

**Supplementary Materials: “A new biogeochemical modelling framework (FLaMe v1.0) for lake methane emissions on the regional scale: Development and application to the European domain”**

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## 20 S1. Overview of the lake physical model–CLSM

21 The Canadian Small Lake Model (CSLM) is a 1D thermal model designed to simulate the water  
22 temperature profiles (T) and relevant variables (e.g., mixing depths ( $h_{\text{mix}}$ ) and ice cover dynamics)  
23 (MacKay, 2012; MacKay *et al.*, 2017). Here, we introduce the CSLM briefly, and the detailed  
24 mathematical descriptions, numerical schemes, and parameterizations of CSLM can be found in  
25 MacKay (2012). The CSLM computes the profiles of water temperature and light penetration, mixing  
26 depths, and ice dynamics, which set the basis for the simulation of the biogeochemical modules  
27 (organic carbon, oxygen, and methane) implemented in the FLaMe model. CSLM adopts a turbulent  
28 mixing parameterization, which relies on a depth-integrated turbulent kinetic energy (TKE) approach  
29 (MacKay 2012).

30 The meteorological variables required to force CSLM include shortwave and longwave radiation,  
31 precipitation, atmospheric temperature and pressure, near-surface wind speed, and specific humidity.  
32 The general heat transfer process (without turbulence and mixing layer) within the lake is described  
33 as follows:

34 
$$\frac{\partial T}{\partial t} = -\frac{1}{\rho c_w} \frac{\partial F}{\partial z} - \frac{1}{\rho c_w} \frac{\partial Q}{\partial z} \quad (1)$$

35 
$$F(z) = -K \frac{\partial T}{\partial z} \quad (2)$$

36 where  $T$  is the temperature of the lake at a given depth  $z$  and time  $t$ ,  $\rho$  is the water density,  $c_w$  is the  
37 water specific heat capacity,  $F$  is the heat flux resulting from thermal conduction,  $K$  is the water  
38 thermal conductivity, and  $Q$  is the radiative energy flux, which decreases with depth according to  
39 Beer's law. This equation is solved numerically using an explicit forward finite difference scheme in  
40 time.

41 To constrain the temperature at the lake surface, the surface energy balance is described as  
 42 follows:

$$43 \quad \frac{\partial T_0}{\partial t} = -\frac{l}{\delta_0 \rho c_w} [F_0 - (L^* - H_S - H_E) + Q_0 - Q^*] \quad (3)$$

44 where  $T_0$  is the surface layer temperature, also called “skin” temperature,  $\delta_0$  is the thickness of the  
 45 skin,  $F_0$  and  $Q_0$  are thermal and solar energy flux, respectively, transmitted from the skin to the water  
 46 column.  $L^*$  is the surface net longwave radiation,  $H_S$  and  $H_E$  are the turbulent sensible and latent heat  
 47 fluxes into the atmosphere, respectively.  $Q^*$  is the surface net solar radiation that penetrates the lake  
 48 water. This description allows ice formation when  $T_0 < 0^\circ\text{C}$ . The details of the parameterization of  
 49 this equation can be found in MacKay (2012).

50 If we consider the turbulence and resultant mixed layer, the depth-integrated turbulent kinetic  
 51 energy (TKE) approach can be described briefly as follows. Driven by wind, the upper portion of the  
 52 lake water is mixed by TKE, leading to a surface mixed layer. Within this mixed layer, the TKE is  
 53 dynamically calculated as follows:

$$54 \quad \frac{d}{dt} (u h_{mix}) = u_*^2 \quad (4)$$

$$55 \quad \frac{d}{dt} \left( \frac{l}{2} h_{mix} E_s \right) = \frac{h_{mix}}{2} \frac{dE_s}{dt} + \frac{E_s}{2} \frac{dh_{mix}}{dt} \quad (5)$$

56 where  $u$  is the mean horizontal velocity within the mixed layer,  $h_{mix}$  is the depth at the bottom of the  
 57 mixed layer,  $u_*$  is the surface friction velocity induced by near-surface wind, and  $E_s$  is the average  
 58 TKE per unit mass in the mixed layer. Equations (4) and (5) specifies the momentum balance and the  
 59 depth-integrated TKE budget, respectively, within the mixed layer.

60 The surface friction velocity  $u_*$  is estimated from the atmospheric near-surface wind speed  $U_a$  as

61 follows:

62 
$$u_* = \frac{C_D \rho_a^{1/2}}{\rho} U_a \quad (6)$$

63 where  $C_D$  is the drag coefficient and  $\rho_a$  is the air density.

64 To constrain  $E_S$  in Eq. (5), the energy budget within the mixed layer is described as follows:

65 
$$\frac{d}{dt} \left( \frac{l}{2} h_{mix} E_s \right) = F_q - F_d + F_s - F_p - F_L \quad (7)$$

66 where  $F_q$  is the mechanical energy caused by wind and buoyancy effects,  $F_s$  is the shear production  
67 flux caused by the stabilization effect of the thermocline at the bottom of the mixed layer,  $F_d$  is the  
68 energy dissipation flux within the mixed layer,  $F_i$  is the transport of TKE to the thermocline,  $F_L$  is the  
69 TKE leakage flux caused by internal wave propagation away from the mixed layer, and  $F_p$  is the  
70 change in potential energy flux caused by the change in thermocline depth. To close the system of  
71 equations, the energy balance within the mixed layer can be separated into two components:

72 
$$\frac{h_{mix}}{2} \frac{dE_s}{dt} = F_q - F_d - F_i \quad (8)$$

73 
$$\frac{E_s}{2} \frac{dh_{mix}}{dt} = F_i + F_s - F_p - F_L \quad (9)$$

74 These two equations can be used to solve the dynamics of  $E_S$  and  $h_{mix}$ , respectively, which rely  
75 only on the model parameters and meteorological forcings (see details in MacKay 2012).

## 76 S2. Methods for estimating the threshold depth for the split of diffusion and ebullition

77 Following Langenegger *et al.* (2019), we assumed an exponentially decreasing function to  
 78 describe the decay of methane production rate versus sediment depth, and adopted a threshold depth  
 79  $z_{eb,min}$  to split the CH<sub>4</sub> production into diffusive and ebullitive transport pathway. Driven by the CH<sub>4</sub>  
 80 production, the CH<sub>4</sub> concentration increases as the sediment depth increases. Therefore, at a certain  
 81 threshold sediment depth, the CH<sub>4</sub> concentration may exceed its solubility limit and bubbles may start  
 82 forming (Fig. 3 in the main text). This threshold depth,  $z_{eb,min}$ , is the depth at which the sum of partial  
 83 pressure of CH<sub>4</sub> and N<sub>2</sub> exceed the sum of the hydrostatic and atmospheric pressure. Following  
 84 Langenegger *et al.*, (2019),  $z_{eb,min}$  is determined by the following equation:

$$85 \quad \underbrace{K_{H,CH_4}[CH_4]_s + K_{H,N_2}[N_2]_s}_{\text{total dissolved gas pressure}} = \underbrace{\rho gh}_{\text{hydrostatic pressure}} + \underbrace{P_{atm}}_{\text{atmosphere pressure}} - \underbrace{P_{H_2O}}_{\text{water vapor saturation pressure}} \quad (10)$$

86 where  $K_{H,CH_4}$  and  $K_{H,N_2}$  are Henry's constants of CH<sub>4</sub> and N<sub>2</sub>, respectively. The dissolved gas  
 87 pressure of CH<sub>4</sub> ( $[CH_4]_s$ ) is determined by the CH<sub>4</sub> production rate and its exponentially decreasing  
 88 distribution versus sediment depth  $z_s$ , while the dissolved gas pressure of N<sub>2</sub> ( $[N_2]_s$ ) is assumed to  
 89 be constant at  $0.78P_{atm}$ . Using a zero-flux boundary condition at the bottom of the sediment and a  
 90 constant CH<sub>4</sub> concentration equal to that in the overlying water layer at the sediment-water interface,  
 91 the threshold depth  $z_{eb,min}$  can then be solved numerically with the CH<sub>4</sub> concentration profile versus  
 92 depth in the sediment:

$$93 \quad 0 = 1 - z_{eb,min} \alpha \exp(-z_{eb,min} \alpha) - \exp(-z_{eb,min} \alpha) - \alpha^2 \frac{k_{diff,CH_4}}{F_{Met,0} K_{H,CH_4}} P_{res} \quad (11)$$

94 where  $P_{res} = \rho gh + P_{atm} - P_{H_2O} - 0.78P_{atm} - K_{H,CH_4}[CH_4]$ , and  $k_{diff,CH_4}$  is the effective molecular  
 95 diffusion of CH<sub>4</sub>.  $\alpha$  is the shape parameter specifying the decay rate of CH<sub>4</sub> production rate versus  
 96 sediment depth, and  $F_{Met,0}$  is the maximum CH<sub>4</sub> production rate at the sediment-water interface. More

97 details of the derivation of the above equations can be found in Langenegger *et al.* (2019).

### 98 **S3. Two representative lakes: Deep oligotrophic lake in a cold climate vs. shallow eutrophic** 99 **lake in a warm climate**

100 The model set-ups for these two representative lakes (a deep oligotrophic lake in a cold climate  
101 vs. a shallow eutrophic lake in a warm climate) are described in section 2.5.1, with their main results  
102 summarized in section 3.1. In this section, we introduce further details of numerical simulations  
103 related to these two representative lakes, i.e., the key physical–biogeochemical patterns as well as the  
104 seasonality of CH<sub>4</sub> productions and emissions.

#### 105 **S3.1 Physical–biogeochemical patterns of these two lakes**

106 Here, we first illustrate the time series of mean water temperature ( $T_{mean}$ ), mixing depth ( $z_{mix}$ ),  
107 depth integrated primary production ( $F_{PP}$ ) and mineralization ( $F_{Min}$ ) as well as the labile carbon  
108 concentration ( $C_{OC,auto}$ ) (Fig. 5 in the main text). For the deep oligotrophic lake,  $T_{mean}$  varies within a  
109 narrow range of  $\sim 3\text{--}8^\circ\text{C}$  across seasons, while  $z_{mix}$  has a much larger variability comprised between  
110 0.5m and 35m (i.e.,  $h_{max}$ ) (Fig. 5a in the main text).  $F_{PP}$  has a low yearly mean of  $\sim 13 \text{ gC m}^{-2} \text{ yr}^{-1}$ ,  
111 with peaks during the productive season about twice as large.  $F_{Min}$  essentially follows the temporal  
112 evolution of  $F_{PP}$ , except with  $\sim 3$  times lower peaks; however, the mineralization can also be higher  
113 than  $F_{PP}$  as high rates may persist for a longer duration (Fig. 5c in the main). As a result, the yearly  
114 mean of  $F_{min}$  amounts to  $8 \text{ gC m}^{-2} \text{ yr}^{-1}$ , with the difference between  $F_{PP}$  and  $F_{min}$  sustaining the burial  
115 flux and a weak accumulation in  $C_{OC,auto}$  (Fig. 5e in the main). As a result of the dynamic interplay  
116 between  $F_{PP}$  and  $F_{Min}$ , the vertically homogeneous  $C_{OC,auto}$  concentrations are comprised between  
117  $0.04 \text{ gC m}^{-3}$  and  $0.41 \text{ gC m}^{-3}$ , with a yearly mean of  $0.19 \text{ gC m}^{-3}$  (Fig. 5e in the main text).

118 For the shallow eutrophic lake,  $T_{mean}$  is characterized by much larger variations, ranging from  $5^\circ$   
119 C in winter to  $\sim 15^\circ\text{C}$  in summer, while in the mixed depth  $z_{mix}$  has a narrower range between 2 m in

120 warm seasons and 10 m in cold seasons (i.e.,  $h_{max}$ ) (Fig. 5b in the main).  $F_{PP}$  has a much higher yearly  
121 mean than the deep oligotrophic lake, reaching up to  $490 \text{ gC m}^{-2} \text{ yr}^{-1}$ , with peaks even exceeding 1000  
122  $\text{gC m}^{-2} \text{ yr}^{-1}$  (Fig. 5d in the main text).  $F_{Min}$  also follows the temporal evolution of  $F_{PP}$ , with the yearly  
123 mean reaching 62% of  $F_{PP}$ . The dynamic balance between production and mineralization lead to  
124  $C_{OC,auto}$  concentration comprised between about 10 and  $33 \text{ gC m}^{-3}$  and a yearly mean of  $21 \text{ gC m}^{-3}$   
125 (Fig. 5f in the main text).

126 Unsurprisingly, in the shallow eutrophic lake,  $T_{mean}$  responds more quickly to variations in  $T_a$   
127 than in the deep lake because of a lower water volume. The dynamics of  $z_{mix}$  is more complex and  
128 requires investigating the vertical distribution of the temperature and density profiles (see below). In  
129 terms of biogeochemistry, the values of  $F_{PP}$  and  $C_{OC,auto}$  simulated here are broadly consistent with  
130 those reported in the literature for lakes of similar characteristics (Wetzel, 2001). In shallow eutrophic  
131 lakes, the much higher  $F_{PP}$  and  $C_{OC,auto}$  values are not only triggered by the higher phosphorus loads,  
132 but also by higher water temperatures, despite this latter factor also increasing  $F_{min}$ . Overall, the  
133 maximum  $C_{OC,auto}$  concentrations in the eutrophic lake are about a factor of 100 larger than that in the  
134 oligotrophic lake, and these differences are instrumental in controlling the  $\text{CH}_4$  production dynamics.  
135 The periods of significant  $C_{OC,auto}$  densities ( $15 \text{ gC m}^{-2}$ ) are also substantially longer in the eutrophic  
136 lake than in the oligotrophic lake.

137 In addition, FLaMe also reproduces observed vertical profiles of  $T$ ,  $[\text{O}_2]$  and  $[\text{CH}_4]$  well (Fig. S9).  
138 In terms of physics and for both representative cases, as the temperature in the first water layer  
139 approaches  $4^\circ\text{C}$ , the thermocline (black curve in top panels) deepens sharply towards the lake bottom,  
140 effectively mixing the entire water column. In the deep oligotrophic lake (Fig. S9a, 9c, and 9e),  
141 stratification is almost permanently maintained and is only interrupted each year by short but intense



142 turnover events during late falls. In Winter, the stratification is particularly stable due to a long  
143 occurrence (six months) of ice cover; while in Summer, the stratification is gradually weakened as  
144 the warming of lake water occurs. The lake turnover events, by inducing full mixing across the entire  
145 water column, are key control factors of the spatio-temporal dynamics in O<sub>2</sub> and CH<sub>4</sub>. Indeed, during  
146 lake turnovers, [O<sub>2</sub>] is effectively mixed across all water layers and reach close to saturation  
147 concentrations with the atmosphere (14.6 and 9.1 g O<sub>2</sub> m<sup>-3</sup> at a 0 and 20 °C, respectively). During the  
148 summer, slight oversaturation occurs in the upper portion of the lake as a result of photosynthetic  
149 activity, followed by a minimum in both the upper and deeper portions of the lake in the late  
150 summer/fall, when the mineralization rate ( $F_{Min}$ ) is higher than the production rate ( $F_{PP}$ ) and the  
151 oxygen diffusion from atmosphere cannot compensate oxygen net consumption. Except for this latter  
152 period, the combined effects of photosynthesis and transport from atmosphere via turnover events  
153 and eddy diffusion generally lead to well oxygenated conditions during most of the year, and the O<sub>2</sub>  
154 concentration is largely dominated by the solubility, itself controlled by the water temperature. That  
155 is, lake physics plays a dominant role in controlling the biogeochemical variables. Due to the  
156 oligotrophic status and well oxygenated conditions, the CH<sub>4</sub> concentrations are overall extremely low  
157 except near the lake bottom during the late summer/fall transition.

158 In the shallow eutrophic lake (Fig. S9b, 9d and 9f), there is no ice cover during the whole year  
159 due to a warmer climate, and the water temperature can adjust more quickly to the variations in  
160 atmospheric temperature due to lower lake depth. Thus, the stratification is weaker than in the deep  
161 oligotrophic lake, resulting in a less pronounced vertical gradients of water temperature. Similar to  
162 the deep oligotrophic lake, during summer and in the upper portion of the lake, the O<sub>2</sub> concentration  
163 are slightly supersaturated due to photosynthesis, followed by gradual decreases in O<sub>2</sub> concentration

164 as the  $F_{min}$  exceeds the  $F_{PP}$ . Due to the high production under eutrophic status, significant amounts of  
165 organic matter are exported to the lower portion of the lake (below thermocline), consuming  
166 progressively all of the  $O_2$  and leading to the development of an anaerobic zone in the hypolimnion.  
167 That is, the vertical  $O_2$  profile is controlled not only by the lake physics (temperature and  $O_2$  solubility;  
168 as in the case of the deep oligotrophic lake) but also by the intense biogeochemical processes. As a  
169 result, the combined effects of high  $F_{Min}$  and low  $O_2$  concentrations lead to maximal  $CH_4$   
170 concentrations ( $3.0 \text{ g } CH_4 \text{ m}^{-3}$ ) in late summer at the bottom of the lake, which are about 600-fold  
171 higher than that in the deep oligotrophic lake. In early Fall, the turnover events can bring oxygen  
172 down to lake bottom and abruptly interrupt the anaerobic condition for  $CH_4$  production.

### 173 **S3.2 Seasonality of $CH_4$ productions and emissions in these two lakes**

174 The comparison of the seasonality in  $CH_4$  productions and emissions for these two representative  
175 lakes is shown in Fig. S10. In the deep oligotrophic lake, the sedimentary  $CH_4$  production ranges  
176 from  $8.0 \times 10^{-5}$  to  $8.0 \times 10^{-3} \text{ g } CH_4 \text{ m}^{-2} \text{ d}^{-1}$ , with the highest values in August and the lowest values in  
177 Winter and Spring. Due to the low production rate and high-water pressure, bubble formation is very  
178 limited and almost all the produced  $CH_4$  escapes the sediment via diffusion. Since the oxygen  
179 availability is high throughout the year and the transport distance is long for the non-littoral zone of  
180 the lake, the diffusive  $CH_4$  flux is almost completely oxidized before reaching the atmosphere.  
181 Moreover, the ice cover during winter and early spring (December–April) blocks the  $CH_4$  emissions,  
182 such that all  $CH_4$  emissions occur in the other seven months (May–November). As a result, the total  
183  $CH_4$  emissions range from 0 to  $2.4 \times 10^{-4} \text{ g } CH_4 \text{ m}^{-2} \text{ d}^{-1}$  with a tiny peak in October, which results from  
184 lake turnovers and the additional contribution of the storage flux to diffusion.

185

186 In the shallow eutrophic lake, the CH<sub>4</sub> production is >1000 times higher than that in deep  
187 oligotrophic lake, ranging from 0.02 to 0.35 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>. This much higher production is attributed  
188 to the higher nutrient loads, higher irradiance and higher temperature during summer which trigger  
189 higher  $F_{PP}$ ,  $COC_{auto}$ ,  $F_{Min}$ , and  $F_{Met}$  (Fig. 5 in the main text), and thus, higher  $F_{Met}$  under a lower O<sub>2</sub>  
190 concentration. This high CH<sub>4</sub> production not only favors CH<sub>4</sub> emissions crossing the sediment-water  
191 interface through the diffusive pathway ( $F_{s,diff}$ ) but also through the ebullitive pathway ( $F_{s,ebul}$ ).  
192 Compared to the deep oligotrophic lake, the shallow eutrophic lake also has weaker stratification,  
193 shorter transport distance from sediment to atmosphere (in non-littoral zones), and therefore a lower  
194 fraction of diffusive CH<sub>4</sub> is oxidized, leading to ~900 times higher total CH<sub>4</sub> emissions. Moreover,  
195 the total emissions have a seasonal pattern similar to that of production, with highest (0.21 g CH<sub>4</sub> m<sup>-2</sup>  
196 d<sup>-1</sup>) and lowest (0.02 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) emissions in September and February, respectively. Due to the  
197 warm climate, the water temperature is too high to form ice on the lake surface, leading to emissions  
198 during the whole year, about twice longer than for the deep oligotrophic lake under cold climate.  
199 Overall, with these two representative lakes, we show that FLame can well capture the wide range of  
200 CH<sub>4</sub> emissions (0–0.3 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) from oligotrophic to eutrophic lakes in real world, as  
201 demonstrated by the cases in Tan *et al.* (2024).

## S4. Effects of trophic status, lake depths, climate conditions on seasonal CH<sub>4</sub> dynamics

In this section, we illustrate the responses of CH<sub>4</sub> dynamics to variations in environmental gradients and lake morphology, hence decomposing the collective effects of these environmental factors analyzed in section S3). First, we set as baseline case a mesotrophic lake with  $h_{max} = 15$  m (and  $h_{mean} = 7.5$  m), [TP]=30  $\mu\text{gP L}^{-1}$ , forced by a temperate climate (48.75°N, 8.75°E) (Fig. S3). Based on this baseline, we then explore the responses of the CH<sub>4</sub> dynamics induced by individual effects of trophic status, climate condition and lake depth, by varying one parameter at a time: (1) increasing the [TP] from 8 to 80  $\mu\text{g P L}^{-1}$ ; and (2) changing the climate from warm (43.75°N, -6.25°E; Fig. S1) to cold conditions (63.75°N, 26.25°E; Fig. S2); (3) changing the maximal lake depth ( $h_{max}$ ) from 10 to 35 m;.

### S4.1 Baseline of lake CH<sub>4</sub> seasonality

In this baseline (Fig. S11), the CH<sub>4</sub> production in lake sediment has its highest rates (0.08 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) in September and lowest rates (0.003 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) in March. The seasonal patterns of CH<sub>4</sub> emissions follows those of CH<sub>4</sub> production, but they are interrupted by the storage flux induced by the weakened stratification and turnover during late fall. As a result, the CH<sub>4</sub> emissions are highest (0.04 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) in November and lowest (0.001 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) in March. Overall, annual mean CH<sub>4</sub> production and emission are 0.037 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>, and 0.019 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>, respectively, with the peaks of total CH<sub>4</sub> production and emission about 3 and 2 times higher than their means. The magnitudes of CH<sub>4</sub> emissions in this mesotrophic lake are very close to the cases (Erssjön and Fei-Tsui) as demonstrated by Tan *et al.* (2024).

### S4.2 Effects of trophic status on CH<sub>4</sub> seasonality

Under different trophic status, the total CH<sub>4</sub> productions have similar seasonality (Fig. S12) as

224 that of the baseline case (Fig. S11), with peaks and valleys of CH<sub>4</sub> production in August and February,  
225 respectively. From oligotrophic to trophic status ([TDP] is increased by 10 times), the peak of CH<sub>4</sub>  
226 production grows by a factor of 18 (from 0.013 to 0.24 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>), and the mean of CH<sub>4</sub> production  
227 is increased by a factor of 30 (from 0.003 to 0.089 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>). This nonlinear response of CH<sub>4</sub>  
228 production to [TDP] can be explained by the effect of P limitation on  $F_{PP}$  (see Eq. (3) in the main) (6  
229 times, from 0.08 to 0.47 from oligotrophic to eutrophic status), as well as the oxygen control on  
230 benthic CH<sub>4</sub> production, i.e., the bottom oxygen concentration is much lower and the anoxic condition  
231 persists for a longer time in the eutrophic lake (Fig. S16a vs. 16b). In the oligotrophic lake, the peak  
232 of CH<sub>4</sub> emission occurs in September, indicating a one-month time lag compared to the production,  
233 which is attributed to the lake stratification and inhibited transport from lake bottom to surface. The  
234 blocking effect of lake stratification on CH<sub>4</sub> emissions is more evident when the CH<sub>4</sub> production is  
235 higher in the eutrophic lake, postponing the peak of CH<sub>4</sub> emissions to November.

236 As shown in Fig. S12, the ratio of ebullition to diffusion from sediments is also sensitive to the  
237 trophic status, suggesting a higher (78%) fraction of diffusion for the oligotrophic case compared to  
238 the eutrophic case (59%). This is expected because, due to higher production and lower oxygen  
239 compared to the oligotrophic case, the CH<sub>4</sub> production is much higher in the eutrophic case, thus  
240 promoting much higher CH<sub>4</sub> supersaturation and bubble production (indicated by lower  $z_{eb,min}$ ). The  
241 oligotrophic and eutrophic lakes have similar seasonal patterns of CH<sub>4</sub> emissions except the high  
242 storage flux occurred in eutrophic case. The oligotrophic lake has slightly higher shares (60% vs.  
243 56%) of ebullition to atmosphere highlighting the more efficient CH<sub>4</sub> oxidation in the water column  
244 for the oligotrophic lake, as revealed in Fig. S16.

### 245 S4.3 Effects of climate conditions on CH<sub>4</sub> seasonality

246 The climate condition changes not only the magnitudes of CH<sub>4</sub> production and emission but also  
247 the temporal patterns or seasonality (Fig. S13). From cold to warm climate, the peak of CH<sub>4</sub>  
248 production grows by a factor of 4 (0.04 to 0.17 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>), and the mean of CH<sub>4</sub> production grows  
249 by a factor of 6 (0.0094 to 0.059 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>), which is mainly attributed to the higher solar radiation  
250 and temperature (Fig. S1 and Fig. S2). Across this climate gradient, both the peak and mean of CH<sub>4</sub>  
251 emissions grow by a factor of ~4 (from 0.02 to 0.07 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> and from 0.0057 to 0.03 g CH<sub>4</sub> m<sup>-2</sup>  
252 d<sup>-1</sup>). This implies that a larger fraction of CH<sub>4</sub> production is oxidized in the warmer lake, due to  
253 higher water temperature (Fig. S15). Climate also controls the ratio of ebullition to diffusion in  
254 production, with the ebullitive pathway being favored by higher temperature in warmer climate (42%  
255 versus 36%) (through lower gas solubility and higher methanogenesis). After accounting for CH<sub>4</sub>  
256 oxidations during diffusion, the fraction of ebullition in CH<sub>4</sub> emissions is still higher in warmer  
257 climate (78% vs. 65%), probably due to the higher CH<sub>4</sub> oxidation rate during the diffusive transport  
258 in water column in the warmer lake.

259 Driven by a cold climate in the boreal region (Fig. S13a), the seasonality of CH<sub>4</sub> production and  
260 emissions are highly affected by the ice cover in winter and spring when the methanogenesis is  
261 strongly reduced due to low temperature and emissions are blocked by the ice. The CH<sub>4</sub> production  
262 starts to increase from June, reaches its maximum in August, and returns to a low level in December.  
263 The CH<sub>4</sub> emissions follow the seasonal pattern of production but is interrupted by the storage flux  
264 induced by lake stratification, leading to a peak in October. With a warmer climate (located in the  
265 temperate region) (Fig. S13b), both CH<sub>4</sub> productions and emissions have later peaks (September and  
266 November, respectively). In the warmer lake, the contribution of the storage flux is more evident,

probably due to higher CH<sub>4</sub> production during summer coinciding with the period of strong lake stratification.

#### S4.4 Effects of lake depth on CH<sub>4</sub> seasonality

By comparing Fig. S14b and Fig. S11, we found that the lakes with a maximum depth of 15 m and 35 m have almost the same CH<sub>4</sub> production rate (0.03 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> for the mean and 0.08 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> for the peak), consistent with our common knowledge and the inferences from formulas of depth-integrated  $F_{PP}$ ,  $F_{Min}$ , and  $F_{Met}$  (Eqs. (3), (8), and (9) in the main). However, the lake with a shallower depth ( $h_{max} = 10$  m) has a slightly lower CH<sub>4</sub> production rate (0.025 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> for the mean and 0.07 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> for the peak). This is attributed to the photic zone being truncated by morphological constraints (i.e., the lakebed is shallower than the “natural” photic depth), which decreases the depth-integrated primary production and further the mineralization and CH<sub>4</sub> production (note that our model does not account for benthic algal production). Lake depth has a more evident effect on the fraction of sedimentary CH<sub>4</sub> production channeled into the ebullitive pathway (52% versus 28%), because this pathway is favored in a shallower lake with lower CH<sub>4</sub> solubility due to higher temperature and lower hydrostatic pressure.

Consistent with our common knowledge, the CH<sub>4</sub> emissions are overall lower for a deeper lake, due to the higher CH<sub>4</sub> solubility, efficient oxidation during the diffusive transport of CH<sub>4</sub> through longer distance, and larger dissolution of the ebullitive flux during the transport of gas bubbles. Interestingly, the lower CH<sub>4</sub> emissions in the deeper lake during the production season could be attributed to a stronger lake stratification, and this effect may also be reflected by the more evident storage flux in November, December and January, which partly offsets the lower emissions of the deeper lake in other months of the year. Except for these three months, the fraction of ebullitive

emissions is slightly higher in deeper lakes (87% vs. 84%), despite a lower fraction of ebullition in the production. However, if we take these three months into account, the fraction of ebullitive emissions in deeper lake becomes 64%, much lower than that in shallower lake (82%). This implies that the lake stratification also plays a crucial role in the regulating the emission pathways especially for deep lakes.

## **S5. Estimates of CH<sub>4</sub> emissions from large lakes ( $\geq 1000$ km<sup>2</sup>) within European domain**

To estimate CH<sub>4</sub> emissions from European lakes  $\geq 1000$  km<sup>2</sup>, we came up with two strategies as follows:

(1) We used FLaMe model directly to simulate lakes  $\geq 1000$  km<sup>2</sup> within the European domain and found that the CH<sub>4</sub> emissions from European lakes  $\geq 1000$  km<sup>2</sup> accounts for only 6% of those from smaller lakes, i.e., 0.06 Tg CH<sub>4</sub> yr<sup>-1</sup>. Thus, the European lakes have a total CH<sub>4</sub> emissions of 1.03 Tg CH<sub>4</sub> yr<sup>-1</sup>.

(2) Despite with very limited samples, Johnson *et al.* (2022) found that the total CH<sub>4</sub> emission (sum of diffusion and ebullition) rates per unit area from lakes  $\geq 5000$  km<sup>2</sup> are in the range of 0–25% of emissions from smaller lakes. Following Johnson *et al.* (2022), we assumed that the CH<sub>4</sub> emission rates per unit area from lakes  $\geq 1000$  km<sup>2</sup> falls in the higher end of 0–25% and adopt a value of 20% to estimate the CH<sub>4</sub> emission rates for lakes  $\geq 1000$  km<sup>2</sup>.

From our simulations for lakes smaller than  $<1000$  km<sup>2</sup>, we obtained that the mean CH<sub>4</sub> emission rate per unit lake area amounts to 7.39 g CH<sub>4</sub> m<sup>-2</sup> yr<sup>-1</sup>. Thus, the mean CH<sub>4</sub> emission rate per unit lake area for lakes larger than 1000 km<sup>2</sup> is estimated as  $7.39 \cdot 20\% = 1.48$  g CH<sub>4</sub> m<sup>-2</sup> yr<sup>-1</sup>. By multiplying this mean CH<sub>4</sub> emission rate (1.48 g CH<sub>4</sub> m<sup>-2</sup> yr<sup>-1</sup>) with the area of  $0.88 \cdot 10^5$  km<sup>2</sup> for lakes  $\geq 1000$  km<sup>2</sup>,



310 we obtain CH<sub>4</sub> emissions from large lakes as 0.13 Tg CH<sub>4</sub> yr<sup>-1</sup>, broadly similar to what is obtained  
311 with the first method. Thus, the European lake emissions reach in this case a total CH<sub>4</sub> of 1.10 Tg  
312 CH<sub>4</sub> yr<sup>-1</sup>.

313 Combining these two strategies, we provide a back of the envelope estimate for the total CH<sub>4</sub>  
314 emissions from European lakes as 1.03–1.10 Tg CH<sub>4</sub> yr<sup>-1</sup>, which can be directly compared to previous  
315 estimates.

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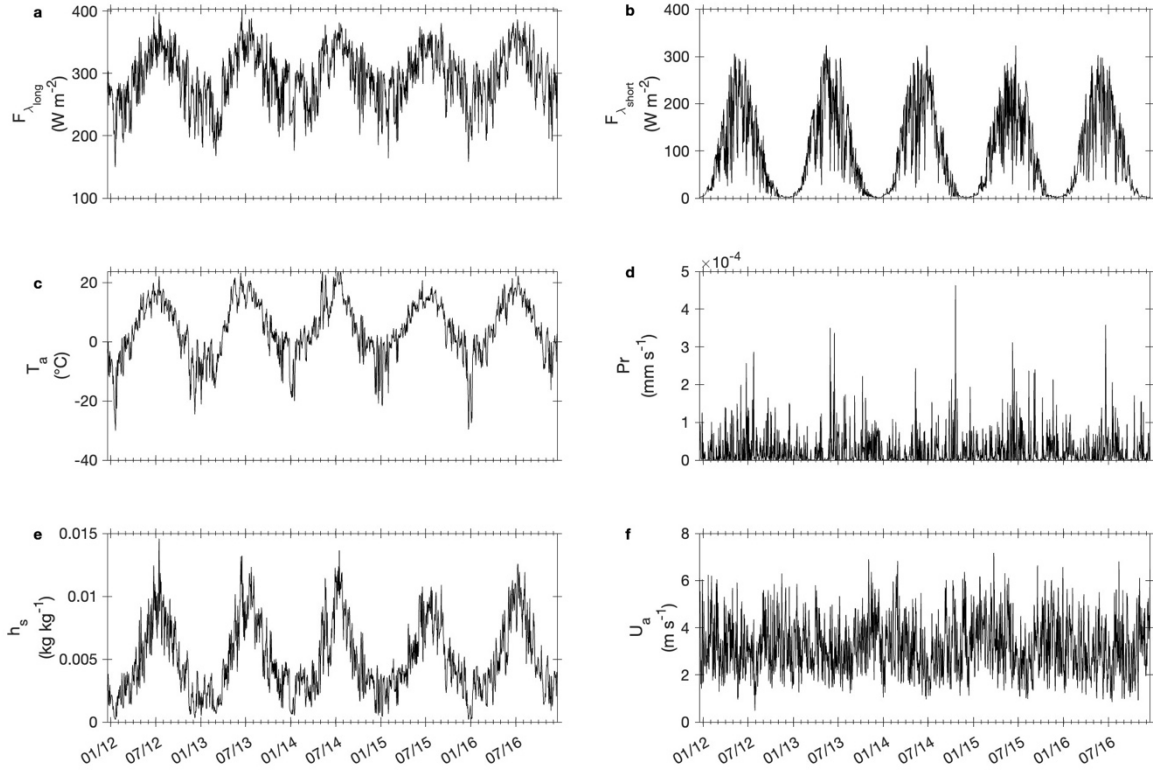
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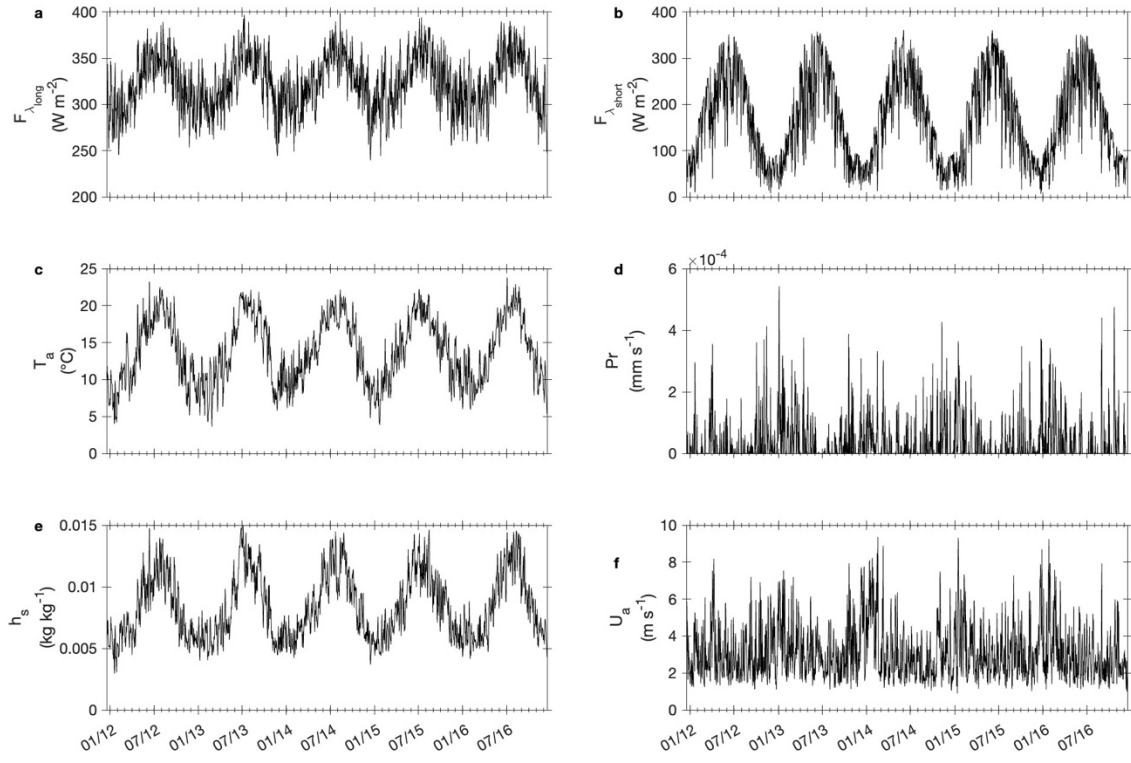
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**Fig. S1.** Meteorological data (2012-2016) for a cold climate ( $63.75^{\circ}\text{N}$ ,  $26.25^{\circ}\text{E}$ ) extracted from the ISIMIP3a. (a) long-wave radiation ( $F_{\lambda, \text{long}}$ ); (b) short-wave radiation ( $F_{\lambda, \text{short}}$ ) (c) atmospheric temperature near the surface ( $T_a$ ); (d) precipitation ( $P_r$ ); (e) specific humidity ( $h_s$ ); and (f) near-surface wind ( $U_a$ ).

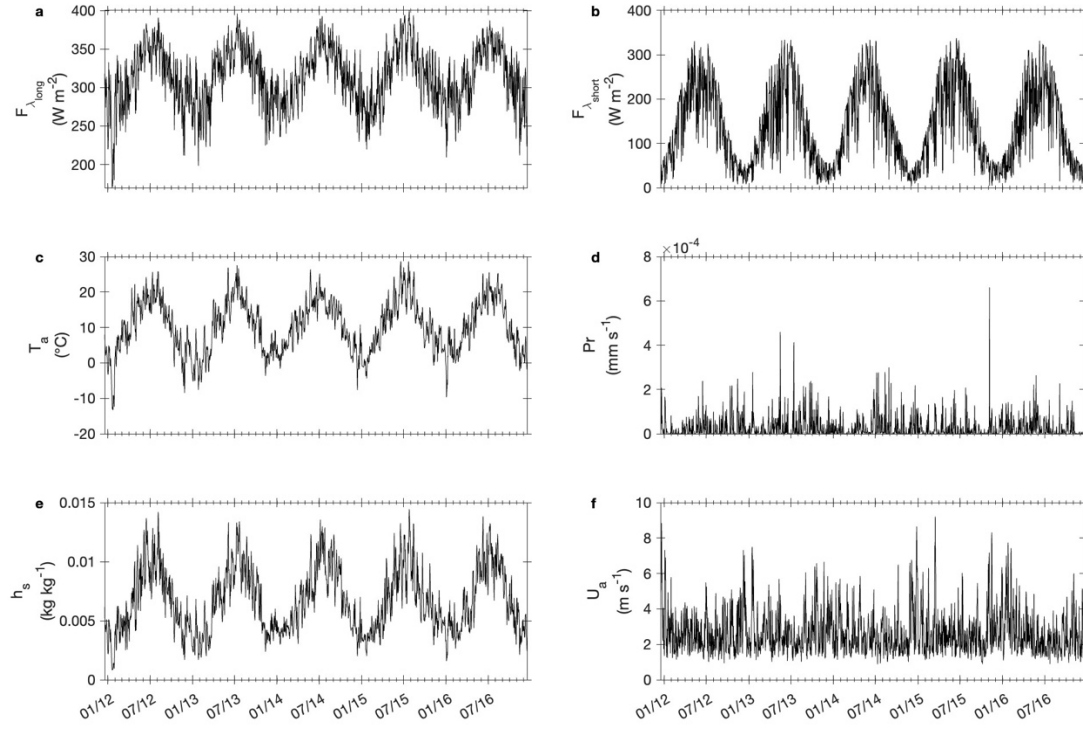


**Fig. S2.** Meteorological data (2012-2016) for a warm temperate climate ( $43.75^{\circ}N$ ,  $-6.25^{\circ}E$ ) extracted from the ISIMIP3a. (a) long-wave radiation ( $F_{\lambda, long}$ ); (b) short-wave radiation ( $F_{\lambda, short}$ ) (c) atmospheric temperature near the surface ( $T_a$ ); (d) precipitation ( $P_r$ ); (e) specific humidity ( $h_s$ ); and (f) near-surface wind ( $U_a$ ).

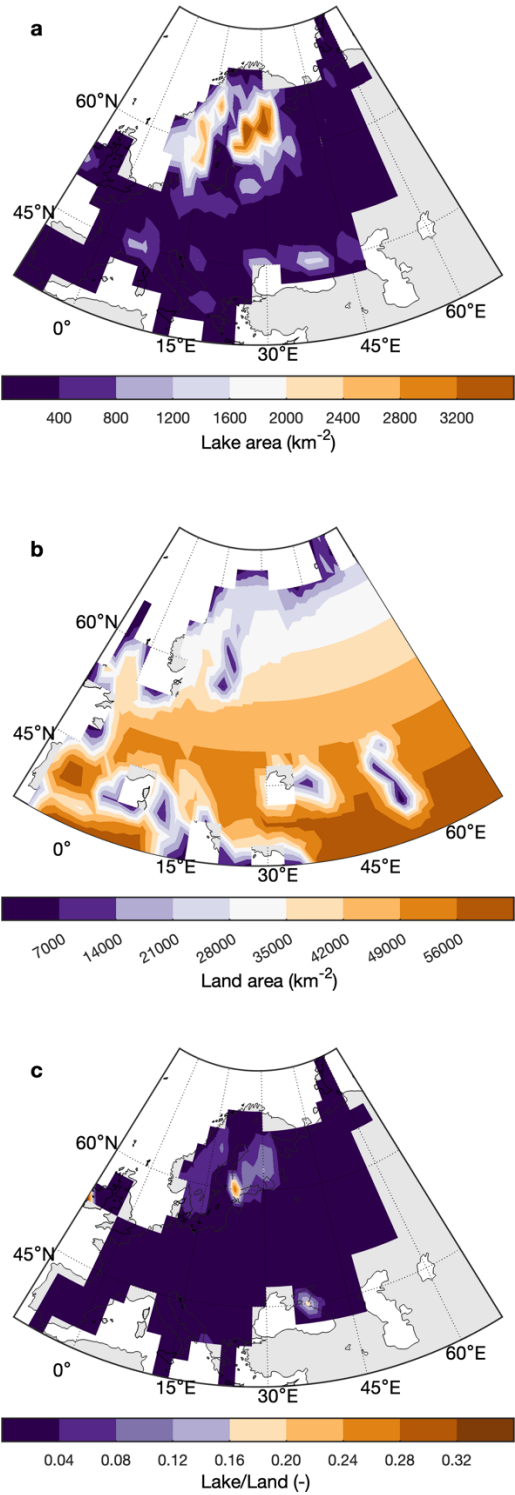


**Fig. S3.** Meteorological data (2012-2016) in Finland (48.75°N, 8.75°E) extracted from the ISIMIP3a.

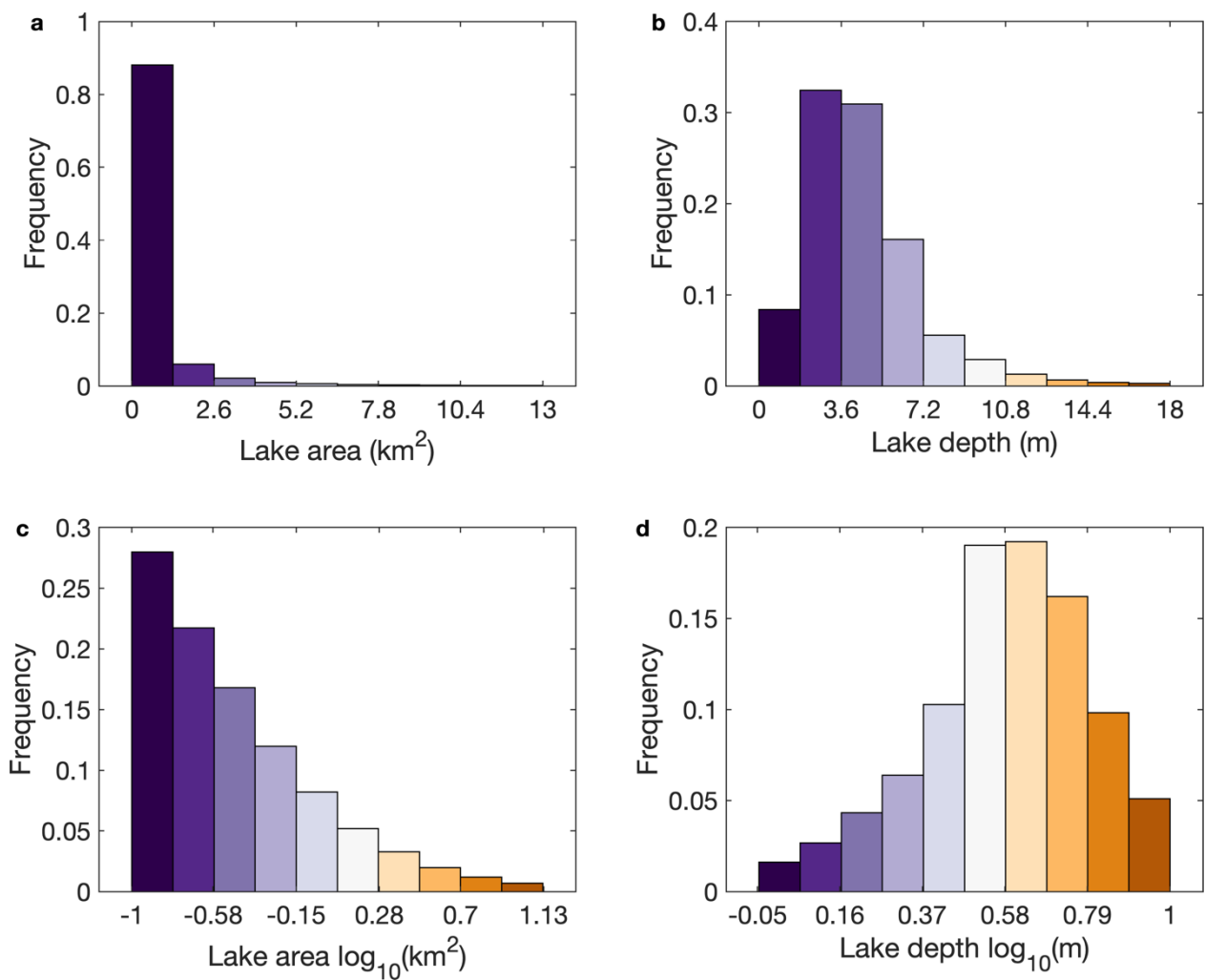
(a) long-wave radiation ( $F_{\lambda, long}$ ); (b) short-wave radiation ( $F_{\lambda, short}$ ) (c) atmospheric temperature near the surface ( $T_a$ ); (d) precipitation ( $P_r$ ); (e) specific humidity ( $h_s$ ); and (f) near-surface wind ( $U_a$ ).



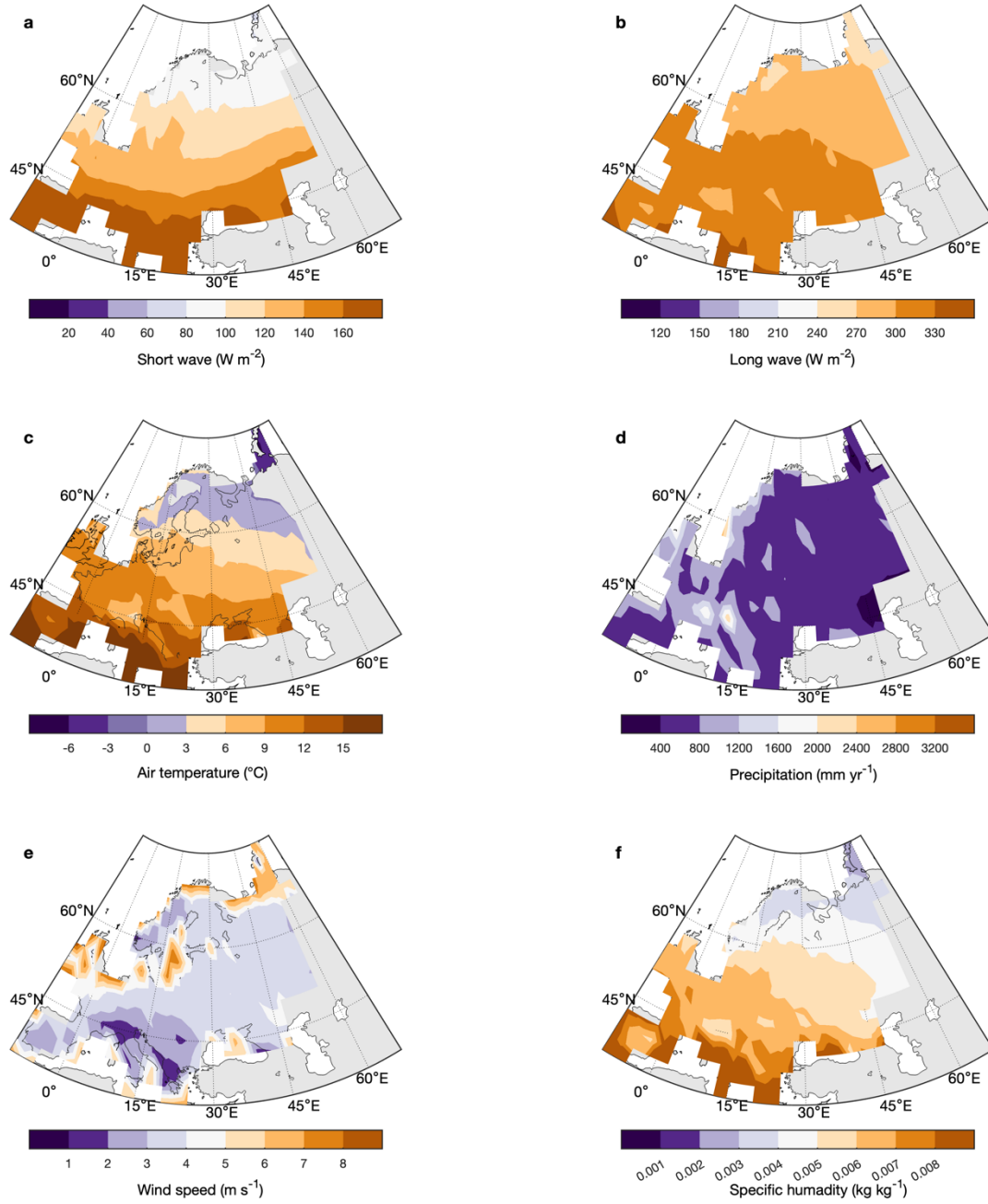
**Fig. S4.** Spatial distribution of lake areas within the European domain. (a) **Lake** area within each grid cell ( $\text{km}^{-2}$ ); (b) **Land** area within each grid cell ( $\text{km}^{-2}$ ); (c) The ratio of lake area to land area within each grid cell.



**Fig. S5.** Frequency distributions of lake area and depth within European domain. Frequency distributions of (a) lake area (km<sup>2</sup>), (b) depth (m), (c) lake area (log<sub>10</sub>(km<sup>2</sup>)), and (d) lake depth (log<sub>10</sub>(m)).

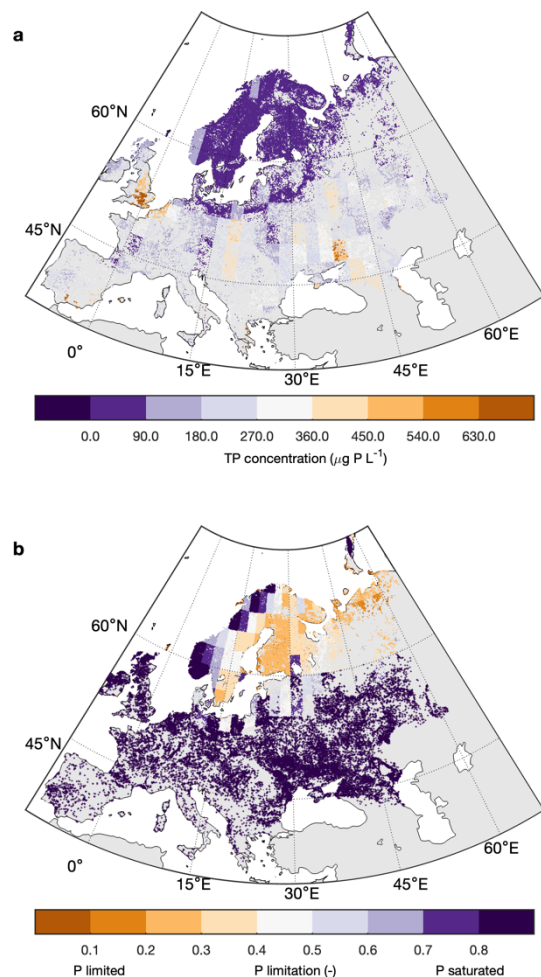


**Fig. S6.** Spatial distribution of climate forcings in the European domain. (a) Short wave radiation ( $\text{W m}^{-2}$ ); (b) Long wave radiation ( $\text{W m}^{-2}$ ); (c) Air temperature ( $^{\circ}\text{C}$ ); (d) Precipitation ( $\text{mm yr}^{-1}$ ); (e) Wind speed ( $\text{m s}^{-1}$ ); (f) Specific humidity ( $\text{kg kg}^{-1}$ ).

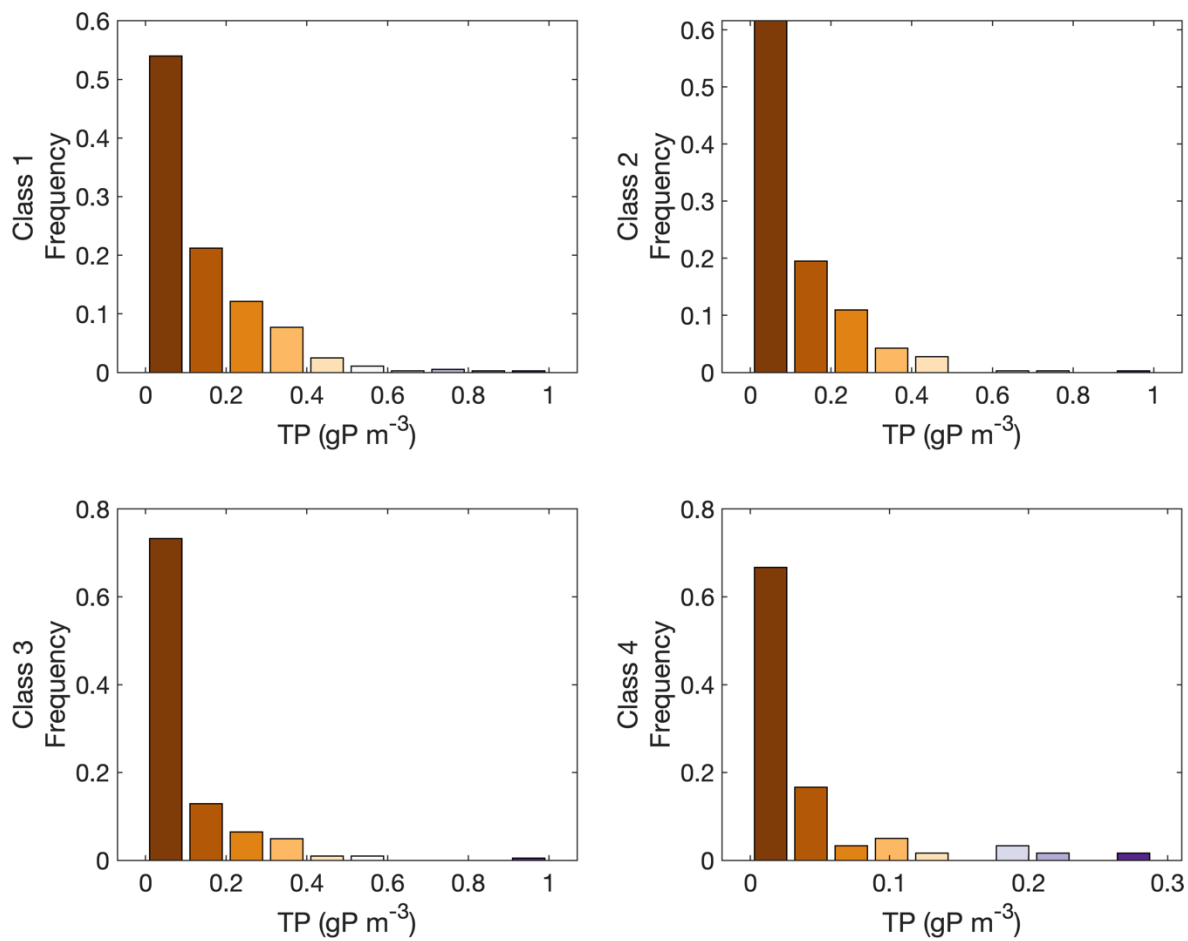




**Fig. S7.** Spatial distribution of (a) total phosphorus concentration ([TP],  $\mu\text{g L}^{-1}$ ) and (b) P limitation in European lakes. The P limitation is evaluated based on the Michaelis-Menten equation with total phosphorus concentration from IMAGE-GNM model.

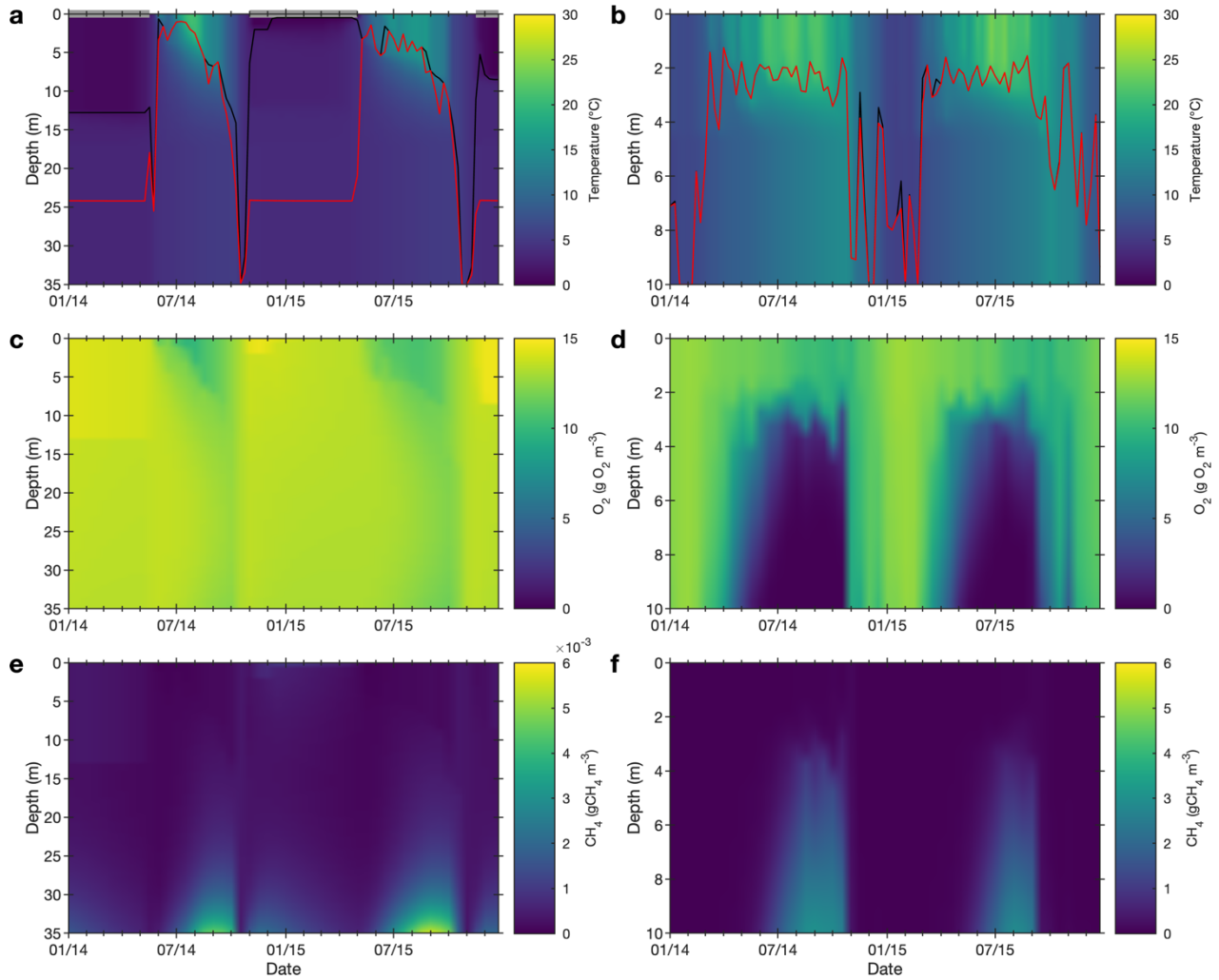


**Fig. S8.** Frequency distribution of total phosphorus concentration (TP;  $\text{g P m}^{-3}$ ) for lakes of different sizes. (a) Class 1:  $0.1\text{--}1 \text{ km}^2$ ; (b) Class 2:  $1\text{--}10 \text{ km}^2$ ; (c) Class 3:  $10\text{--}100 \text{ km}^2$ ; (d) Class 4:  $100\text{--}1000 \text{ km}^2$ .

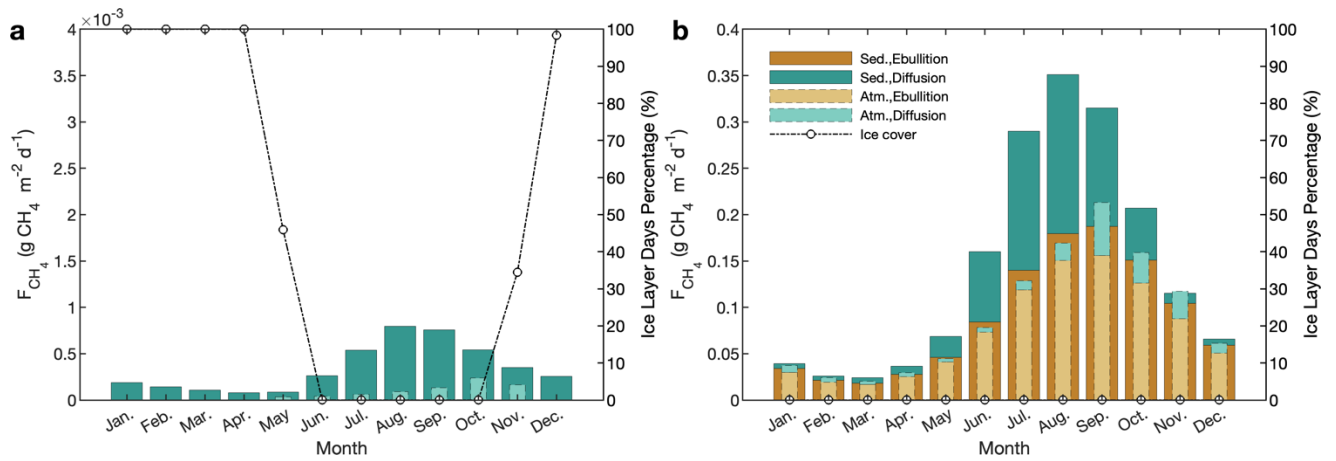


**Fig. S9.** Vertical profiles of  $T$ ,  $[O_2]$  and  $[CH_4]$  for the deep oligotrophic lake (left) and the shallow eutrophic lake (right). Top row: Temperature, mixing depth (black line), photic depth (red curve) and period of ice cover (grey shaded areas at the lake surface). Middle row: Oxygen. Bottom row:  $CH_4$ .

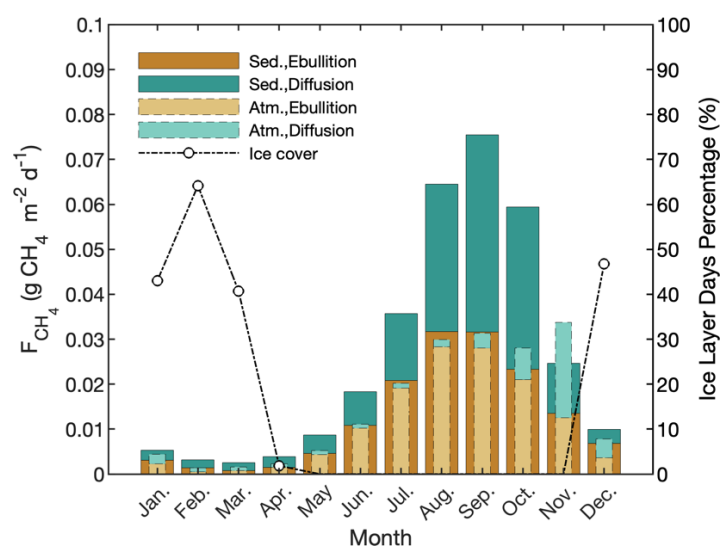
*Note the different scales for  $CH_4$  in panels e and f.*



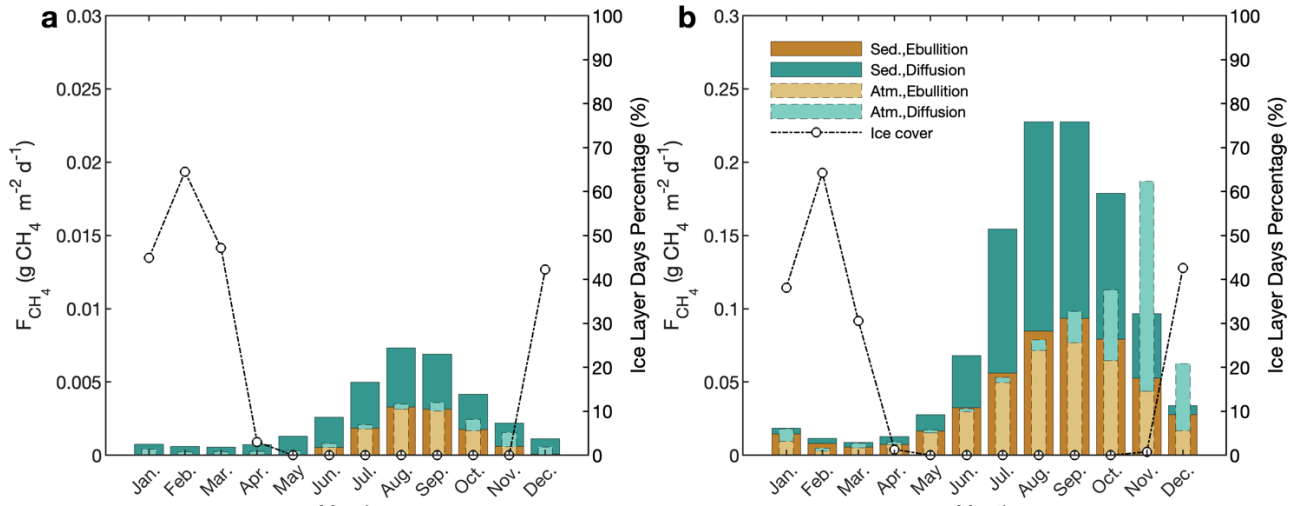
**Fig. S10.** Comparison of seasonality of sedimentary methane ( $\text{CH}_4$ ) production (fainted colors) and  $\text{CH}_4$  emissions to the atmosphere (plain colors) for the two representative lakes. (a) deep oligotrophic lake, driven by a cold climate ( $63.75^\circ\text{N}$ ,  $26.25^\circ\text{E}$ ), and (b) the shallow eutrophic lake, driven by a warm climate ( $43.75^\circ\text{N}$ ,  $-6.25^\circ\text{E}$ ). Orange: diffusive flux; Cyan: Ebullitive flux. Dashed curve: Percent days with ice cover. *Note the different scales in panels a and b.*



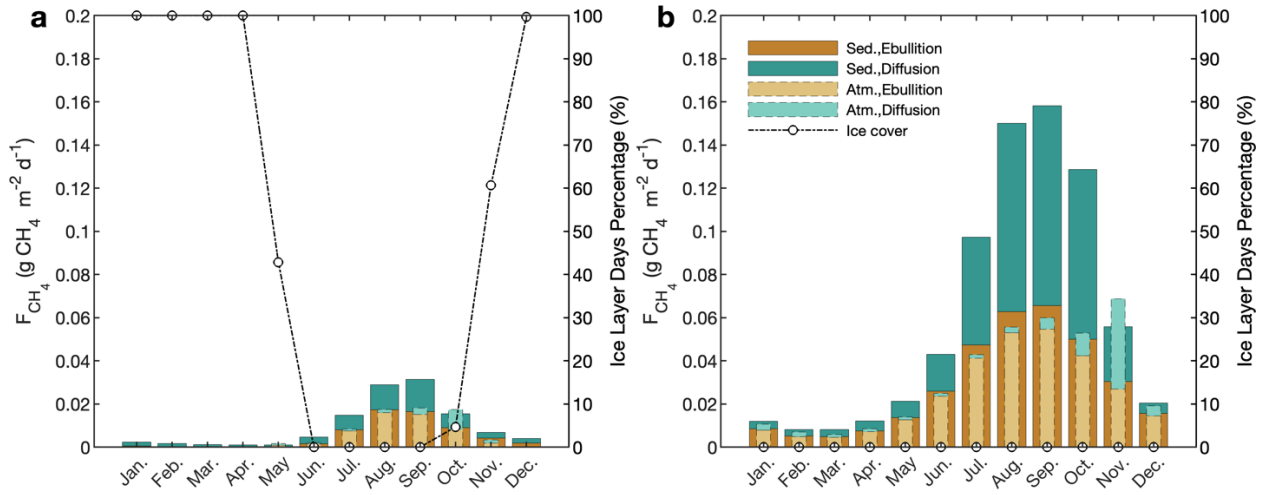
**Fig. S11.** Baseline of the seasonality of CH<sub>4</sub> productions and emissions. The baseline mesotrophic lake has a maximal depth of 15 m (a mean depth of 7.5 m), and a [TDP] of 30 µg P L<sup>-1</sup>, and climate forcings at the grid cell of 48.75°N, 8.75°E. Note that the diffusive emission is higher than the production in November, indicating the occurrence of storage flux due to lake turnover.



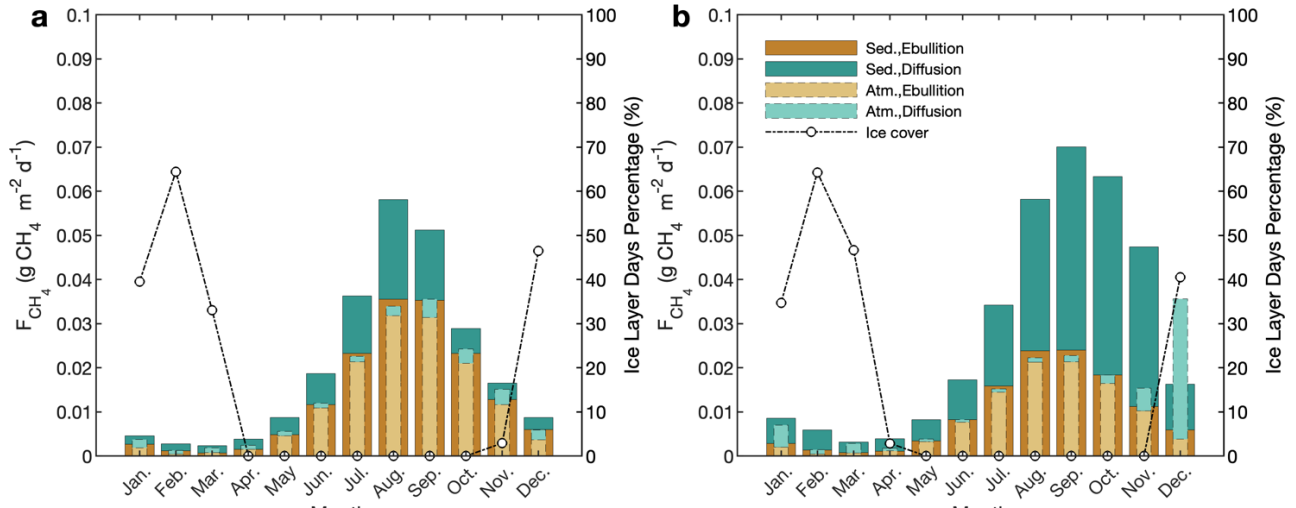
**Fig. S12.** Effects of lake trophic status on  $\text{CH}_4$  dynamics. The lake has a [TDP] of (a)  $8 \mu\text{gP L}^{-1}$  and (b)  $80 \mu\text{gP L}^{-1}$ . The lake depth, area and climate conditions are set the same as those in the baseline. Note that the diffusive emission is higher than the production in November and December, indicating the occurrence of storage flux due to lake turnover. *Note the different scales in panels a and b.*



**Fig. S13.** Effects of climate conditions on CH<sub>4</sub> dynamics. The climate conditions are set as (a) warm (43.75°N, -6.25°E) and (b) cold (63.75°N, 26.25°E). The [TDP] and lake depth are set the same as those in the baseline. Note that the higher diffusive emission than the production (October in a and November in b) indicates the occurrence of storage flux due to lake turnover.

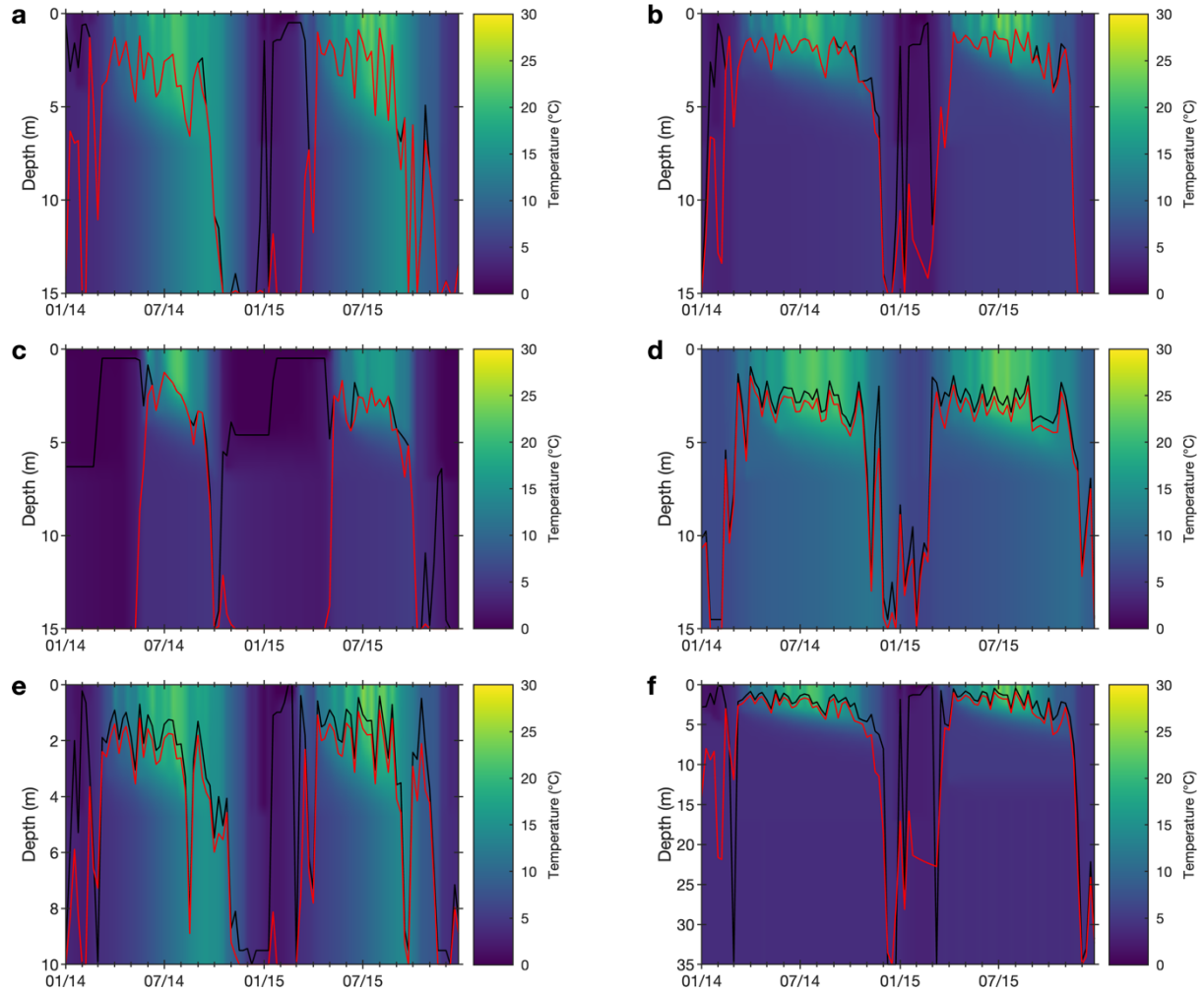


**Fig. S14.** Effects of lake depth on  $\text{CH}_4$  dynamics. The lake depth is set as (a)  $h_{\text{max}} = 10 \text{ m}$  ( $h_{\text{mean}} = 5 \text{ m}$ ) and (b)  $h_{\text{max}} = 35 \text{ m}$  ( $h_{\text{mean}} = 17.5 \text{ m}$ ). The [TDP] and climate conditions are set the same as those in the baseline. Note that the higher diffusive emission than the production indicates the occurrence of storage flux due to lake turnover.

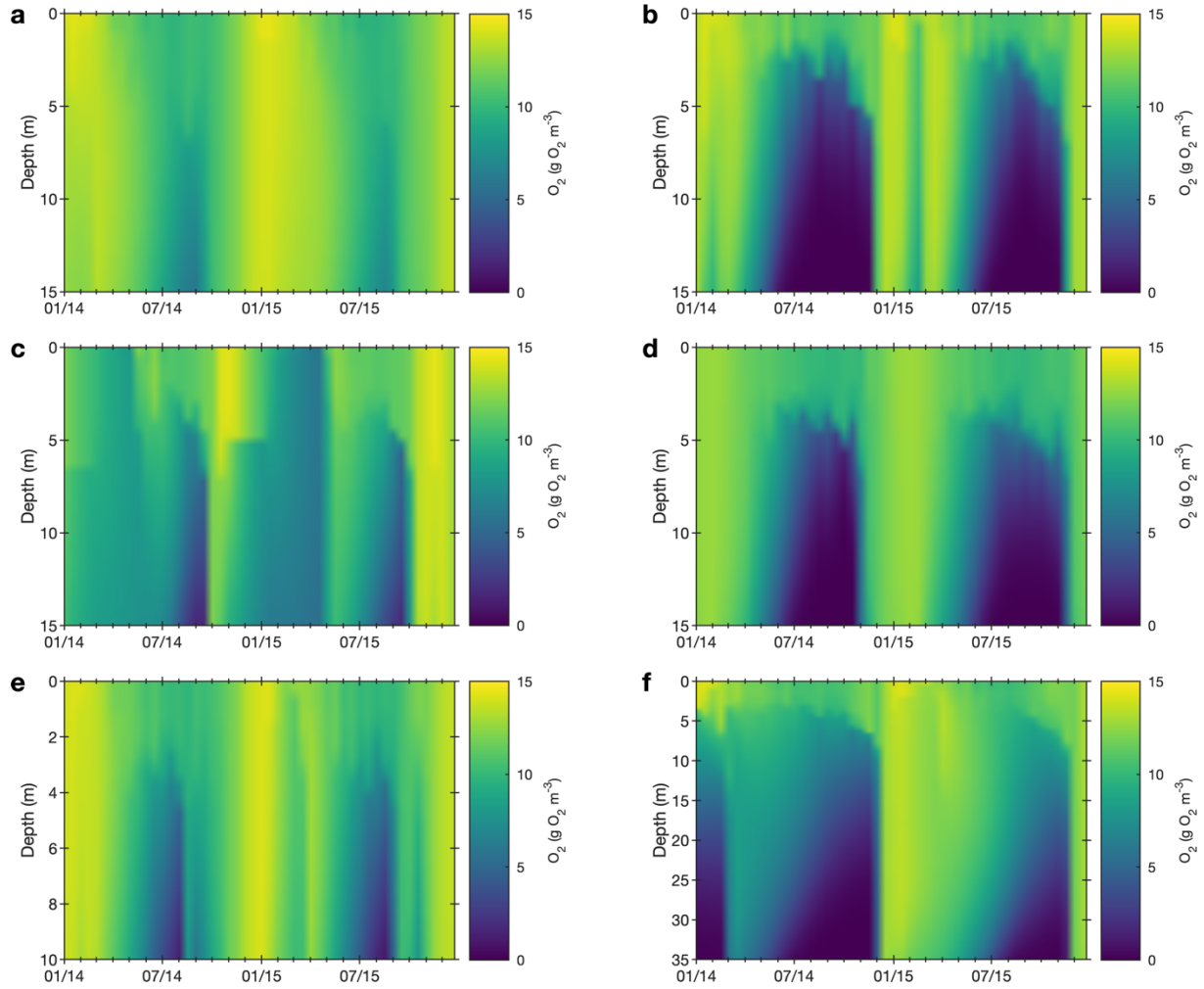




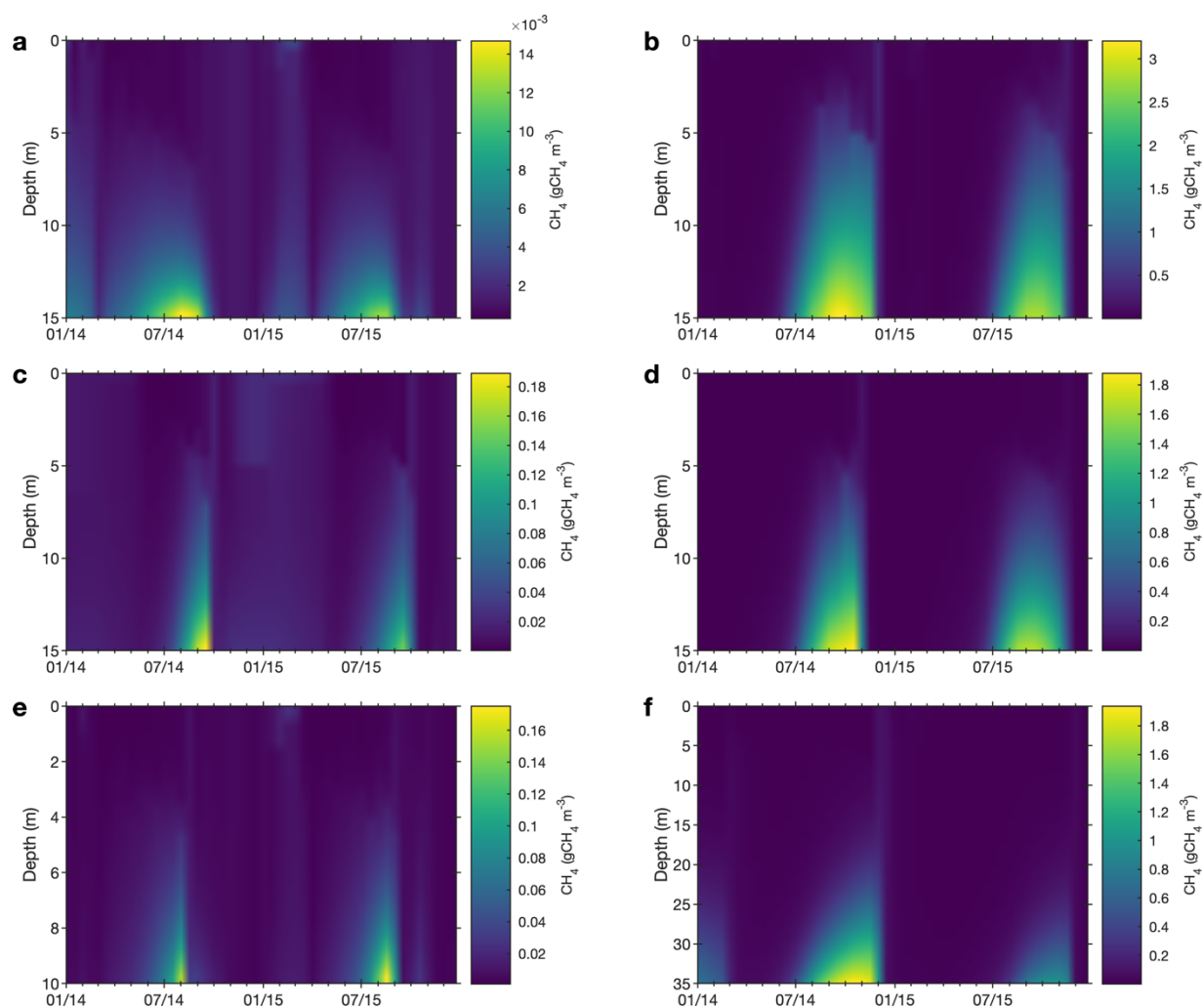
**Fig. S15.** Vertical profiles of lake temperature across environmental gradients. (a) and (b): Lakes with different trophic status ( $[TDP]=8\ \mu\text{g L}^{-1}$  and  $80\ \mu\text{g L}^{-1}$ ); (c) and (d): Lakes driven by different climate condition (cold vs. warm climate); (e) and (f): Lakes with different depth (10 m vs. 35 m).



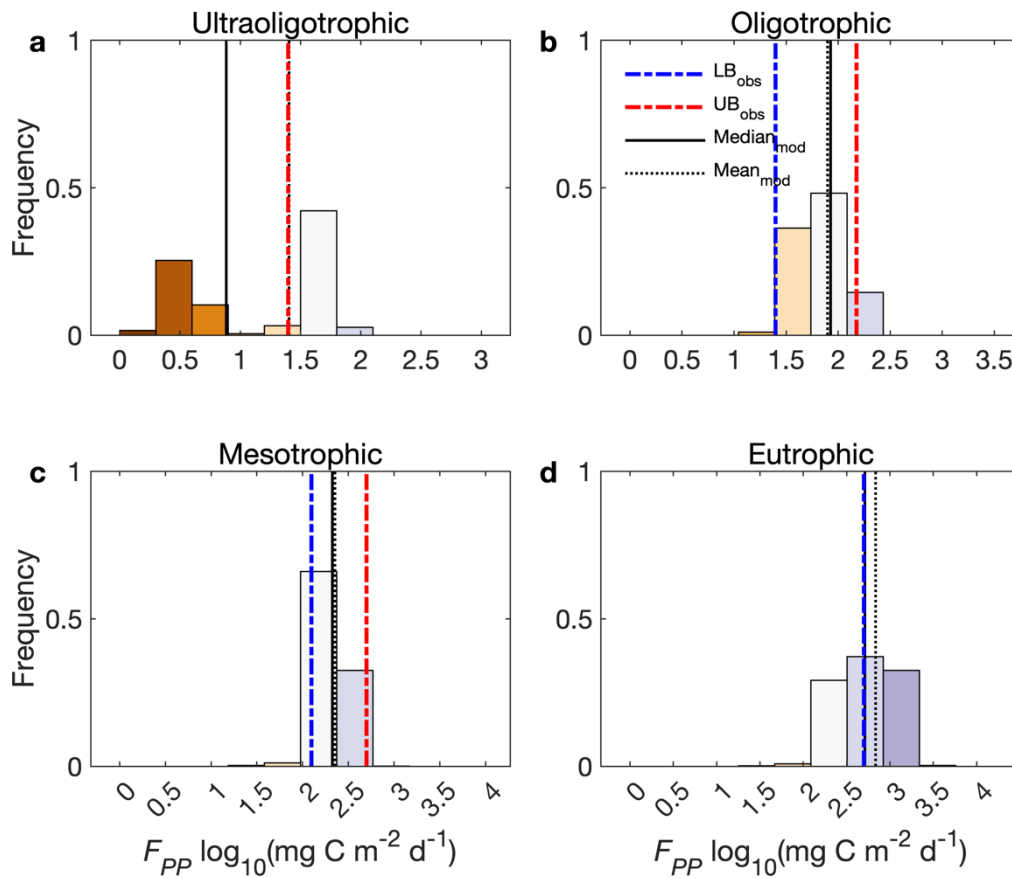
**Fig. S16.** Vertical lake oxygen profiles across environmental gradients. (a) and (b): Lakes with different trophic status ( $[TDP]=8 \mu\text{g L}^{-1}$  and  $80 \mu\text{g L}^{-1}$ ); (c) and (d): Lakes driven by different climate condition (cold vs. warm climate); (e) and (f): Lakes with different depth (10 m vs. 35 m).



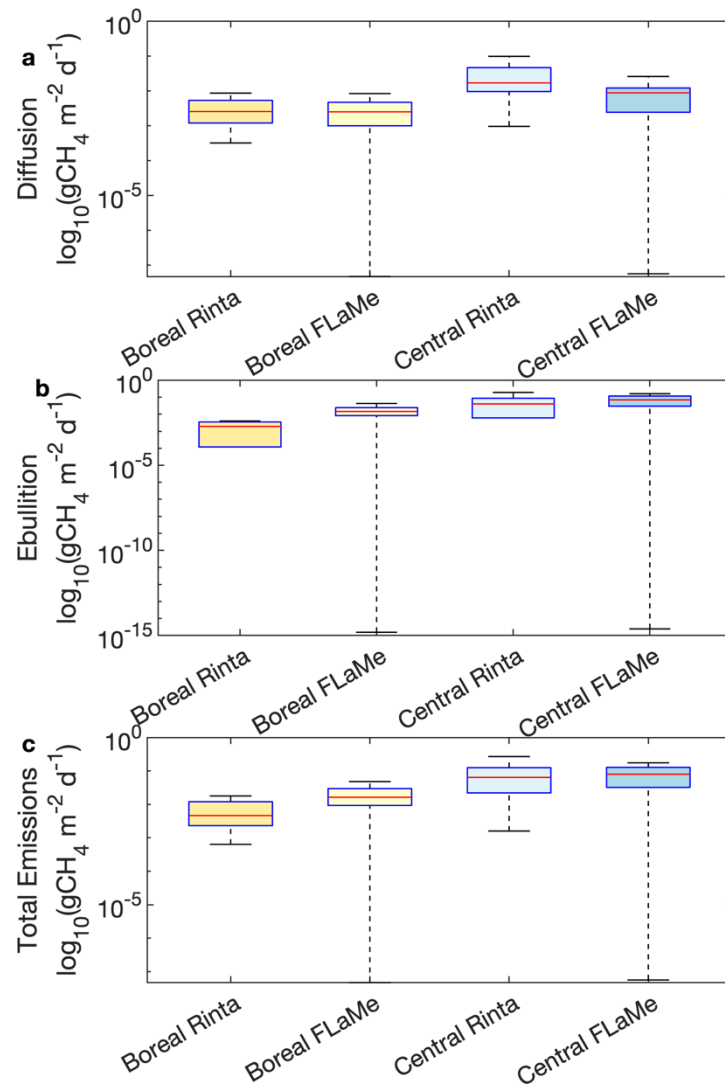
**Fig. S17.** Vertical lake methane profiles across environmental gradients. (a) and (b): Lakes with different trophic status ( $[TDP]=8 \mu\text{g L}^{-1}$  and  $80 \mu\text{g L}^{-1}$ ); (c) and (d): Lakes driven by different climate condition (cold vs. warm climate); (e) and (f): Lakes with different depth (10 m vs. 35 m).



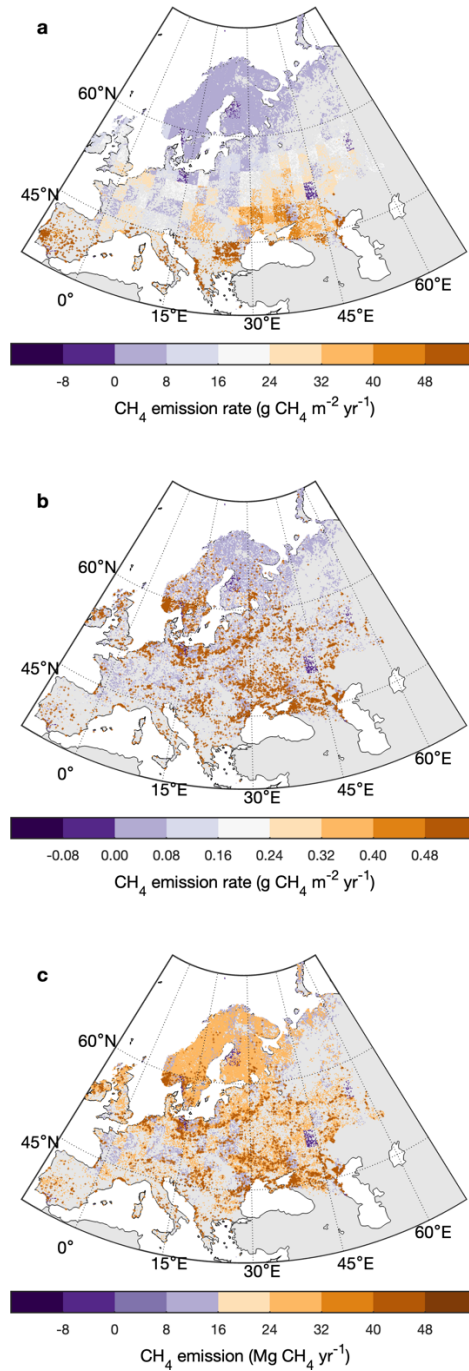
**Fig. S18.** Comparison of simulated depth-integrated primary production ( $F_{PP}$ ) with empirical estimates reported by Wetzel (2001). The histograms show the frequency distributions of simulated  $F_{PP}$  (log scale) in all European lakes ( $n = 108407$ ) that are grouped into ultraoligotrophic ( $0\text{--}5\ \mu\text{gP L}^{-1}$ ), oligotrophic ( $5\text{--}10\ \mu\text{gP L}^{-1}$ ), mesotrophic ( $10\text{--}30\ \mu\text{gP L}^{-1}$ ), and eutrophic ( $>30\ \mu\text{gP L}^{-1}$ ) lakes. In the figure, blue and red dashed lines are the lower and upper bounds ( $\text{LB}_{\text{obs}}$  and  $\text{UB}_{\text{obs}}$ ), respectively; Black solid and dotted lines are the  $\text{median}_{\text{mod}}$  and  $\text{mean}_{\text{mod}}$ , respectively, of simulated  $F_{PP}$  in this class of lakes.



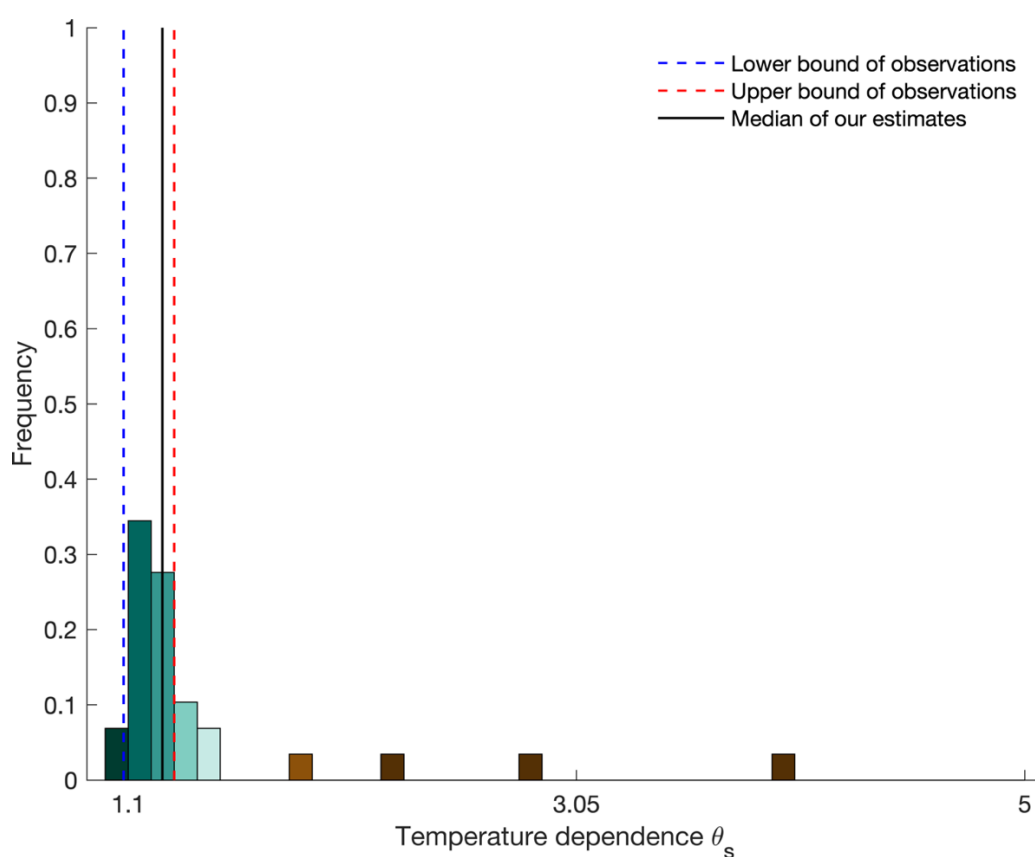
**Fig. S19.** Comparison of simulated diffusive (top row), ebullitive (middle row) and total (bottom row)  $\text{CH}_4$  emission rates (in log scale) with the measurements complied by Rinta *et al.* (2017). The datasets reported by Rinta *et al.* (2017) comprises the diffusive, ebullitive and total emission rates from 17 boreal lakes in Finland and Sweden and 30 lakes of central European lakes in The Netherlands, Germany and Switzerland. The boxes represent the 25% and 75% quartiles, and the whiskers cover the 95% confidence intervals. The same figure with an arithmetic scale is presented in Fig. 7.



**Fig. S20.** Comparison of spatial distributions of CH<sub>4</sub> emission rates. (a) CH<sub>4</sub> emission rate (g CH<sub>4</sub> m<sup>-2</sup> yr<sup>-1</sup>, per *lake* area); (b) CH<sub>4</sub> emission rate (g CH<sub>4</sub> m<sup>-2</sup> yr<sup>-1</sup>, per *land* area) within each 0.1°×0.1° grid cell; (c) aggregated CH<sub>4</sub> fluxes within 0.1°×0.1° grid cell (Mg CH<sub>4</sub> yr<sup>-1</sup>).



**Fig. S21.** Comparison of the simulated temperature dependence ( $\theta_s$ ) of net ebullitive methane emissions against observed ranges reported by Aben *et al.* (2017). The histogram shows the frequency distribution of annual mean values of temperature dependence estimated from 953 representative lakes (an annual mean estimate for each lake). The black bold line is the median of our estimates, which falls well within the lower (blue dashed line) and upper (red dashed line) bounds of observed range.



**Table S1. The comparison of FLaMe-v1.0 with the existing lake models**

	GLM3.0	LAKE2.0	bLake4Me	ALBM	FLaMe-v1.0
<b>Lake shape</b>	User-specified	Bucket/Valley	Bucket	User-specified/Valley	Valley
<b>Application</b>	Site	Site/Regional	Site/Global	Site/Global	Regional/Global
<b>Physical processes</b>					
Water temperature fields	✓	✓	✓	✓	✓
Lake stratification & turnover	✓	✓	✓	✓	✓
Ice dynamics	✓	✓	✓	✓	✓
Sedimentary temperature fields	✓	✓	✓	✓	✗
Hydrological routing	✓	✗	✗	✗	✗
<b>Biogeochemical processes</b>					
Autochthonous C dynamics	✗	✓	✗	✓	✓
P limitation with C	✗	✗	✗	✓	✓
N limitation with C	✗	✗	✗	✗	✗
Allochthonous C input from thawing	✗	✓	✓	✓	✗
Oxygen profile	✗	✗	✗	✓	✓
Sedimentary methane production & its split between diffusion and ebullition	✗	✓	✓	✓	✓
Methane oxidation in water and dissolution in gas bubbles	✗	✓	✓	✓	✓
Gas exchanges with air	✗	✓	✓	✓	✓

a. Red ticks indicate the model has the capability but it was only applied at site level and not to regional or global to scales.



**Table S2. Comparison of means and medians (g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) between observed and simulated methane emission rates in boreal and central European lakes. See Fig. 8 and Fig. S19 for a graphical representation.**

	Mean				Median			
	Boreal		Central Europe		Boreal		Central Europe	
	Obs.	Sim.	Obs.	Sim	Obs.	Sim.	Obs.	Sim.
Diffusion	0.0042	0.0029	0.0337	0.0084	0.0026	0.0025	0.0170	0.0088
Ebullition	0.0051	0.0179	0.0846	0.0722	0.0019	0.0146	0.0402	0.0700
Total	0.0089	0.0199	0.1177	0.0807	0.0046	0.0164	0.0642	0.0801