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Article

Enhancing Surface Methane Fluxes from an Oligotrophic Lake: Exploring the Microbubble Hypothesis

MCGINNIS, Daniel Frank, et al.

Abstract

Exchange of the greenhouse gases carbon dioxide (CO2) and methane (CH4)across inland water surfaces is an important component of the terrestrial carbon (C) balance. We investigated the fluxes of these two gases across the surface of oligotrophic Lake Stechlin using a floating chamber approach. The normalized gas transfer rate for CH4 (k600,CH4) was on average 2.5 times higher than that for CO2 (k600,CO2) and consequently higher than Fickian transport. Because of its low solubility relative to CO2, the enhanced CH4 flux is possibly explained by the presence of microbubbles in the lake's surface layer. These microbubbles may originate from atmospheric bubble entrainment or gas supersaturation (i.e., O2) or both. Irrespective of the source, we determined that an average of 145 L m−2 d−1 of gas is required to exit the surface layer via microbubbles to produce the observed elevated k600,CH4. As k600 values are used to estimate CH4 pathways in aquatic systems, the presence of microbubbles could alter the resulting CH4 and perhaps C balances. These microbubbles will also a the surface fluxes of other sparingly [...]

## Reference

MCGINNIS, Daniel Frank, *et al*. Enhancing Surface Methane Fluxes from an Oligotrophic Lake: Exploring the Microbubble Hypothesis. *Environmental science & technology*, 2015, vol. 49, no. 2, p. 873-880

DOI: 10.1021/es503385d

Available at: http://archive-ouverte.unige.ch/unige:45939

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### **Supporting Information**

# Enhancing Surface Methane Fluxes from an Oligotrophic Lake: Exploring the Microbubble Hypothesis

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#### 13 Pages

Pg S2 – S3: Supplemental methods: Meteorological sensor specifications and turbulent kinetic energy dissipation

Pg S3–S12: 9 Figures

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#### **Supplemental Methods**

#### Weather station specifications

The wind speed/direction, air temperature and humidity data were obtained from the Vaisala Weather Transmitter WXT520 (<u>https://store.vaisala.com/eu</u>). Data are measured and reported every 1 minute. The sensor specifications are

Wind Speed	
Range	$0 - 60 \text{ m s}^{-1}$
Accuracy	$\pm 3\%$ at 10m s <sup>-1</sup>
Output resolutions and unit	0.1 m s <sup>-1</sup> , 0.1 km h <sup>-1</sup>
Wind Direction	
Accuracy	$\pm 3^{\circ}$
Output resolution and unit	1°
Relative Humidity	
Range	0 – 100% RH
Accuracy	±3% RH within 0-90% RH, ±5% RH 90-100% RH
Output resolution and unit	0.1% RH
Air Temperature	
Range	-52 – +60 °C
Accuracy for sensor at +20 °C	±0.3 °C
Output resolutions and unit	0.1 °C

Turbulent kinetic energy (TKE) dissipation. The velocity structure function

$$D(z,r) = [(v'(z)-v'(z+r))^2]$$
(1)

was calculated from the HR-Aquadopp data and was used to estimate total kinetic energy dissipation rate.<sup>see 1</sup> Here, v'(z) is the along-beam velocity fluctuation at distance *z*, *r* is the depth range of the dissipation estimation, square brackets denote time averaging. Three estimations of the velocity fluctuations were determined by extracting the mean velocity value for the averaging periods of 10, 20, and 30 min. Three values of the maximum estimation range for the velocity correlation r = 0.4, 0.5, and 0.6 m were tested, covering the range recommended by *Wiles et al.*<sup>1</sup> for weakly stratified turbulence. The TKE dissipation rate  $\varepsilon$  was estimated by fitting the equation

$$C_{v}^{-3}D(z, r)^{3/2} = r\varepsilon(z) + Noise.$$
 (2)

Here, the constant  $C_v = 3^{1/3}$ .<sup>see e.g. 2</sup> The fitting constant *Noise* representing the average effect of the acoustic noise on the velocity fluctuations was used for the goodness-of-fit check, using the condition

*Noise* > 
$$[C_v^{-3}D^{3/2}]$$
 (3)

to discard corresponding  $\varepsilon$  values with subsequent interpolation between the neighboring values. A further quality check was performed by comparison of the 27 arrays of the TKE dissipation rate calculated from the three HR-Aquadopp beams with three different values of r and three averaging periods. The discrepancy between the different estimations did not exceed 10%. The  $\varepsilon$  estimations based on the averaging time of 20 min and r = 0.4 m had a minimum number of bad values according to the *Noise* condition. Therefore, they were adopted for further analysis instead of an average among the 27 estimations.

- Wiles, P. J.; Rippeth, T. P.; Simpson, J. H.; Hendricks, P. J., A novel technique for measuring the rate of turbulent dissipation in the marine environment. *Geophys. Res. Lett.* 2006, *33*, (21), L21608.
- 2. Lien, R. C.; D'Asaro, E. A., The Kolmogorov constant for the Lagrangian velocity spectrum and structure function. *Physics of Fluids* **2002**, *14*, 4456-4459.





Figure S2. Picture of floating chamber, tubing, and listed dimensions.











Figure S7. Left: August 24 - 25, 2013 time series of CH<sub>4</sub>, CO<sub>2</sub>, O<sub>2</sub> and temperature from probes mounted to the side of the boat. Hashed areas are times when boat was underway or in the harbor. Non-hashed areas are while boat was drifting during transects (transect number indicated at top of plot) towards the middle of the lake. Right: Corresponding CH<sub>4</sub> profiles during the campaign.



1a.  $R^2$  with all data points is 0.65.

