2009; Wakamatsu et al., 2010; Baba et al., 2012; Miura et al., 2013). Therefore, these findings may suggest that the sulfate concentration in the bottom sediment does not change significantly due to the strong stratification effect in a brackish lake. Kasten and Jørgensen (2000) suggested that anaerobic methane oxidation is induced by sulfate reduction in brackish environments. In this study, sulfate concentration decreased rapidly from about 900 mg/L in the water column above bottom sediment to 0.0 mg/L in the first sediment layer (**Fig. 6**). Sediments a few cm thick existed above the first sediment layer with a water content of about 80%. There is thus the possibility that anaerobic CH_4 oxidation due to sulfate reduction occurs in the high-water content layer. The CH_4 production layer thickness is more than 20 cm, resulting in a reduction in CH_4 production to the range of 1,400–1,800 μmol/m^2/day, which is more significant than the reduction in CH_4 in the high-water content layer. Therefore, CH_4 was probably released from the first or a deeper sediment layer to the water column without anaerobic CH_4 oxidation induced by sulfate reduction.

Comment E-2

Comment E-2

Comment E-2

Although it has been established that CH_4 is produced under anaerobic conditions (Capone and Kiene, 1988; Minami et al., 2012), the effect of DO concentrations on CH_4 production has not been sufficiently clarified. Since Lake Abashiri is a typical brackish lake and has strong stratification, DO concentrations in water overlying the sediment do not change for more than a few months a year. Therefore, Lake Abashiri is considered a highly suitable lake to investigate the effect of overlying DO concentrations on CH_4 production from the sediment. CH_4 production was thus plotted against DO concentrations in the overlying water (**Fig. 7**), based on seven samples from four stations (**Table 1**). In the cultivation experiment performed at St. B on June 29, 2012, the incubation temperature was 15°C, which is slightly lower than that of other cultivation experiments (generally 20°C). However, experiments using the same DO concentration (St. I on August 7, 2013) showed that the CH_4 production was almost the same under different temperatures (**Fig. 7** and **Table 1**). Although methanogenic archaea activity varies with water temperature changes (Conrad et al., 1987), the water temperature may have little influence on CH_4 production in Lake Abashiri. Note that the water temperature of the lower layer is generally from 10.0°C to 20.0°C throughout the year in Lake Abashiri. To

estimate the CH_4 production from bottom sediments using DO concentrations in water overlying the sediments, we propose a model equation with a logistic function as follows:

$$P_M = \frac{a}{[1+\exp\{b \cdot (DO + c)\}]} \quad (6)$$

where P_M is the CH_4 production from sediment (μmol/m^2/day), DO is the DO concentration (mg/L), and a, b and c are the coefficients for the logistic function (a = 1,400.0, b = 2.33, c = –3.97), respectively.

Comment E-2

2.3 Application of the methane model to a naturally eutrophic lake

Wakamatsu et al. (2010) revealed that CH_4 emission from Lake Abashiri is 355 μmol/m^2/day. Since the DM concentration in the upper layer of Lake Abashiri is 587 μmol/m^3 (Sasaki and Endo, 2014), the CH_4 release rate can be estimated as 355 μmol/m^2/day / 587 μmol/m^3 = 0.60 m/day. Because the upper layer thickness is about 6.0 m in the absence of strong winds over 10.0 m/sec, which could cause the pycnocline to fluctuate (Maruya et al., 2010), the exchange time can be estimated as 6.0 m / 0.60 m/day = 10.0 d. On the other hand, assuming that DO concentrations were 0.0 mg/L, the CH4 production in Lake Abashiri is estimated as 27,900 mol/day using Eq. (6), because the bottom area below the pycnocline is about 19.96 km^2. As the surface area of Lake Abashiri is 32.3 km^2, the CH_4 production converted to a unit area for the water surface is 27,900 mol/day / 32.3 km^2 = 864 μmol/m^2/day. Therefore, CH_4 in the upper layer under aerobic conditions is estimated to be reduced by 355 μmol/m^2/day / 864 μmol/m^2/day = 0.41 (41%) compared to CH_4 in the lower layer. The time required to reduce CH_4 by 41% is estimated as 9.1 and 10.3 days based on incubator temperatures of 15℃ and 30℃, respectively. Cultivation experiments revealed that DM concentrations decreased to 50% for about 8 days due to oxidation under aerobic conditions (**Fig. 8**), close to the exchange times of 9.1 and 10.3 days. Therefore, oxidation is expected to be the dominant reduction mechanism of DM concentrations in the upper aerobic layers in Lake Abashiri.

Comment E-2

Comment E-2

The wind was the only significant factor controlling the upper layer mixing during the study period because the inflow effect was negligible (**Fig. 4**). Since wind speed was low and did not vary substantially, the entrainment velocity within the pycnocline was constant. Maruya et al. (2010) revealed that wind speed over 10 m/sec induces large-amplitude internal waves, which changes the pycnocline level. Maruya et al. (2010) also suggested that the vertical diffusion coefficient is constant when wind speed is weak, i.e., less than 10 m/sec, as in this study. Therefore, we used a constant vertical diffusion coefficient. The diffusion coefficient of CH_4 between the upper and lower layers (aerobic and anoxic layers) in Lake Abashiri was

Comment E-2

Comment E-2

revealed to be 1.284×10^{-7} m^2/sec, as estimated from field observations (Wakamatsu et al., 2010). Therefore, CH_4 flux from the upper to lower layers was suppressed due to the strong stratification. Moreover, DM concentrations in 2008 were almost constant in the upper layer, at about 0.2 μmol/L (Miura et al., 2013). To evaluate the applicability of the methane model, the DM concentration in the upper layer was thus given a constant of 0.2 μmol/L. Since the diffusion coefficient for CH_4 on the bottom is higher than that of the pycnocline, CH_4 is easily mixed due to turbulence owing to internal wave motions. Therefore, the diffusion coefficient was given linearly from 1.0×10^{-6} m^2/sec at the pycnocline to 1.083×10^{-6} m^2/sec at the bottom layer by referring to three-dimensional numerical simulations (Maruya et al., 2010). The methane model successfully reproduced the vertical CH_4 profiles under calm conditions, discounting significant disturbance events such as strong winds that cause blue tides, since no such winds occurred during the field observations of 2008 (Miura et al., 2013) (**Fig. 3**).

Comment E-2

Comment E-2

Comment E-2

3 Discussion

In this study, we observed that methane was produced in the accumulated bottom sediment at Lake Abashiri at high rates of 1,400–1,800 μmol/m^2/day under low DO concentrations due to the strong stratification. Additionally, methane production from the bottom sediment decreased with increased DO concentrations around the pycnocline. To confirm that our findings on methane production were comparable to those observed at another eutrophic lake, we compared our results with those of Takii et al. (1997). Takii et al. (1997) sampled sediments from the sediment surface to a sediment depth of 0.12 m in Lake Kizaki from 1992 to 1994. Lake Kizaki is a typical eutrophic lake. DM concentrations were measured using sealed vial containers, in which the headspace above the sediment sample was filled with nitrogen gas. They conducted cultivation experiments with an incubator temperature of 6℃. In contrast, we used DM concentrations from 1993 to 1994, because hypoxic water was expected to have existed continuously without any change in stratification over that period. At Lake Kizaki, the annual mean CH_4 production was 1,470 μmol/m^2/day (800 to 1,800 μmol/m^2/day) under anaerobic conditions (Takii et al., 1997). In our study, the CH_4 production was 1,400 μmol/m^2/day from Eq. (6) when the DO concentration was 0.0 mg/L, which is almost the same as that in Takii et al. (1997). These findings may suggest that 1,400 μmol/m^2/day is the approximate universal value of the maximum methane production from the bottom sediment in a stratified eutrophic lake. However, a deeper understanding and clarification of the methane production from the bottom sediment will be needed to develop a universal model equation in future studies.

Comment E-2

Comment E-2

Equation (6) may be a first step toward the detailed estimation of methane diffusive emission for implementing more adequate and accurate mitigation measures.

Based on their field observations in 2009, Wakamatsu et al. (2010) revealed that diffusive CH_4 flux from the water surface to the atmosphere is about 350 μmol/m^2/day under average meteorological conditions. In the present study, the maximum methane production from the bottom sediment was about 1,400 μmol/m^2/day, which is much larger than the diffusive CH_4 flux from the water surface to the atmosphere. Diffusive CH_4 flux can be reduced in two ways: by reducing oxidation in the upper layer or by reducing vertical flux suppression due to stratification. Maruya (2014) suggested that the suppression of the vertical methane flux plays a more significant role in reducing diffusive CH_4 flux from the water surface to the atmosphere than does the suppression of the oxidation in the upper layer. Consequently, methane is accumulated in the lower layer, resulting in a high methane concentration of about 400 μmol/L in the lower layer, as Wakamatsu et al. (2010) found. The accumulation of a high methane concentration will not cause any problem in the ecosystem if the methane stays in the lower layer. However, a strong wind will sometimes cause upwelling, potentially causing a large amount of methane to be released from the water surface to the atmosphere for a short period. Therefore, the outcome of this study may contribute to research on the significant burst-like diffusive methane release from the water surface to the atmosphere. Also, it may suggest the necessity of developing a method for estimating methane concentration in a lake efficiently and accurately, such as by using a simple one-dimensional vertical column model.

Comment E-2

Comment E-2

Comment E-2

The one-dimensional methane model can estimate CH_4 for a wide variety of lake characteristics. For example, stratification varies seasonally in freshwater lakes, resulting in an aerobic to anoxic lower layer from winter to summer. The developed methane model can evaluate how DO affects the vertical CH_4 profile of the lower layer by giving the different DO rates. Since CH_4 production varies from 0 μmol/m^2/day to 1,400 μmol/m^2/day, we applied a DO range from 0 to 6 mg/L to the methane model to investigate the effect of DO on the DM vertical profile (**Fig. 9**). As expected from Eq. (6), the DM concentrations decreased rapidly with DO changes from 3 mg/L to 5 mg/L. Since the maximum DO concentration is about 8 mg L^{-1} in Lake Abashiri, DO of 5 mg/L is considered an aerobic condition, which shows that the DM volume is about 10% when the lower layer is anoxic. Hypoxia occurs more readily in a eutrophic lake than an oligotrophic lake because of the more significant accumulation of particulate organic matter at the eutrophic lake bottom. We revealed that the lower the DO concentration, the higher the DM production, as shown in **Fig. 7** and Eq. (6). Therefore, CH_4

Comment E-2

Comment E-2

production due to diffusion tends to be significant in Lake Abashiri, a brackish and eutrophic lake. According to Sasaki and Endo (2014), the DM concentration in Lake Abashiri is about ten times larger than that in other oligotrophic lakes of the same size (**Fig. 2a**). Note that diffusion, rather than ebullition, was the primary source of CH_4 emission in the lakes investigated by Sasaki and Endo (2014).

4 Conclusions

A methane production equation with a logistic function was proposed to estimate the CH_4 production from bottom sediments in a eutrophic lake using the DO concentrations in water overlying the sediments. Our results showed that CH_4 production from the bottom sediments was 1,400 μmol/m^2/day when the DO concentrations in water overlying the sediments ranged from 0 mg/L to 3 mg/L. In addition, the developed vertical methane model was successfully applied to evaluate a vertical CH_4 profile in Lake Abashiri with a fair agreement to the field observations. The DM and DO concentrations vary due to internal waves in stratified flow fields, which determine the vertical diffusion coefficients in a one-dimensional model. Therefore, applying a detailed three-dimensional numerical model may provide a more accurate spatiotemporal estimation of the methane concentration in a eutrophic lake. However, since the running cost of the vertical methane model is low, it will be easy to apply the model to many different lakes over a long-term investigation. Therefore, the developed model could project methane flux due to climate change in future studies.

Comment 1-1

Comment E-2

Comment 1-1

Comment 1-1

Acknowledgments

This work was supported by JSPS KAKENHI Grant Numbers JP26420491, JP14J03382, JP18KK0119, JP18H01545, JP18K13835, and JP21H05178.

Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version.

References

Baba, Y., Hirokawa, M., Sasaki, M., Endoh, N., 2011. Air-Lake exchange of methane in Hokkaido. Proc. of Jpn. Soc. Mech. Eng. 50, 79–80. (in Japanese)

Bastviken, D., Ejlertsson, J., Tranvik, L., 2002. Measurement of Methane Oxidation in Lakes: A Comparison of Methods, Environ. Sci. Technol. 36, 3354−3361.

491 Bastviken, D., Cole, J.J., Pace, M.L., Tranvik, L., 2004. Methane emissions from lakes:
492 Dependence of lake characteristics, two regional assessments, and a global estimate.
493 Global Biogeochem. Cycles 18, GB4009, doi:10.1029/2004GB002238
494 Bastviken, D., Cole, J., Pace, M.L., Van de Boger, M.C., 2008. Fates of methane from different
495 lake habitats: Connecting whole-lake budgets and CH_4 emissions. J. Geophys. Res. 113,
496 G02024, doi:10.1029/2007JG000608
497 Bastviken, D., Tranvik, L.J., Downing, J.A., Crill, P.M., Enrich-Prast, A., 2011. Freshwater
498 methane emissions offset the continental carbon sink. Science 331, 50,
499 DOI:10.1126/science
500 Caldwell, S. L., Laidler, J.R., Brewer, E.A., Eberly, J.O., Sandborgh, S.C., Colwell, F.S., 2008.
501 Anaerobic oxidation of methane: mechanisms, bioenergetics, and the ecology of
502 associated microorganisms. Environ. Sci. Technol. 42(18), 6791.
503 Capone, D.G., Kiene, R.P., 1988. Comparison of microbial dynamics in marine and freshwater
504 sediments: Contrasts in anaerobic carbon catabolism. Limnol. Oceanogr. 33, 725-749,
505 DOI:10.4319/lo.1988.33.4_part_2.0725
506 Cicerone, R.J., Oremland R S., 1988. Biogeochemical aspects of atmospheric methane. Global
507 Biogeochem. Cycles 2, 299-327, DOI: 10.1029/GB002i004p00299
508 Conrad, R., Schutz, H., Babbel, M., 1987. Temperature limitation of hydrogen turnover and
509 methanogenesis in anoxic paddy soil. FEMS Microbiol. Ecol. 45, 281-289, DOI:
510 10.1111/j.1574-6968.1987.tb02378.x
511 Di Tero, D.M., Paquin, P.R., Subburamu, K., Gruber, D.A., 1990. Sediment oxygen demand
512 model: methane and ammonia oxidation. J. Environ. Eng. 116(5), 945–986,
513 https://doi.org/10.1061/(ASCE)0733-9372(1990)116:5(945).
514 Fernández, J.F., Peeters, F., Hofmann, H., 2014. Importance of the Autumn Overturn and
515 Anoxic Conditions in the Hypolimnion for the Annual Methane Emissions from a
516 Temperate Lake. Environ. Sci. Technol. 48(13), 7297-7304, DOI: 10.1021/es4056164.
517 Galchenko, V.F., Lein, A., Ivanov, M., 1989. Biological sinks of methane. in: Andreae, M.O.,
518 Schimel, D.S. (Eds.), Exchange of Trace Gases Between Terrestrial Ecosystems and the
519 Atmosphere, Chichester , New York : Wiley, 59–71.
520 Hantush, M.M., 2007. Modeling nitrogen-carbon cycling and oxygen consumption in bottom
521 sediments. Adv Water Resour 30, 59–79, doi.10.1026/j.advwatres.2006.02.007.
522 Imberger, J., Patterson, J.C., 1990. Physical limnology. in: Hutchinson, J., Wu, T. (Eds.),
523 Advances in applied mechanics, Elsevier, 27, 303–475.

IPCC, 2007. Climate Change 2007. The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. 4. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

IPCC, 2013. In: Stocker, T.F., et al. (Eds.), Climate Change 2013: The Physical Science Basis in Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (Cambridge and New York).

Jin, T., Sasaki, M., Endoh, N., Kato, T., 2008. Air-lake exchange of methane in Hokkaido. Proc. of Jpn. Soc. Mech. Eng. 47, 157–158. (in Japanese)

Kasten, S., Jørgensen, B.B., 2000. Sulfate reduction in marine sediments, Marine geochemistry. Springer, pp. 263–281.

Kato, T., Fujii, H., Tomita, T., Endo, N., Sasaki, M., 2006. Methane exchange between inland waters and the atmosphere. Proc. of Jpn. Soc. Mech. Eng. 45, 189–190. (in Japanese)

Kato, T., Fujiwara, T., Endo, N., Sasaki, M., 2007. Air-lakes exchange of methane in Hokkaido. Proc. of Jpn. Soc. Mech. Eng. 46, 153–154. (in Japanese)

Kiene, R.P., 1991. Production and consumption of methane in aquatic systems, in: Rogers, J., Whitman W. (Eds.), Microbial Production and Consumption of Greenhouse Gases: Methane, Nitrogen Oxides, and Halomethanes, American Society for Microbiology, pp. 111–146.

Komori, D., Nakamura, S., Kiguchi, M., Nishijima, A., Yamazaki, D., Suzuki, S., Kawasaki, A., Oki, K., Oki, T., 2012. Characteristic of the 2011 Chao Phraya River flood in Central Thailand. Hydrol. Res. Lett. 6, 41–46, doi:10.3178/hrl.6.41.

Liu, Y.Q., Moy, B., Kong, Y.H., Tay, J.H., 2010. Formation, physical characteristics and microbial community structure of aerobic granules in a pilot-scale sequencing batch reactor for real wastewater treatment. Enzyme Microb. Technol. 46, 520–525, doi:10.1016/j.enzmictec.2010.02.001

Martinez, D., Anderson, M.A., 2013. Methane production and ebullition in a shallow, artificially aerated, eutrophic temperate lake (Lake Elsinore, CA). Sci. Total Environ. 454–455, 457-465.

Maruya, Y., Nakayama, K., Shintani, T., Yonemoto M., 2010. Evaluation of entrainment velocity induced by wind stress in a two-layer system. Hydrol. Res. Lett. 4, 70–74, doi:10.3178/hrl.4.70

556 Maruya, Y., 2014. Mechanisms of production of methane from benthic sediment in enclosed
557 water body. (Ph.D. thesis). Kitami Institute of Technology, Kitami, Hokkaido, Japan. (in
558 Japanese)
559 Minami, H., Tatsumi, K., Hachikubo, A., Yamashita, S., Sakagami, H., Takahashi, N., Shoji,
560 H., Jin, Y.K., Obzhirov, A., Nikolaeva, N., Derkachev, A., 2012. Possible variation in
561 methane flux caused by gas hydrate formation on the northeastern continental slope off
562 Sakhalin Island, Russia. Geo-Mar. Lett. 32, 525–534, DOI 10.1007/s00367-012-0287-x
563 Miura, A., Sasaki, M., Endoh, N., Hirano, S., 2013. Analysis of methane balance in a saline
564 lake (Lake Abashiri-ko). Proc. of Jpn. Soc. Mech. Eng. 52, 67–68. (in Japanese)
565 Mwangangi, J.M., Muturi, E.J., Shililu, J.I., Muriu, S., Jacob, B., Kabiru, E.W., Mbogo, C.M.,
566 Githure, J.I., Novak, R.J., 2007. Environmental covariates of Anopheles arabiensis in a
567 rice agroecosystem in Mwea, Central Kenya. J. Am. Mosq. Control Assoc. 23, 371–377,
568 doi:http://dx.doi.org/10.2987/5605.1
569 Nakayama, K., Shintani, T., Kokubo, K., Kakinuma, T., Maruya, Y., Komai, K., Okada, T.,
570 2012. Residual currents over a uniform slope due to breaking of internal waves in a two-
571 layer system. J. Geophys. Res. 117, C10002, doi:10.1029/2012JC008155.
572 Nakayama, K., Shintani, T., Shimizu, K., Okada, T., Hinata, H., Komai, K., 2014. Horizontal
573 and residual circulations driven by wind stress curl in Tokyo Bay. J. Geophys. Res. 119,
574 1977–1992, doi:10.1002/2013JC009396.
575 Nakayama, K., Nguyen, H. D., Shintani, T., Komai, K., 2016. Reversal of secondary
576 circulations in a sharp channel bend. Coast. Eng. J. 58, 165002,
577 https://doi.org/10.1142/S0578563416500029
578 Nakayama, K., Sato, T., Shimizu, K., Boegman, L., 2019. Classification of internal solitary
579 wave breaking over a slope. Phys. Rev. Fluids 4, 014801,
580 https://doi.org/10.1103/PhysRevFluids.4.014801
581 Nakayama, K., Sato, T., Tani, K., Boegman, L., Fujita, I., Shintani, T., 2020. Breaking of
582 Internal Kelvin Waves Shoaling on a Slope. J. Geophys. Res. 125, e2020JC016120,
583 https://doi.org/10.1029/2020JC016120.
584 Okada, T., Nakayama, K., 2007. Density intrusion and variation in dissolved oxygen
585 concentrations in a bay with a sill at its mouth. J. Environ. Eng., 133, 447–453,
586 doi:10.1061/(ASCE)0733-9372(2007)133:4(447).
587 Reeburgh, W.S., 2003. Global methane biogeochemistry, in: Keeling R. F. (Ed.), Treatise on
588 Geochemistry, Elsevier, pp. 65–89.

589 Roulet, N.T., Crill, P.M., Comer, N.T., Dove, A., Boubonniere, R.A., 1997. CO_2 and CH_4 flux
590 between a boreal beaver pond and the atmosphere. J. Geophys. Res. 102, 29313–29319,
591 DOI: 10.1029/97JD01237
592 Rudd, J.W.M., Hamilton, R.D., 1978. Methane cycling in a eutrophic shield lake and its effects
593 on whole lake metabolism. Limnol. Oceanogr. 23, 337–348,
594 DOI: 10.4319/lo.1978.23.2.0337
595 Sampei, Y., Kurakado, Y., Shimizu, A., 1997. Distribution of organic carbon, nitrogen and
596 sulfur in surface sediments of Lake Saroma and Lake Abashiri, Hokkaido, Japan. Res. Org.
597 Geochem. 12, 51–60, https://doi.org/10.20612/rog.12.0_51 (in Japanese)
598 Sasaki, M., Imura, S., Kudoh, S., Yamanouchi, T., Morimoto, S., Hashida, G., 2009. Methane
599 efflux from bubbles suspended in ice-covered lakes in Syowa Oasis, East Antarctica. J.
600 Geophys. Res. 114, D18114, doi:10.1029/2009JD011849
601 Sasaki, M., Endoh, N., Imura, S., Kudoh, S., Yamanouchi, T., Morimoto, S., Hashida, G., 2010.
602 Air-lake exchange of methane during the open water season in Syowa Oasis, East
603 Antarctica. J. Geophys. Res. 115, D16313, doi:10.1029/2010JD013822
604 Sasaki, M., Endoh, N., 2014. Exchanges of methane between lakes and the atmosphere in
605 Hokkaido, Subarctic Climate Region, Japan. J. Water Resour. Ocean Sci. 3, 89–94, doi:
606 10.11648/j.wros.20140306.14
607 Schubert, C.J., Diem, T., Eugster, W., 2012. Methane emissions from a small wind shielded
608 lake determined by eddy covariance, flux chambers, anchored funnels, and boundary
609 model calculations: A comparison. Environ. Sci. Technol. 46(8), 4515−4522, DOI:
610 10.1021/es203465x.
611 Shintani, T., A de la Fuente Nino, Y., Imberger J., 2010. Generalizations of the Wedderburn
612 number: Parameterizing upwelling in stratified lakes. Limnol. Oceanogr. 55, 1377–1389,
613 doi: 10.4319/lo.2010.55.3.1377
614 Striegl, R.G., Michmerhuizen, C.M., 1998. Hydrological influence on methane and carbon
615 dioxide dynamics at two north-central Minnesota lakes. Limnol. Oceanogr. 43, 1519–1529,
616 doi:10.4319/lo.1998.43.7.1519
617 Takii, S., Li, J.H., Hayashi, H., 1997. Methane production and sulfate reduction in profundal
618 sediments in Lake Kizaki, Japan. Japan. Jpn. J. Limnol. 58, 373–384,
619 doi.org/10.3739/rikusui.58.373
620 Taoka, T., Iwata, H., Hirata, R., Takahashi, Y., Miyabara, Y., Itoh, M. 2020. Environmental
621 controls of diffusive and ebullitive methane emissions at a subdaily time scale in the littoral

zone of a midlatitude shallow lake. J. Geophys. Res. Biogeosci. 125, e2020JG005753, doi: 10.1029/2020JG005753.

Tokida, T., Miyazaki, T., Mizoguchi, M., 2005. Ebullition of methane from peat with falling atmospheric pressure. Geophys. Res. Lett. 32, L13823, doi:10.1029/2005GL022949.

Utsumi, M., Nojiri, Y., Nakamura, T., Takeshi, N., Otsuki, A., Seki, H., 1998. Oxidation of dissolved methane in a eutrophic, shallow lake: Lake Kasumigaura, Japan. Limnol. Oceanogr., 43, 471–480, https://doi.org/10.4319/lo.1998.43.3.0471.

Wakamatsu, T., Sasaki, M., Endoh, N., Jin, T., 2009. Air-lake exchange of methane in Lake-Abashiri-ko. Proc. of Jpn. Soc. Mech. Eng. 48, 165–166. (in Japanese)

Wakamatsu, T., Sasaki, M., Endoh, N., Ito, Y., Baba, T., 2010. Air–Lake exchange of methane in Lake Abashiri-ko. Proc. of Jpn. Soc. Mech. Eng. 49, 131–132. (in Japanese)

Walter, K.M., Zimov, S.A., Chanton, J.P., Verbyla, D., Chapin, III F.S., 2006. Methane bubbling from Siverian thaw lakes as a positive feedback to climate warming. Nature 443, 71–75, doi:10.1038/nature05040

Watson, A., Nedwell, D.B., 1998. Methane production and emission from peat: the influence of anions (sulphate, nitrate) from acid rain. Atmos. Environ. 32, 3239–3245, DOI:10.1016/S1352-2310(97)00501-3

Yan, X., Xu, X., Ji, M., Zhang, Z., Wang, M., Wu, S., Wang, G., Zhang, C., Liu, H., 2019. Cyanobacteria blooms: A neglected facilitator of CH_4 production in eutrophic lakes. Sci. Total Environ. 651, 466–474.

643 **List of tables**

644 **Table 1**

645 Bottom sediment samples from 2012 to 2013. Incubation temperature, dissolved oxygen
646 concentration of the overlying water, and methane production are shown.

Station	Date	Incubation temperature (°C)	DO (mg/L)	P_M (μmol/m^2/day)
St. Y	August 8, 2013	20	5.88	25.79
St. S	October 23, 2012	20	4.08	601.9
St. B	June 29, 2012	15	0.14	1,335
St. B	October 23, 2012	20	1.48	1,274
St. B	November 26, 2012	20	3.02	1,241
St. I	October 23, 2012	20	1.09	1,463
St. I	August 7, 2013	20	0.05	1,398

Comment E-2

647

648

649 **Table 2**

650 Ratios of geochemical components in the bottom sediment in **Fig. 5**. Unit: %.

	St. B	St. F	St. H	St. I	St. K	St. M
Na_2O	1.18	1.54	1.47	1.23	1.66	1.26
MgO	1.35	1.26	1.28	1.19	1.15	1.20
Al_2O_3	14.91	11.90	12.92	13.01	13.11	12.20
SiO_2	57.62	56.28	57.37	58.15	55.26	55.22
P_2O_5	0.31	0.25	0.23	0.22	0.27	0.22
SO_3	1.49	2.31	2.63	2.39	2.24	2.59
Cl	0.11	0.85	0.66	0.59	0.75	0.60
K_2O	1.75	1.38	1.37	1.38	1.33	1.28
CaO	1.07	0.80	0.79	0.76	0.89	0.68
TiO_2	0.00	0.00	0.64	0.00	0.63	0.51
MnO	0.00	0.02	0.02	0.00	0.00	0.02
Fe_2O_3	0.03	0.02	6.35	0.03	0.03	0.02
Co_2O_3	6.03	5.95	0.02	6.40	5.83	6.45
ZnO	0.01	0.02	0.02	0.01	0.01	0.02
Rb_2O	0.02	0.01	0.01	0.00	0.00	0.01
SrO	0.01	0.02	0.01	0.01	0.02	0.01
C	14.11	17.39	14.22	14.61	16.82	17.71

651

652

List of figures

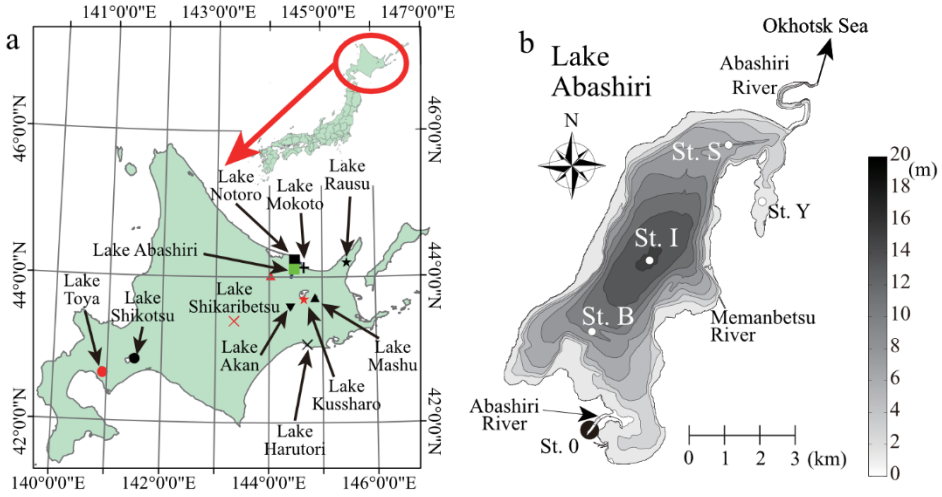


Fig. 1 Dissolved methane concentration adjacent to the water surface in lakes of Hokkaido, and field observation stations in Lake Abashiri. a: Hokkaido lakes from which water samples were collected. b: Lake Abashiri and field observation stations.

Comment E-2

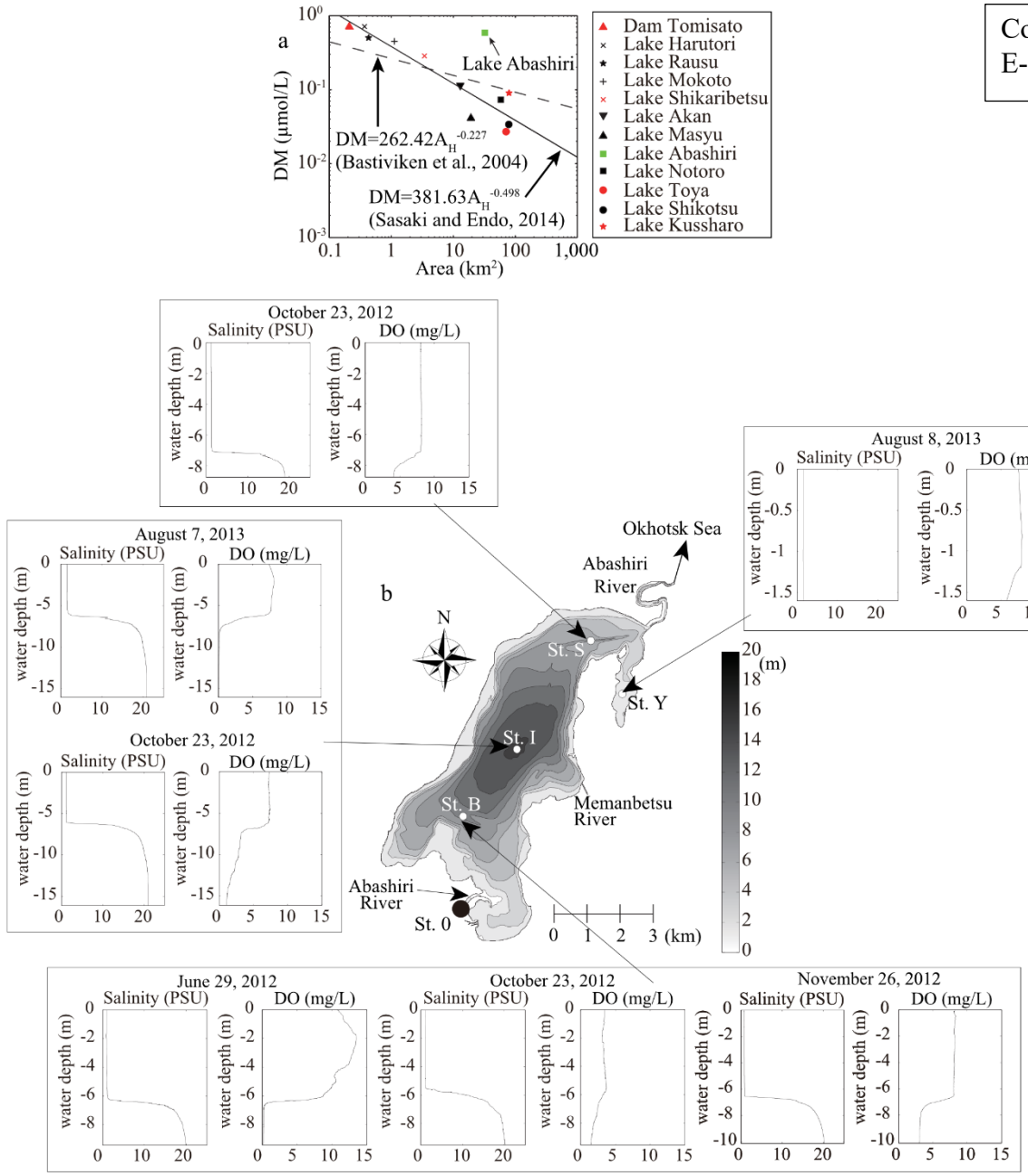
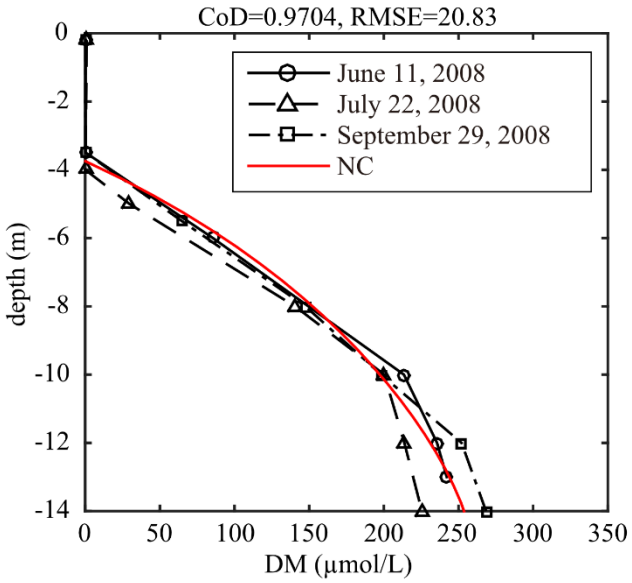


Fig. 2 a: Surface area of lakes and dissolved methane concentration (μmol/L) (Sasaki and Endo, 2014). b: The vertical profile of salinity (PSU) and dissolved oxygen concentrations (mg/L) in Lake Abashiri.

Comment E-2

Comment
E-2



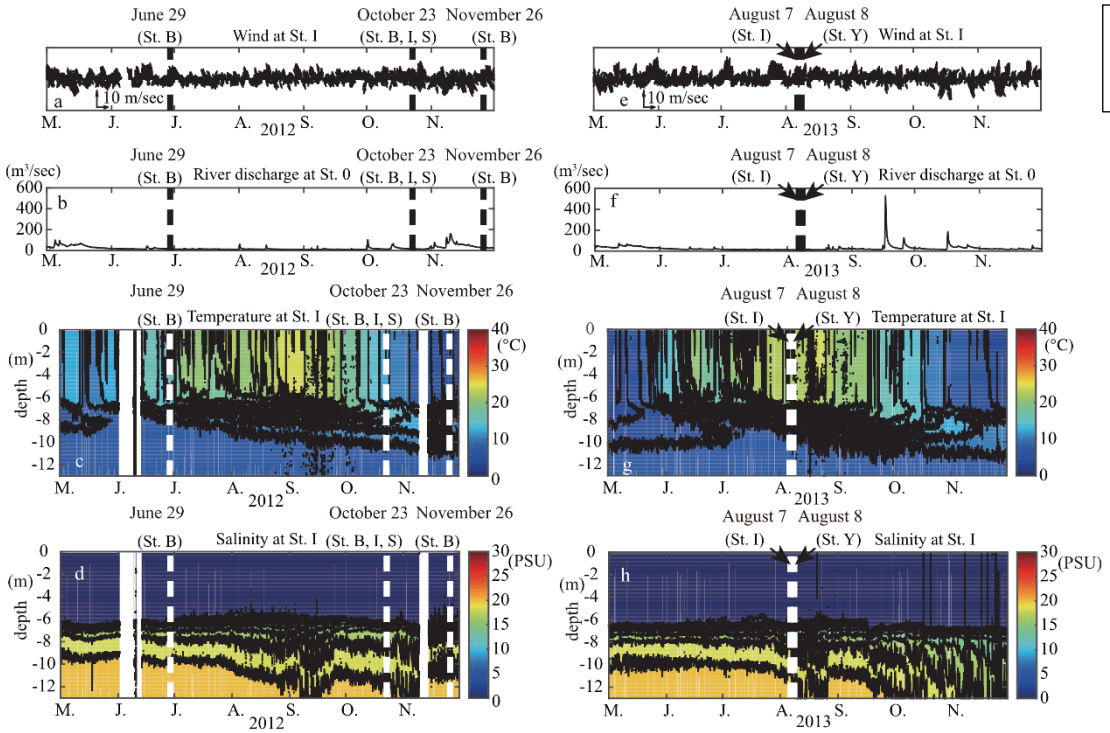
664

665 **Fig. 3** Vertical profile of dissolved methane concentrations at St. I in 2008 (Miura et al., 2013),
666 and comparison between observations and numerical simulations. NC indicates numerical
667 computation. The coefficients of determination (CoD) and root-mean-square error (RMSE)
668 were calculated by comparing the NC and average observation data.

669

670

Comment E-2

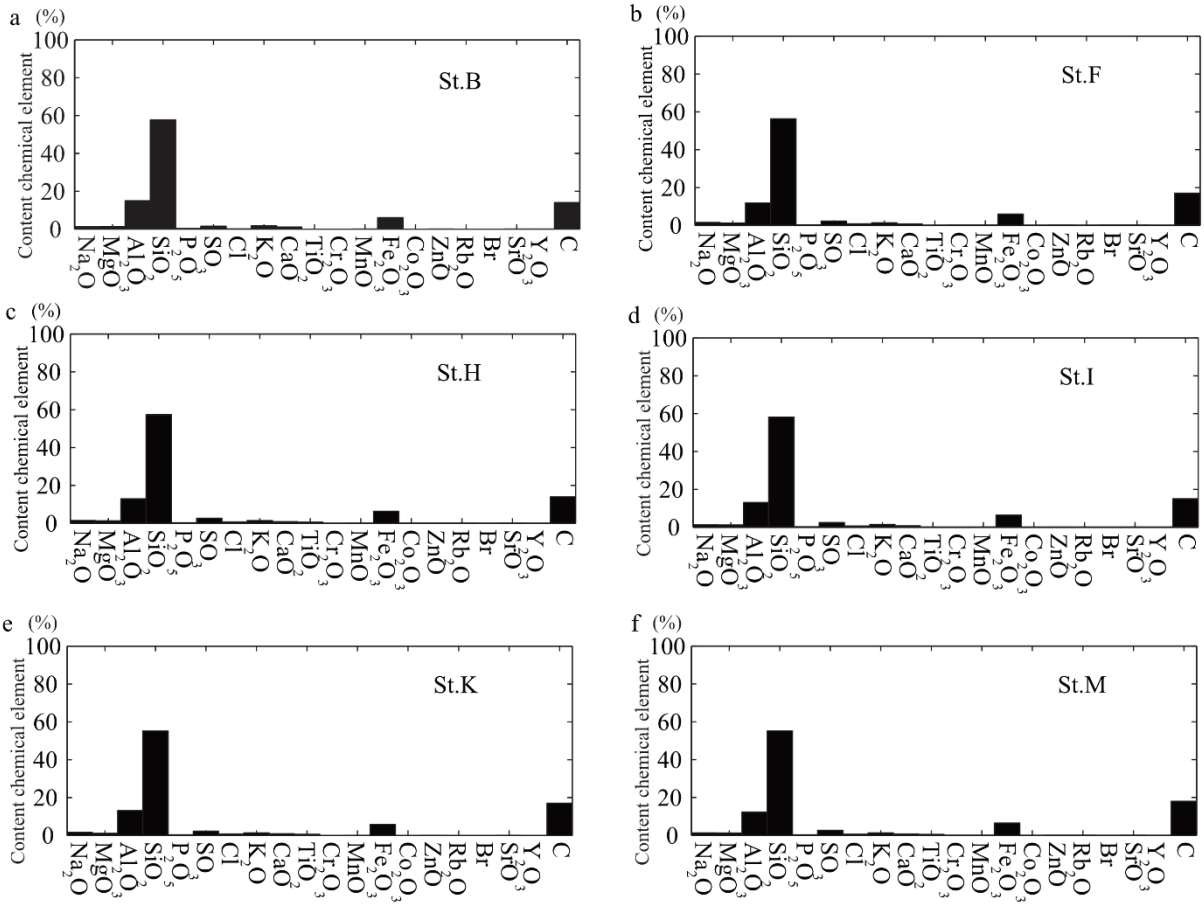


671

672 **Fig. 4** Wind vector, hydrograph, water temperature, and salinity in Lake Abashiri. White broken
673 lines indicate the date of sampling. a: Wind vector (m/sec) at St. I in 2012. b: River discharge
674 (m^3/sec) at St. 0 in 2012. c: Vertical profile of water temperature (°C) at St. I in 2012. d: Vertical
675 profile of salinity (PSU) at St. I in 2012. e: Wind vector (m s^{-1}) at St. I in 2013. f: River discharge
676 (m^3/sec) at St. 0 in 2013. g: Vertical profile of water temperature (°C) at St. I in 2013. h: Vertical
677 profile of salinity (PSU) at St. I in 2013.

Comment E-2

678



679

680 **Fig. 5** Geochemical composition in the bottom sediment by X-ray fluorescence analysis at a:

681 St. B; b: St. F; c: St. H; d: St. I; e: St. K; and f: St. M.

682

683

Comment E-2

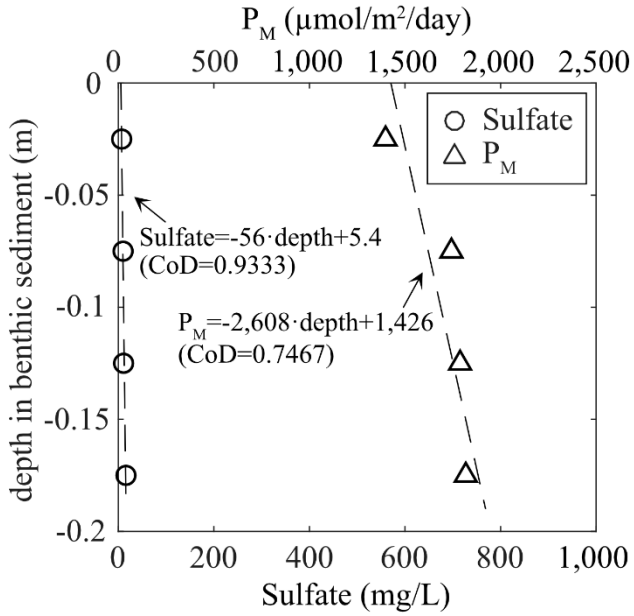


Fig. 6 Sulfate and dissolved methane concentrations across the first to fourth layers at St. I on August 7, 2013. Circles and triangles indicate sulfate concentrations and methane production, respectively.

Comment E-2

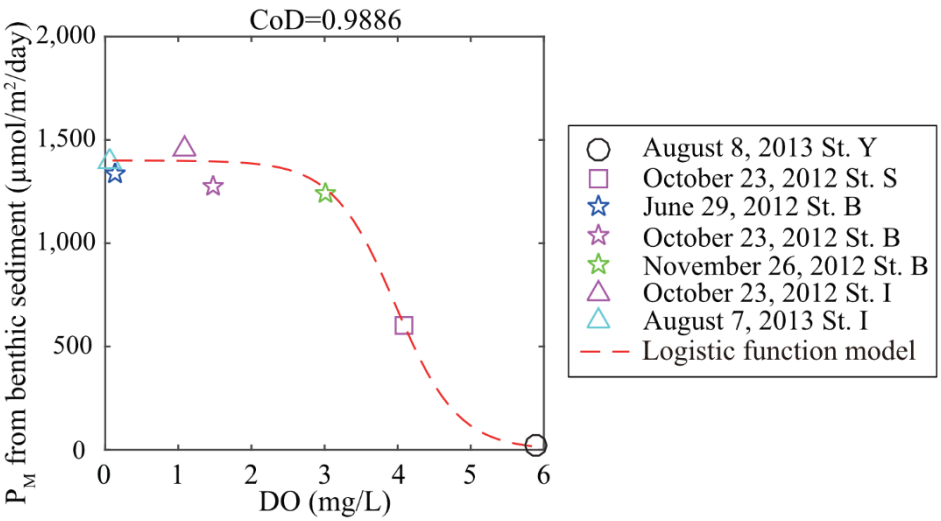
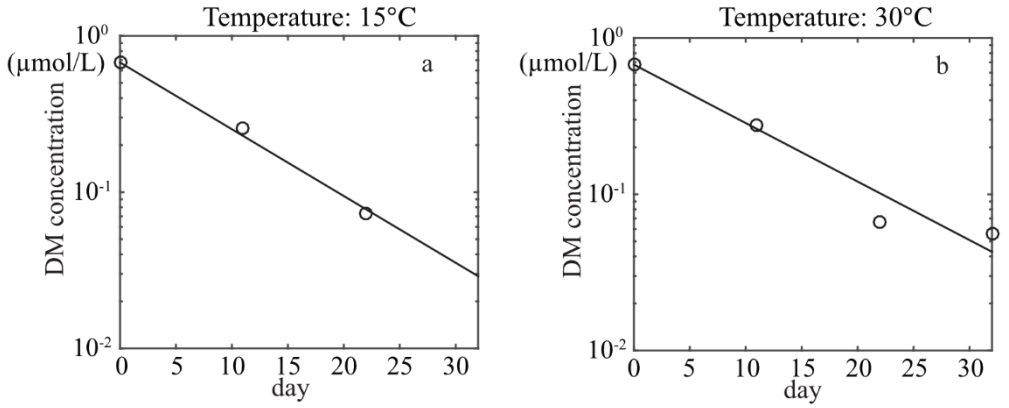


Fig. 7 Dissolved oxygen concentrations in the overlying water and methane production from the bottom sediment. Red broken lines indicate the logistic function, Eq. (6).



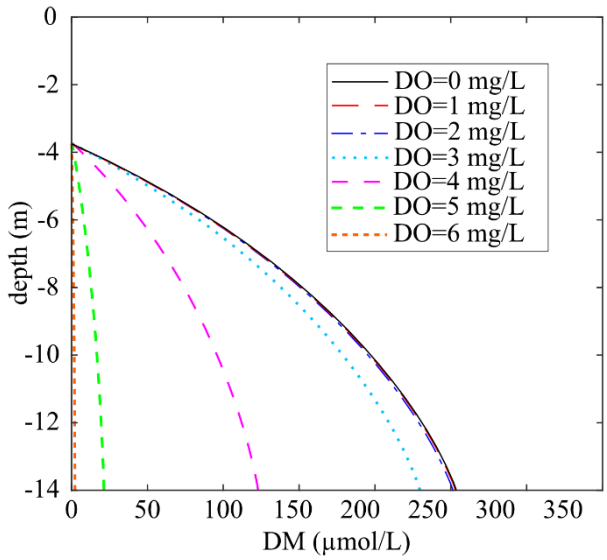
Comment E-2

692

693 **Fig. 8** Oxidation-reduction of dissolved methane concentrations under aerobic conditions. a:

694 Water temperature of 15℃. b: Water temperature of 30℃.

695



Comment E-2

696

697 **Fig. 9** The vertical profile of dissolved methane concentrations when the dissolved oxygen

698 concentration was calculated using one-dimensional numerical simulations varies from 0

699 mg/L to 6 mg/L.

Comment E-2

700

701

Appendix A. Supplementary data

Effect of dissolved oxygen on methane production from bottom sediment in a eutrophic stratified lake

Yasuyuki Maruya[1,*], Keisuke Nakayama[2], Masafumi Sasaki[3], Katsuaki Komai[3]

1. Graduate School of Engineering, Kyushu University, 744 Motooka, Nishi-Ku, Fukuoka 819-0395, Japan. E-mail: ysyk.mry@gmail.com
2. Graduate School of Engineering, Kobe University, Rokkodai-Cho 1-1, Nada-Ku, Kobe 657-8501, Japan
3. Department of Engineering, Kitami Institute of Technology, Kohen-Cho 165, Kitami 090-8507, Japan

Received October 26, 2021
Revised January 11, 2022
Accepted

---

* Corresponding author. E-mail: ysyk.mry@gmail.com (Yasuyuki Maruya)

Contents of this file

Fig. S1

Introduction

The Supplementary Information (SI) includes the following: **Fig. S1**, which provides an image of the non-disturbing core sampler used to sample anaerobic sediments as described in the main body of the manuscript.

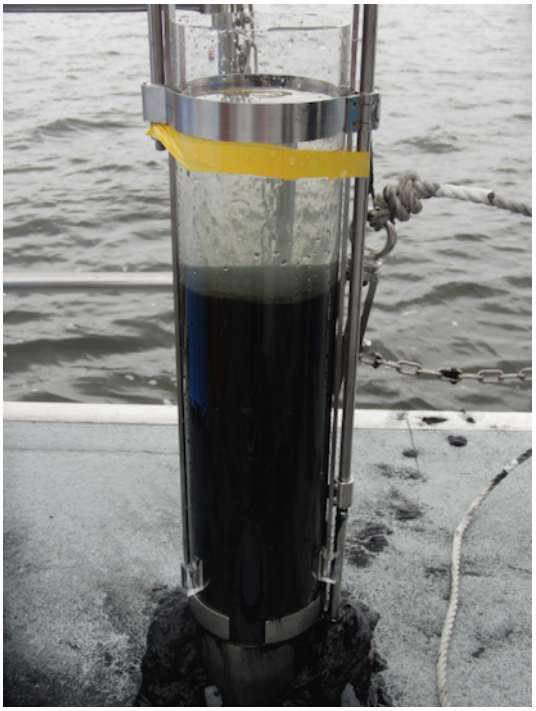


Fig. S1 Undisturbed core sampler.

Appendix A. Supplementary data

Effect of dissolved oxygen on methane production from bottom sediment in a eutrophic stratified lake

Yasuyuki Maruya[1,*], Keisuke Nakayama[2], Masafumi Sasaki[3], Katsuaki Komai[3]

1. Graduate School of Engineering, Kyushu University, 744 Motooka, Nishi-Ku, Fukuoka 819-0395, Japan. E-mail: ysyk.mry@gmail.com
2. Graduate School of Engineering, Kobe University, Rokkodai-Cho 1-1, Nada-Ku, Kobe 657-8501, Japan
3. Department of Engineering, Kitami Institute of Technology, Kohen-Cho 165, Kitami 090-8507, Japan

Received October 26, 2021
Revised January 11, 2022
Accepted

---

* Corresponding author. E-mail: ysyk.mry@gmail.com (Yasuyuki Maruya)

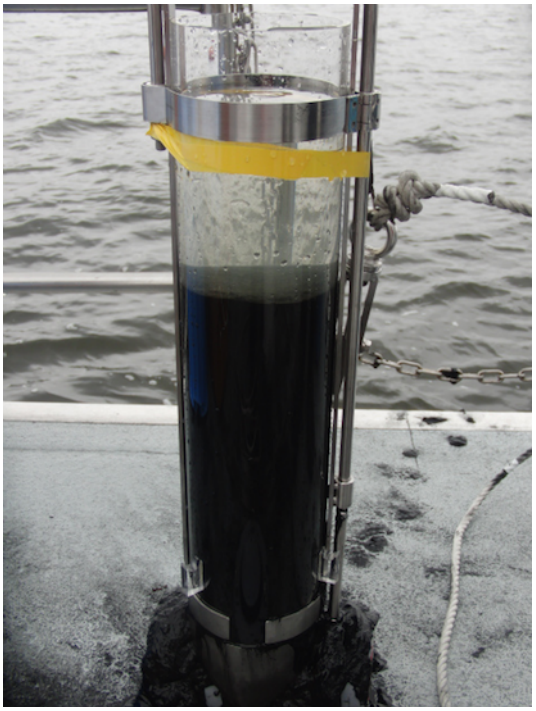
Contents of this file

Fig. S1

Introduction

The Supplementary Information (SI) includes the following: **Fig. S1**, which provides an image of the non-disturbing core sampler used to sample anaerobic sediments as described in the main body of the manuscript.

763



764 **Fig. S1** Undisturbed core sampler.

765