

1 SUPPORTING INFORMATION

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3 **Spatial variations of methane emission in a large and shallow eutrophic lake in**
4 **subtropical climate**

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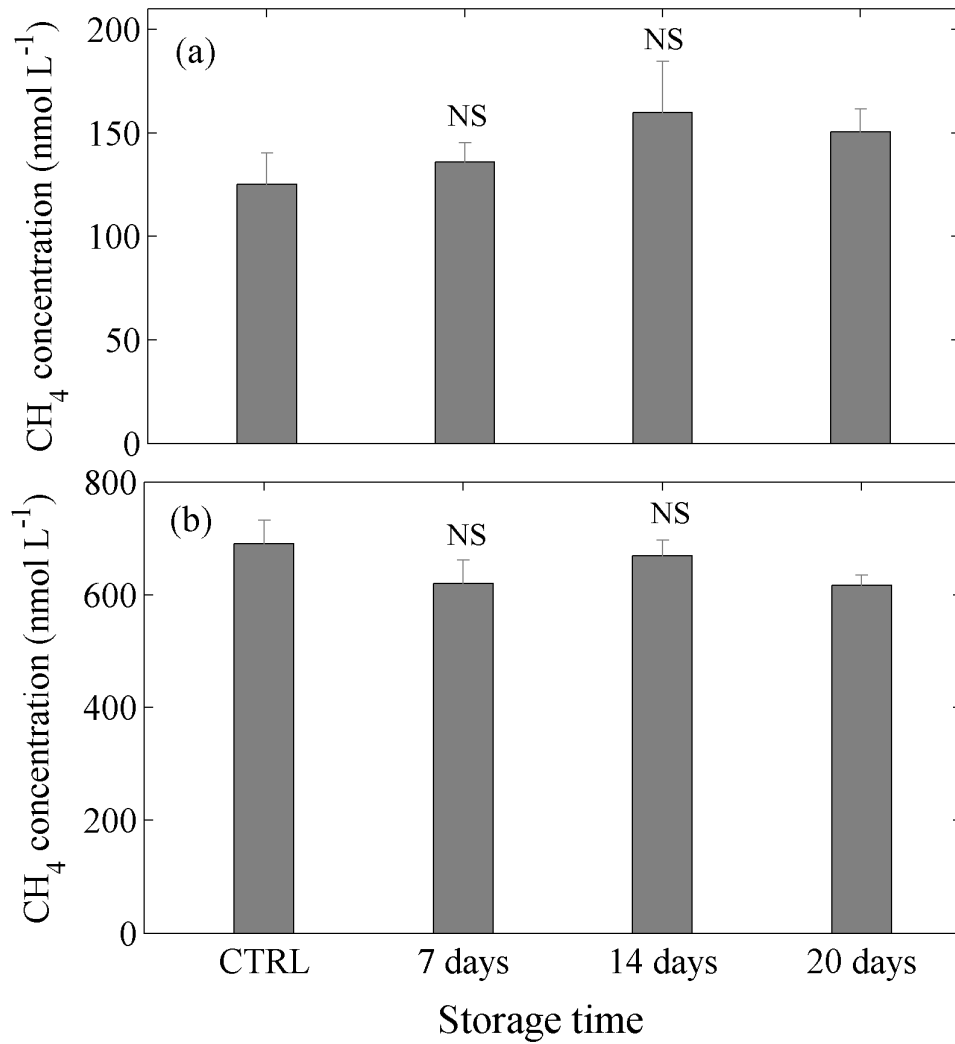
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34 Supplementary text regarding comparison of the gas transfer coefficient k calculated by four
35 different models

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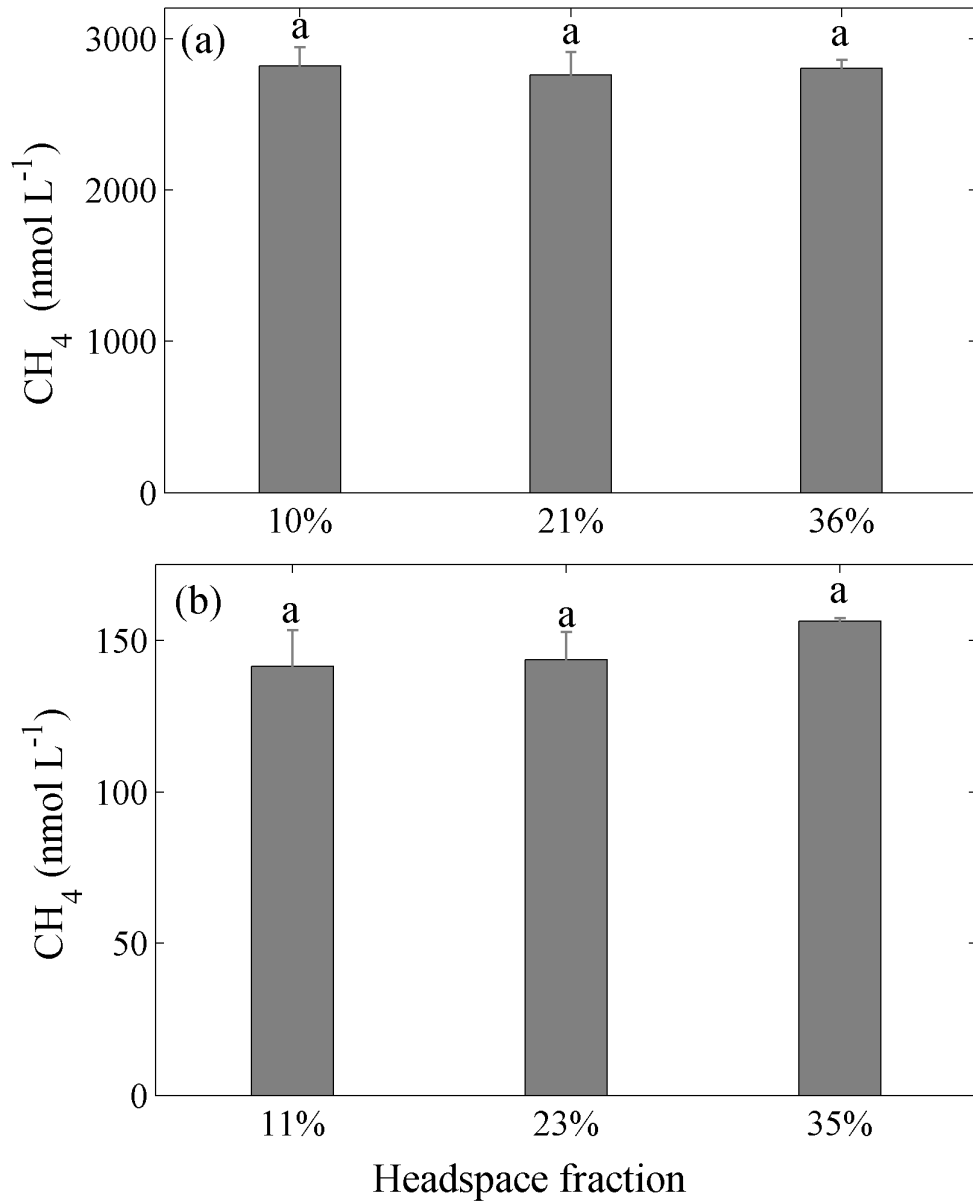
37 Figure S7. Comparison of the diffusion CH₄ flux calculated with four different models of the
38 gas transfer coefficient.

39 **Figure S1.** Effects of storage time on the dissolved methane concentration using water
40 samples collected at BFG (a) and MLW (b). Each treatment was replicated three times. Error
41 bars are one standard deviation. CTRL: measurement was made without delay. NS: difference
42 from CNTRL is not statistically significant.
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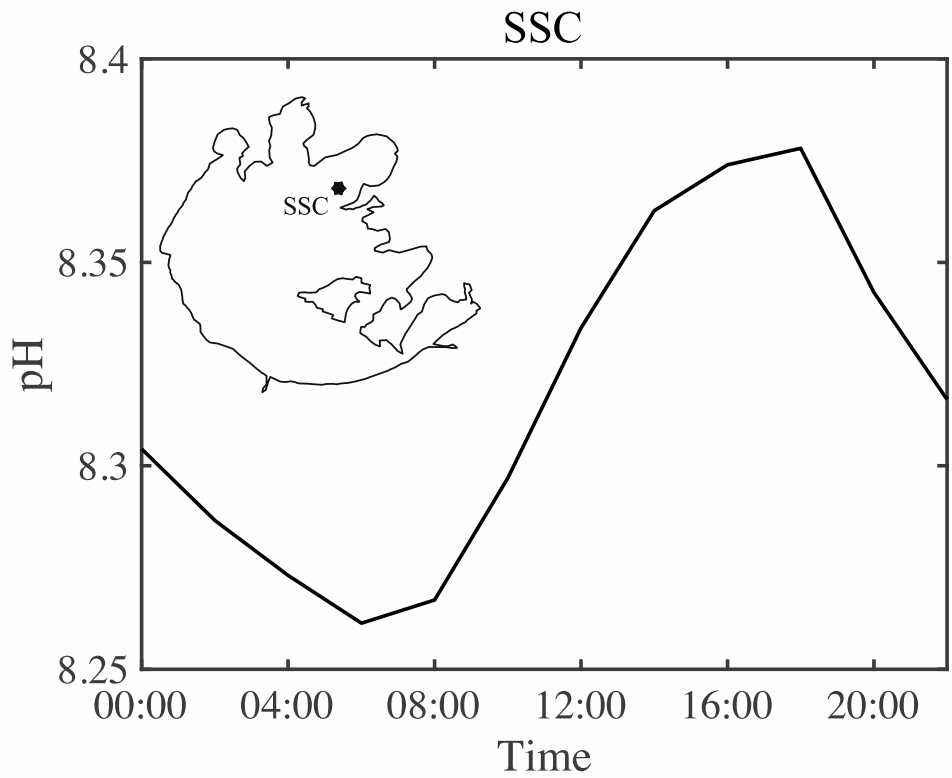
45 **Figure S2.** Effects of headspace fraction on the dissolved methane concentration using water
46 samples collected at a local pond (a) and at MLW (b). Each treatment was replicated three
47 times. Error bars are one standard deviation.



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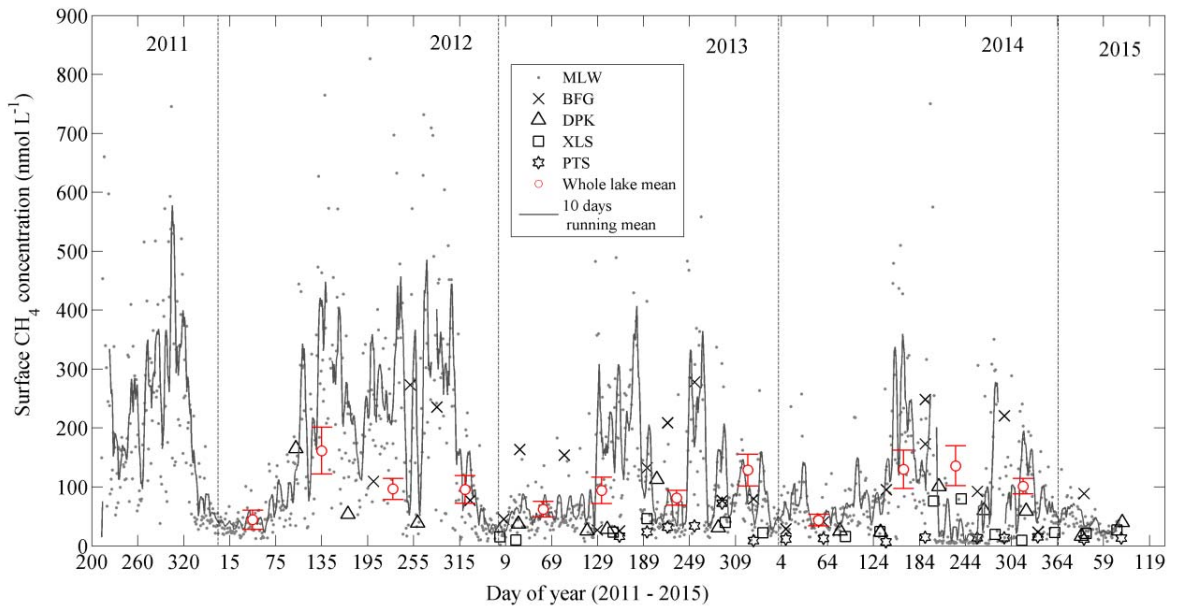
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50 **Figure S3.** A diel composite of pH observed at the 20-cm depth at a buoy site in Gonghu Bay
51 (location labeled as SSC in the map inset). Observations were made over 155 days in the
52 summer of 2009 and in the winter of 2009-2010 (Hu *et al.*, 2015).



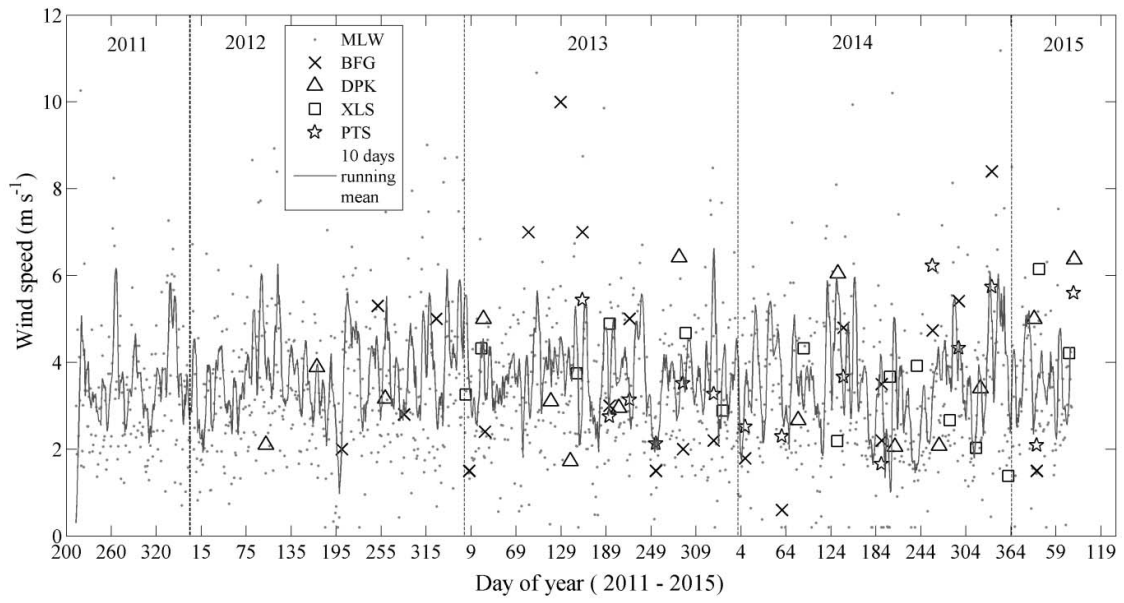
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54 **Figure S4.** Temporal variation of the surface dissolved CH₄ at the five lake observation sites
55 (MLW, BFG, DPK, XLS, and PTS) where frequent water sampling took place. Their
56 locations are shown in Figure 1. Red circles indicate the whole-lake mean dissolved CH₄
57 concentration. The error bar is ±1 standard deviation.



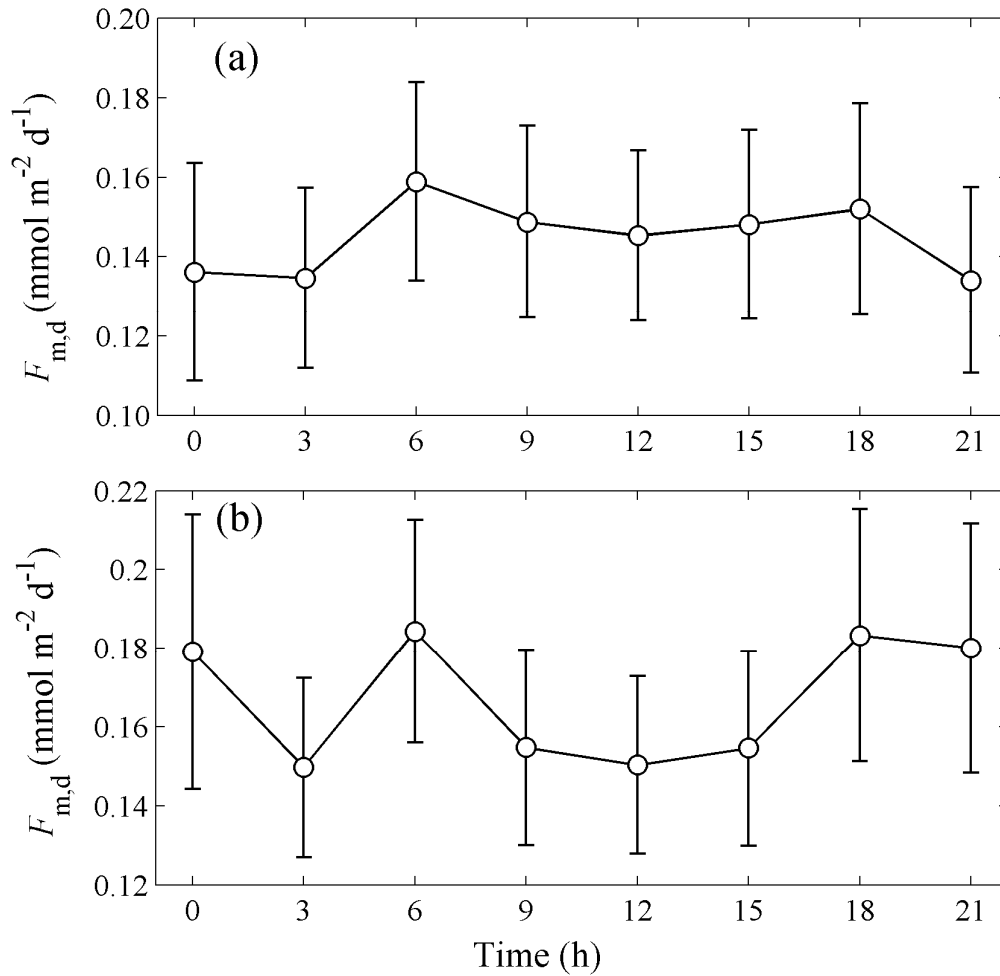
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60 **Figure S5.** Temporal variation of wind speed at the five lake observation sites (MLW, BFG,
61 DPK, XLS, and PTS) where frequent water sampling took place. Their locations are shown in
62 Figure 1.



63
64

65 **Figure S6.** Diel variation of the diffusion CH_4 flux at MLW. The gas transfer coefficient was
66 determined with the model described by Cole *et al.* (1998, a), and with a model that considers
67 both wind speed and waterside convection (Podgrajsek *et al.*, 2015, b). Error bars are ± 1
68 standard error.



69

70 **Table S1.** Annual mean total phosphorous (TP) and total nitrogen (TN) of the seven zones in
 71 Lake Taihu in 2014.

Zones	Area (km ²)	TP (mg L ⁻¹)	TN (mg L ⁻¹)	Trophic class ¹
Meiliang Bay	100	0.087	2.18	Eutrophic
Gonghu Bay	215.6	0.065	1.81	Mesotrophic
East Zone	316.4	0.033	1.23	Mesotrophic
Dongtaihu Bay	131	0.037	0.90	Mesotrophic
Southwest Zone	443.2	0.067	2.01	Mesotrophic
Northwest Zone	394.1	0.094	2.58	Hyper-eutrophic
Central Zone	737.5	0.072	1.89	Mesotrophic
Whole lake	2338	0.069	1.90	Eutrophic

72 Data source: The Health Status Report of Taihu Lake, Taihu Basin Authority of Ministry of
 73 Water Resources and Electric Power, <http://www.tba.gov.cn/>.

74

75 1: Trophic classifications are defined according to OECD (Organization for Economic
 76 Cooperation and Development) (1982), Eutrophication of Waters. Monitoring assessment and
 77 control. Final Report. OECD Cooperative Programme on Monitoring of Inland Waters
 78 (Eutrophication Control), Environment Directorate, OECD, Paris.

79

80 **Table S2.** Temporal correlation of the diffusion CH₄ flux (mmol m⁻² d⁻¹) with wind speed (m
 81 s⁻¹) and dissolved CH₄ concentration (nmol L⁻¹) at five locations (MLW, BFG, DPK, XLS,
 82 and PTS).

83

Site	CH ₄ concentration	Wind speed
MLW	y = 0.0011x - 0.0193 R = 0.92 p < 0.001 n = 1261	y = 0.0934x - 0.1852 R = 0.09 p < 0.001 n = 1264
BFG	y = 0.0015x - 0.0468 R = 0.84 p < 0.001 n = 24	y = 0.0514x - 0.0601 R = 0.21 p = 0.315 n = 24
DPK	y = 0.0010x - 0.0032 R = 0.93 p < 0.001 n = 15	y = -0.0255x + 0.1470 R = -0.21 p = 0.458 n = 15
XLS	y = 0.0016x - 0.0164 R = 0.97 p < 0.001 n = 15	y = 0.0276x - 0.0688 R = 0.33 p = 0.223 n = 15
PTS	y = 0.0010x - 0.0028 R = 0.94 p < 0.001 n = 15	y = 0.0114x - 0.0232 R = 0.13 p = 0.654 n = 15

84

85

86 **Table S3.** Pearson correlation between these explanatory environmental variables measured
 87 at the 29 spatial sampling sites. DO, dissolved oxygen concentration; Chl-a, chlorophyll a
 88 concentration; Spc, specific conductance; ORP, oxidation reduction potential; NTU, turbidity;
 89 Depth, water depth; Clarity, water clarity.

90

	NDVI	DO	pH	Chl-a	NTU	Depth	Spc	ORP
NDVI								
DO	-0.02							
pH	-0.34*	0.72**						
Chl-a	0.31	0.70**	0.48**					
NTU	-0.61**	0.07	0.27	0.23				
Depth	0.01	0.25	0.43*	0.04	0.11			
Spc	0.291	-0.49**	-0.57**	0.43*	-0.35*	0.09		
ORP	-0.01	-0.18	-0.08	0.05	-0.20	-0.04	0.08	
Clarity	0.58**	0.12	-0.11	0.54**	-0.61	-0.23	-0.10	0.01

91 *, ** Correlation is significant at the 0.05, and 0.01 level, respectively.

92

93 **Table S4.** Summary of the general multiple regressions: variance inflation factor (VIF), R^2 ,
 94 significance levels of the predictor variables (p), and the Akaike information criterion (AIC).
 95

Model	Explanatory variables	VIF	R^2	p	AIC
1	NDVI		0.27	0.004	-148.29
2	NDVI	1.51	0.40	0.001	-151.98
	Water clarity	1.51			
3	NDVI	1.52	0.63	<0.001	-164.19
	Water clarity	1.55			
	Dissolved oxygen	1.03			
4	NDVI	1.60	0.78	<0.001	-176.46
	Water clarity	1.75			
	Dissolved oxygen	1.14			
	Water depth	1.21			

96

97 **Comparison of the diffusion flux calculated with four different models for the gas**
98 **transfer coefficient**

99 In this supplementary section, we present a comparison of the diffusion flux calculated using
100 four different models for the gas transfer coefficient. The four models are described by Cole
101 *et al.* (1998, k_1), Read *et al.* (2012, k_2), Heiskanen *et al.* (2014, k_3), and Podgrajsek *et al.*
102 (2015, k_4).

103

104 The first model is that of Cole *et al.* (1998). In this model, the gas transfer coefficient k_1 is
105 wind-dependent and is normalized to a Schmidt number 600 of a gas at temperature of 20 °C,

$$106 \quad k_1 = k_{600} \times (S_c/600)^{-n} \quad (\text{S1})$$

107 where S_c is Schmidt number for CH₄ at in-situ temperature. For the exponent n , we used the
108 value 2/3 at low wind speed ($U_{10} < 3.7 \text{ m s}^{-1}$) according to Huotari *et al.* (2009) and the value
109 of 1/2 at high wind speed ($U_{10} > 3.7 \text{ m s}^{-1}$) according to MacIntyre *et al.* (1995) and Juutinen
110 *et al.* (2009). An empirical relationship was used to determine k_{600} (cm h^{-1} ; Cole and Caraco,
111 1998):

$$112 \quad k_{600} = 2.07 + 0.215U_{10}^{1.7} \quad (\text{S2})$$

113 where U_{10} is wind speed at the 10-m height (m s^{-1}). The required input is U_{10} , which was
114 measured by a wind sensor at PTS in the lake.

115

116 The second model is a surface renewal scheme described Read *et al.* (2012). It considers both
117 wind shear (ε_w) and waterside convection (ε_w),

$$118 \quad k_2 = \eta(\varepsilon_w)^{0.25} S_c^{-n} \quad (\text{S3})$$

119 where η is a proportionality constant, ν is the kinematic viscosity of water, n is a coefficient
 120 representing surface conditions, and

$$121 \quad \varepsilon = \varepsilon_u + \varepsilon_w \quad (S4)$$

122 is the turbulent kinetic energy dissipation rate representing the total contribution from wind
 123 shear (ε_u) and waterside convection (ε_w). The wind shear contribution is given by

$$124 \quad \varepsilon_u = (\tau_t/\rho_w)/(K\delta_v) \quad (S5)$$

125 where τ_t is the tangential shear stress in air, ρ_w is the density of water, K is the von Karman
 126 constant, and δ_v is the thickness of the viscous sublayer given by Soloviev *et al.* (2007),

$$127 \quad \delta_v = c_1\nu/(\tau_t/\rho_w)^{0.5} \quad (S6)$$

128 where c_1 is a dimensionless constant.

129 The contribution by waterside convection (ε_w) is given as,

$$130 \quad \varepsilon_w = -\beta \quad (S7)$$

131 where β is buoyancy flux defined as

$$132 \quad \beta = \frac{gaQ_e}{\rho_w c_p} \quad (S8)$$

133 where g is the acceleration of gravity, a is the thermal expansion coefficient of water, C_p is
 134 the specific heat of water, Q_e is the effective surface heat flux (Imberger, 1985; Jeffery *et al.*,
 135 2007). If the lake is gaining heat from the atmosphere ($Q_e > 0$), ε_w is set to zero.

136

137 We used the air friction velocity measured at PTS to determine τ_t in Equation S5 and S6, and
 138 approximate the surface heat flux Q_e as the residual of the surface energy balance equation,

$$139 \quad Q_e = R_n - H - \lambda E \quad (S9)$$

140 where R_n (net radiation), H (sensible heat flux), and λE (latent heat flux) were measured at

141 PTS. Other coefficients are given by Read *et al.* (2012) as $\eta= 0.29$, $n = 0.5$, and $S_c = 600$.

142

143 The third model, described by MacIntyre *et al.* (2010) and Heiskanen *et al.* (2014), is also a
144 surface renewal parameterization. It uses different fitting coefficients from Read *et al.* (2012)
145 to calculate the gas transfer coefficient,

$$146 \quad k_3 = 0.5(\varepsilon\nu)^{0.25}S_c^{-n} \quad (\text{S10})$$

$$147 \quad \varepsilon = 0.77 (-\beta) + 0.3 (u_w^*)^3/(Kz) \quad (\text{S11})$$

148 where β is buoyancy flux defined by Equation S8, z is a mixed layer depth, u_w^* is the
149 velocity scale for wind shear given by

$$150 \quad u_w^* = u_a^* \sqrt{\frac{\rho_a}{\rho_w}} \quad (\text{S12})$$

151 where ρ_a is the density of air, u_a^* is the air friction velocity measured at PTS in the lake,
152 S_c is the Schmidt number for CH₄ at in-situ temperature, $n = 0.5$, and the mixing layer depth z
153 was set to 0.5 m according to the thermal diffusivity profile calculated with the model of
154 Herb and Stephan (2005) for Lake Taihu.

155

156 The fourth model is that of Podgrajsek *et al.* (2015) which also considers the effect of
157 waterside convection. The gas transfer coefficient k_4 is given as

$$158 \quad k_4 = k_1 + 0.05 \times \exp(1068 \times (\beta z)^{1/3}) \quad (\text{S13})$$

159 where k_1 is determined by Equation S1, β is defined by Equation S8, and z is the mixed layer
160 depth. In this equation, the second term represents the contribution of waterside convection to
161 the gas transfer.

162

163 We estimated the percentage of the gas transfer (k_w) driven by waterside convection from the
164 last three models. In the case of the second model, k_w was computed from Equation S3 by
165 setting ε_u to zero. In the third and the fourth model, k_w was computed from Equation S10 and
166 S13 by setting u_w^* and k_1 to zero, respectively. The percent of the contribution of waterside
167 convection is

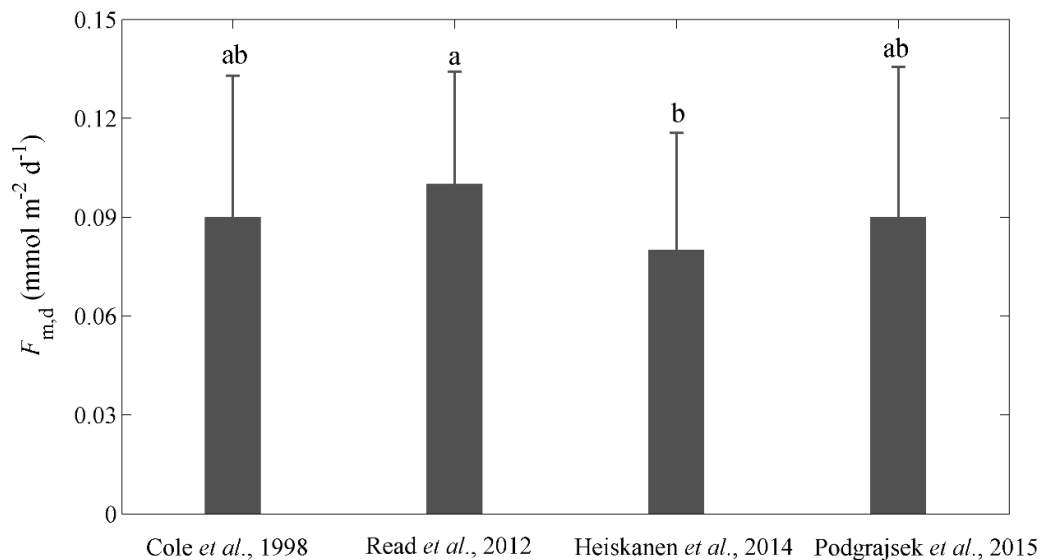
$$168 \quad k_w\% = (k_w/k)100\% \quad (\text{S14})$$

169 where k is the total gas transfer coefficient driven by wind shear and waterside
170 convection.

171

172 Figure S7 compares the annual mean diffusion flux from the four models. The annual mean
173 CH₄ diffusion fluxes based on the four different diffusivity formulations were 0.092 (Cole *et*
174 *al.*, 1998), 0.103 (Read *et al.*, 2012), 0.080 (Heiskanen *et al.*, 2014), and 0.093 mmol m⁻² d⁻¹
175 (Podgrajsek *et al.*, 2015).

176 **Figure S7.** Comparison of the whole-lake diffusion CH₄ flux calculated with four different
177 models of the gas transfer coefficient. Error bars are one standard deviation of the 12 annual
178 mean values for the 29 lake survey locations (Figure 1).



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