SUPPORTING INFORMATION

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

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



**Figure S2.** Effects of headspace fraction on the dissolved methane concentration using water samples collected at a local pond (a) and at MLW (b). Each treatment was replicated three times. Error bars are one standard deviation.



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



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



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



**Figure S6.** Diel variation of the diffusion CH4 flux at MLW. The gas transfer coefficient was determined with the model described by Cole *et al.* (1998, a), and with a model that considers both wind speed and waterside convection (Podgrajsek *et al.,* 2015, b). Error bars are ± 1 standard error.



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

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Zones | Area (km2) | TP (mg L-1) | TN ( mg L-1) | Trophic class1 |
| 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 |

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

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

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

|  |  |  |
| --- | --- | --- |
| Site | CH4 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 |

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

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | 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 |

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

**Table S4.** Summary of the general multiple regressions: variance inflation factor (VIF), R2, significance levels of the predictor variables (*p*), and the Akaike information criterion (AIC).

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Model | Explanatory variables | VIF | R2 | *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 |

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

In this supplementary section, we present a comparison of the diffusion flux calculated using four different models for the gas transfer coefficient. The four models are described by Cole *et al*. (1998, *k*1), Read *et al*. (2012, *k*2), Heiskanen *et al*. (2014, *k*3), and Podgrajsek *et al*. (2015, *k*4).

The first model is that of Cole *et al*. (1998). In this model, the gas transfer coefficient *k*1 is wind-dependent and is normalized to a Schmidt number 600 of a gas at temperature of 20 oC,

*k*1 = *k*600 × (*S*c/600)-*n*(S1)

where *S*c is Schmidt number for CH4 at in-situ temperature. For the exponent *n*, we used the value 2/3 at low wind speed (*U*10 < 3.7 m s-1) according to Huotari *et al*. (2009) and the value of 1/2 at high wind speed (*U*10 > 3.7 m s-1) according to MacIntyre *et al*. (1995) and Juutinen *et al*. (2009). An empirical relationship was used to determine *k*600 (cm h-1; Cole and Caraco, 1998):

*k*600 = 2.07 + 0.215$U\_{10}^{1.7}$ (S2)

where *U*10 is wind speed at the 10-m height (m s-1). The required input is *U*10, which was measured by a wind sensor at PTS in the lake.

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

*k*2 = *η(ε**ν*)0.25*Sc*-*n*(S3)

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

*ε = εu + εw* (S4)

is the turbulent kinetic energy dissipation rate representing the total contribution from wind shear (*ε*u) and waterside convection (*ε*w). The wind shear contribution is given by

*εu =* (*τt/ρw*)*/*(*Κδv*) (S5)

where *τt* is the tangential shear stress in air, *ρw* is the density of water, *K* is the von Karman constant, and *δv* is the thickness of the viscous sublayer given by Soloviev *et al*. (2007),

*δv =* c1*ν*/(*τt /ρw*)0.5 (S6)

where c1 is a dimensionless constant.

The contribution by waterside convection (*ε*w) is given as,

*εw* = -*β* (S7)

where *β* is buoyancy flux defined as

*β* = $\frac{ga Q\_{e}}{ρ\_{wC\_{p}}}$ (S8)

where *g* is the acceleration of gravity, *a* is the thermal expansion coefficient of water, *C*p is the specific heat of water, *Q*e is the effective surface heat flux (Imberger, 1985; Jeffery *et al*., 2007). If the lake is gaining heat from the atmosphere (*Q*e > 0), *ε*w is set to zero.

We used the air friction velocity measured at PTS to determine *τt* in Equation S5 and S6, and approximate the surface heat flux *Q*e as the residual of the surface energy balance equation,

*Qe =* *Rn –* *H –* *λE* (S9)

where *Rn* (net radiation), *H* (sensible heat flux), and *λE* (latent heat flux) were measured at PTS. Other coefficients are given by Read *et al*. (2012) as *η*= 0.29, *n* = 0.5, and *S*c = 600.

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

*k3* = 0.5(*εν*)0.25*Sc*-*n*(S10)

ε = 0.77 (-*β*) + 0.3 ($u\_{w}^{\*}$)3/(*Kz*) (S11)

where *β* is buoyancy flux defined by Equation S8, *z* is a mixed layer depth, $u\_{w}^{\*}$ is the velocity scale for wind shear given by

$u\_{w}^{\*}$ = $u\_{a}^{\*}\sqrt{\frac{ρ\_{a}}{ρ\_{w}}}$ (S12)

where *ρ*a is the density of air, $u\_{a}^{\*}$ is the air friction velocity measured at PTS in the lake,

*S*c is the Schmidt number for CH4 at in-situ temperature, *n* = 0.5, and the mixing layer depth *z* was set to 0.5 m according to the thermal diffusivity profile calculated with the model of Herb and Stephan (2005) for Lake Taihu.

The fourth model is that of Podgrajsek *et al*. (2015) which also considers the effect of waterside convection. The gas transfer coefficient *k*4 is given as

*k*4 = *k*1 + 0.05× exp(1068 × (*β* *z*)1/3)(S13)

where *k*1is determined by Equation S1, *β* is defined by Equation S8, and z is the mixed layer depth. In this equation, the second term represents the contribution of waterside convection to the gas transfer.

We estimated the percentage of the gas transfer (*k*w) driven by waterside convection from the last three models. In the case of the second model, *k*w was computed from Equation S3 by setting *εu* to zero. In the third and the fourth model, *k*w was computed from Equation S10 and S13 by setting $u\_{w}^{\*}$ and *k*1 to zero, respectively. The percent of the contribution of waterside convection is

*kw*%= (*k*w/k)100%(S14)

where *k* is the total gas transfer coefficient driven by wind shear and waterside

convection.

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

**Figure S7**. Comparison of the whole-lake diffusion CH4 flux calculated with four different models of the gas transfer coefficient. Error bars are one standard deviation of the12 annual mean values for the 29 lake survey locations (Figure 1).

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