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 affect the surface fluxes of other sparingly [...]
Supporting Information

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

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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 (https://store.vaisala.com/eu). Data are measured and reported every 1 minute. The sensor specifications are:

**Wind Speed**
- **Range**: 0 – 60 m s\(^{-1}\)
- **Accuracy**: ±3% at 10 m s\(^{-1}\)
- **Output resolutions and unit**: 0.1 m s\(^{-1}\), 0.1 km h\(^{-1}\)

**Wind Direction**
- **Accuracy**: ±3°
- **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

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

was calculated from the HR-Aquadopp data and was used to estimate total kinetic energy dissipation rate.\(^1\) 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.\(^1\) for weakly stratified turbulence. The TKE dissipation rate \(\varepsilon\) was estimated by fitting the equation.
\[ C_v^3 D(z, r)^{3/2} = \varepsilon(z) + \text{Noise}. \] (2)

Here, the constant \( C_v = 3^{1/3} \). See e.g. \(^2\) The fitting constant \( \text{Noise} \) representing the average effect of the acoustic noise on the velocity fluctuations was used for the goodness-of-fit check, using the condition

\[ \text{Noise} > [C_v^3 D^{3/2}] \] (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 \( \text{Noise} \) condition. Therefore, they were adopted for further analysis instead of an average among the 27 estimations.


Figure S1. Bathymetric map of Lake Stechlin. Units of depth contours are in meters.
Figure S2. Picture of floating chamber, tubing, and listed dimensions.

<table>
<thead>
<tr>
<th>Specifications</th>
</tr>
</thead>
<tbody>
<tr>
<td>✷ Bottom area of the chamber: 0.126 m²</td>
</tr>
<tr>
<td>✷ Volume: 19.1 L</td>
</tr>
<tr>
<td>✷ Water penetration: ~ 2 cm</td>
</tr>
</tbody>
</table>
Figure S3. Slopes for CH4 chamber readings.

Transect 1
Fit

Adj. R-Square: 0.99598
[CH4]_ppm

Value

Intercept: 46.07463
Slope: 370.75004

Transect 2
Fit

Adj. R-Square: 0.99574
[CH4]_ppm

Value

Intercept: -1.29489E9
Slope: 540.36022

Transect 3
Fit

Adj. R-Square: 0.99429
[CH4]_ppm

Value

Intercept: -1.53028E9
Slope: 622.94589

Transect 4
Fit

Adj. R-Square: 0.99767
[CH4]_ppm

Value

Intercept: -1.2821E9
Slope: 521.91561

Transect 5
Fit

Adj. R-Square: 0.98455
[CH4]_ppm

Value

Intercept: -6.91781E8
Slope: 281.60915

Transect 6
Fit

Adj. R-Square: 0.99334
[CH4]_ppm

Value

Intercept: -1.20774E9
Slope: 491.64572

Transect 7
Fit

Adj. R-Square: 0.99919
[CH4]_ppm

Value

Intercept: -1.91793E9
Slope: 780.74625

Transect 8
Fit

Adj. R-Square: 0.99594
[CH4]_ppm

Value

Intercept: -2.74041E9
Slope: 1115.56134

Transect 9
Fit

Adj. R-Square: 0.99366
[CH4]_ppm

Value

Intercept: -2.50514E9
Slope: 1019.78731

Transect 10
Fit

Adj. R-Square: 0.99658
[CH4]_ppm

Value

Intercept: -2.75116E9
Slope: 1119.93628

Transect 11
Fit

Adj. R-Square: 0.99328
[CH4]_ppm

Value

Intercept: -1.93795E9
Slope: 788.89566

Transect 12
Fit

Adj. R-Square: 0.9989
[CH4]_ppm

Value

Intercept: -2.40767E9
Slope: 980.10821
Figure S4. Slopes for CO₂ chamber readings.
Figure S5. Contour showing the onset of penetrative mixing in temperature (bottom) and O$_2$ (top). Profiles were obtained at monitoring station located at lake center with a profiling CTD (YSI multiprobe) every 1 hour at 0.5 depth intervals.
Figure S6. August 24 – 25, 2013 high-resolution water column temperature profiles over the campaign period obtained with CTD profiler deployed from the boat. Profile intervals were approximately 10 – 15 cm.
Figure S7. Left: August 24 – 25, 2013 time series of CH₄, CO₂, O₂ and temperature from probes mounted to the side of the boat. Hashed areas are times when boat was underway or in the harbor. Nonhashed areas are while boat was drifting during transects (transect number indicated at top of plot) towards the middle of the lake. Right: Corresponding CH₄ profiles during the campaign.
Figure S8. Log$_{10}$ of dissipation ($\varepsilon$) rate of turbulent kinetic energy (TKE) in surface layer (1-2 meters) as a function of wind speed. Red dots were removed (first ~2 hours) from the correlation due to large divergence between the wind speed and turbulence as seen on Figure 1a. R$^2$ with all data points is 0.65.
Figure S9. Fluxes, $F_{\text{CH}_4}$ and $F_{\text{CO}_2}$ (top) as a function of the dissipation rate of turbulent kinetic energy (TKE).

- Linear Fit, $R^2 = 0.74$
- $F_{\text{CH}_4}$ (mmol m$^{-2}$ d$^{-1}$)
- Log$_{10}$ of Dissipation Rate of TKE (W kg$^{-1}$)
- Linear Fit, $R^2 = 0.67$
- $F_{\text{CO}_2}$ (mmol m$^{-2}$ d$^{-1}$)