Supporting Information for

A flux-gradient system for simultaneous measurement of the CH_4, CO_2 and H_2O fluxes at a lake-air interface

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This file includes:

Instrument calibration

In this research, the WS-CRDS analyzer's CO_2 and CH_4 measurements were calibrated twice. Before installation at the experimental site, they were calibrated in the laboratory against a standard CO_2 gas (490 ppm, National Primary Standard prepared by the National Institute of Metrology, China) and a standard CH_4 gas (3.05 ppm, National Primary Standard) on 2 May 2012. Prior to gain factor adjustments, the analyzer's reading was 486.4 ppm for CO_2 (absolute error -3.6 ppm, relative error -0.7%) and 3.02 ppm for CH_4 (absolute error -0.03 ppm, relative error -1.0%). Its internal gain factors were then adjusted so that its outputs matched the concentrations of these standards.

The second calibration was conducted at the field site on 22 June (DOY 174) 2012. In the case of CH_4, the same standard gas (3.05 ppm) was used, and the measurement of this standard was 3.00 ppm, with a relative error of -1.6%. In the case of CO_2, two standard gases (with concentrations of 490 and 381 ppm, National Primary Standard) were used, and the measurements of these gases were 486.4 and 378.3 ppm, with the same relative error of -0.7%.

The analyzer's water vapor measurement was calibrated on May 1, 2011 against a dew point generator (model LI-610, Li-Cor Inc., Lincoln, Nebraska, USA) at five dew point temperatures of 1, 5, 10, 15 and 18 ^{o}C. Prior to field deployment (on May 3, 2012), it was checked against the dew-point generator at four dew point temperatures of 1, 5, 10, and 15 ^{o}C. The relative bias of the water vapor mixing ratio measured by the two instruments was less than 0.4%. The dew point generator was not available during the field experiment.

Comparison against the independent vapor isotope analyzer indicates that the water vapor calibration was quite stable (Figure S1).

Figure S1. Comparison of the water vapor mixing ratio difference between the two air intake (at the height of 1.1 m and 3.5 m above the water surface) observed using the WS-CRDS and the vapor isotope analyzer.

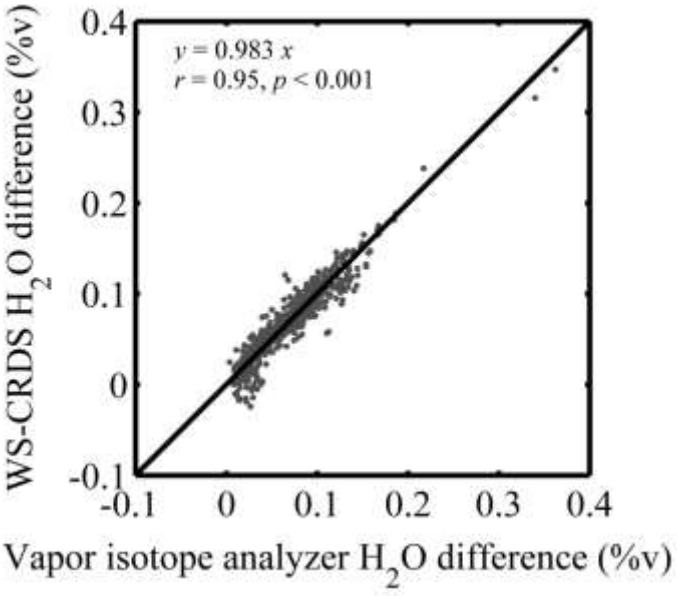

Figure S2. Comparison of CO_2 flux (F_c) and and CH_4 flux (F_m) calculated with the flux-gradient method and the modified Bowen-ratio method (open circles: daytime; closed circles: nighttime).

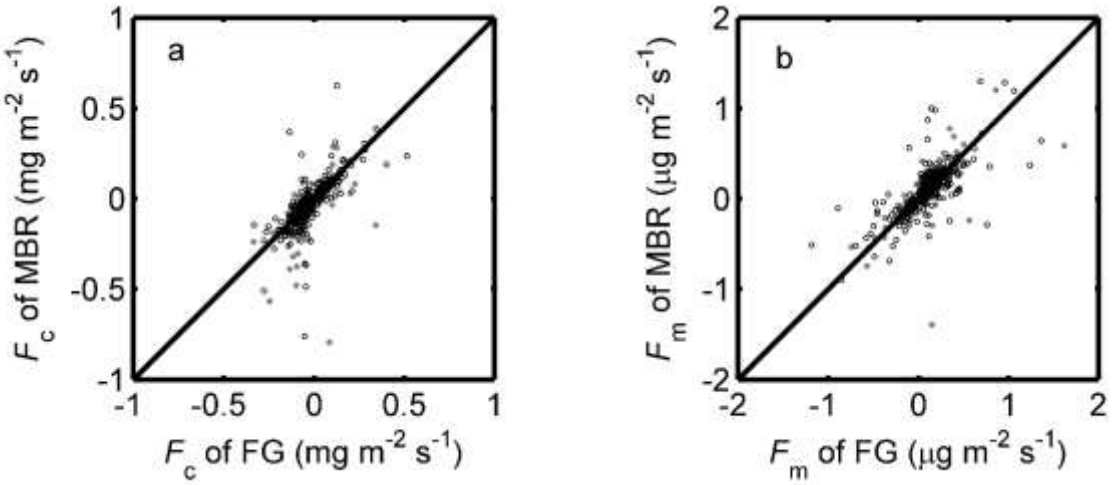

Table S1. Data used for the relationship between F_m and water temperature (T_w) in Figure 7. Observation methods include boundary layer model (BLM), inverted funnels (IF), floating chamber (FC) and eddy covariance (EC).

Markers	Type	Water bodies	Methods	References
Red squares	Oligotrophic water bodies	10 lakes in arctic Alaska	BLM	S1
		Polegar Lake, Brazil	BLM & IF	S2
Green triangles	Mesotrophic water bodies	small wetland lakes, western Siberia	FC	S3
		Hydroelectric reservior Porttipahta, Finland	FC & IF	S4
Grey dots	Eutrophic water bodies	Lake Calado, Amazon Basin	FC	S5
		Lake Kevaton, Finland	FC & IF	S6
		Priest Pot, UK	BLM	S7
		Rotsee, Switzerland	FC	S8
		Lake Taihu	FC	S9
		Donghu	FC	S10
		Dongtinghu, Poyanghu, Chaohu, Nansihu, Hongzehu, Fuxianhu, Erhai, Dianchi	FC	S11
		Biguás Lake, Brazil	BLM & IF	S2
		2 lakes and 3 floodplains, Pantanal, Brazil	FC	S12
		Hydroelectric reservior Lokka, Finland	FC & IF	S4
		Lake Wohlen, Switzerland hydropower reservoir	BLM & IF	S13
Dashed line	A eutrophic lake	Lake Suwa, Japan	BLM & IF	61
Solid line	A eutrophic hydropower reservoir	Lake Wohlen, Switzerland Hydropower reservoir	EC	S14

Table S2. Annual methane emission from lakes worldwide in the literatures (the box plot in Figure 7).

Continent: North America (NA), South America (SA), Europe (E), Asia (A), Australia (AU).

Lake name/Region	Continent	CH_4 emission ($g\ CH_4\ m^{-2}\ yr^{-1}$)	Reference
Amazonian floodplain lakes	SA	43.8	S15, S16
Big Soda	NA	0.2	S17
Bisen	E	0.2	S18
Biwa	A	1.6	S19
Brown	NA	3.5	S18
Canadian thaw ponds	NA	3.4	S20
Coleen	NA	0.5	S1
Constance	E	0.2	S21
Crampton	NA	1.8	S18
Crystal	NA	0.1	S22, S23
Crystal Bog	NA	1.2	S23
Dam	NA	0.7	S1
Dillon	NA	3.6	S24
East Long	NA	4.8	S18
Erie	NA	0.3	S25
Finnish lakes	E	0.4	S26
Fiolen	E	0.1	S18
Gransjön	E	0.8	S18
Grunnen	E	0.7	S18
Gyslättasjön	E	0.6	S18
Hummingbird	NA	2.3	S18
Illersjön	E	2.7	S27
Kasumigaura	A	1.5	S28
Kevätön	E	2.6	S6
Klintsjön	E	0.5	S18
L Sångaren	E	0.2	S18
Lago Calado	SA	24.4	S5, S29
Lake 227	NA	1.0	S30
Lake Donghu 1	A	127.0	S10
Lake Donghu 2	A	115.7	S10
Lake Donghu 3	A	130.5	S10
Lake Mendota	NA	2.9	S31
Lillsjön	E	0.1	S27
Ljustjärn	E	0.7	S18
Long	NA	0.2	S24
Lövtjärn	E	0.9	S18

Mäkijärvi	E	0.3	S6
Manaus 1	SA	256.0	S32
Manaus lake	SA	2576.9	S33
Marn	E	0.3	S27
Mendota	NA	2.9	S31
Mirror	NA	1.2	S34
Mono	NA	6.0	S17
Morris	NA	24.0	S18
N1	NA	0.1	S1
N2-cont	NA	0.8	S1
Nojiri	A	0.4	S35
North Gate	NA	2.0	S18
Onodaga	NA	5.8	S36
Ontario	NA	0.7	S37
Open water, Amazon flood plain	SA	9.3	S38
Orinico river flodplain lakes	SA	8.5	S39
Pantanal	SA	51.3	S40
Pass	NA	3.4	S24
Paul	NA	6.9	S18
Peter	NA	8.1	S18
Pond 386	NA	0.7	S1
Postilampi	E	14.0	S6
Priest Pot	E	74.4	S7
Rägastjärn	E	1.5	S18
Rainbow	NA	4.8	S24
Red Rock	NA	7.8	S24
Roach	NA	0.7	S18
Ryans 1 Billabong	AU	8.8	S41
Sangaren	E	0.3	S18
Searsville	NA	1.0	S17
Siberian taiga lake	NA	1.6	S3
Siberian taiga pond	NA	4.9	S3
Siberian thaw lakes	A	24.9	S42
Siberian tundra lake	NA	0.9	S3
Skärshultssjön	E	0.3	S18
Skottjärn	E	2.6	S18
Soap	NA	0.8	S17
Southern Hudson Bay Lowlands 1	NA	1.5	S32
Sparkling	NA	0.7	S23
Svarttjän	E	2.6	S18
Toolik Lake	NA	1.5	S1
Trout Bog	NA	2.4	S23
Tuesday	NA	4.7	S18
Vehmasjärvi	E	0.3	S6

Ward	NA	15.3	S18
William	NA	0.4	S1
Windy	NA	0.3	S1
Wintergreen	NA	181.2	S24
Wintergreen	NA	107.2	S43
Yukon delta	NA	10.5	S44

Table S3. Lake CO_2 data found in the literatures. Continent: North America (NA), South America (SA), Europe (E), Asia (A), Australia (AU), Antarctic (AT). Numbers in parentheses are mean of the upper and lower limits reported.

Lake name	**Continent**	**CO_2 flux ($mg\ m^{-2}\ s^{-1}$)**	**references**
Toolik Lake	NA	0.005	7
Soppensee	E	0.012	7
Mirror Lake	NA	0.003 – 0.006 (0.005)	14
Williams Lake	NA	-0.007 – 0.119 (0.056)	17
Lake Valkea-Kotinen	E	0.009 – 0.018 (0.014)	18
Lake Gårdsjön	E	0.05	29
Lake Mochou and Tuanjie	AT	-0.02	57
Lake Paajarvi	E	0.003 – 0.070 (0.037)	58
E1	NA	0.030	S1
NE14	NA	0.020	S1
Toolik	NA	0.018	S1
N1	NA	0.010	S1
Island	NA	0.009	S1
N2	NA	0.006	S1
S6	NA	0.004	S1
I8	NA	0.023	S1
NE2	NA	0.021	S1
Windy	NA	0.020	S1
Dam	NA	0.016	S1
Silhouette	NA	0.017	S1
Galibraith	NA	0.015	S1
Carolyn	NA	0.013	S1
S3	NA	0.011	S1
NE12	NA	0.008	S1
Africa	NA	0.008	S1
Charles	NA	0.007	S1
Coleen	NA	0.006	S1
Maxine	NA	0.003	S1

William	NA	0.002	S1
Bern	NA	0.002	S1
George	NA	0.001	S1
Anne	NA	0.000	S1
Camp	NA	-0.003	S1
Mtlake	A	0.036	S3
lake Postilampi	E	0.003 – 0.015 (0.009)	S6
Lake Heialämpi	E	0.007	S6
Lake Kevätön	E	0.007 – 0.008 (0.008)	S6
Lake Vehmasjärvi	E	0.006 – 0.014 (0.010)	S6
Lake Mäkijärvi	E	0.003 – 0.006 (0.005)	S6
Crystal Bog	NA	0.016	S6
Trout Bog	NA	0.023	S6
Crystal Lake	NA	0.000	S6
Sparkling Lake	NA	0.003	S6
Williams Lake	NA	0.0002	S6
Shingobee Lake	NA	0.019	S6
Mirror Lake	NA	0.003	S6
Four Lakes	NA	-0.033	S6
Priest Pot	E	0.020	S6
Lake Donghu	A	0.004	S10
Lake Daming	A	-0.005 – 0.002 (-0.002)	S45
Croche	NA	0.005	S46
Ours	NA	0.009	S46
Achigan	NA	0.006	S46
Blanche	NA	0.006	S46
Connelly	NA	0.006	S46
En Coeur	NA	0.002	S46
Gervais	NA	0.008	S46
Mt. Noire	NA	0.008	S46
du Nord.	NA	0.008	S46
Ouimet	NA	0.011	S46
Pin Rouge	NA	0.004	S46
Rouge	NA	0.006	S46
des Sables	NA	0.010	S46

St. Joseph	NA	0.006	S46
Tremblant	NA	0.007	S46
Truite	NA	0.010	S46
d'Argent	NA	0.014	S46
Bowker	NA	0.003	S46
Brome	NA	0.005	S46
Orford	NA	0.011	S46
Waterloo	NA	0.004	S46
La Peche	NA	0.009	S46
Phillippe	NA	0.005	S46
6 lakes within the Qu'Appelle River catchment	NA	-0.051 – 0.102 (0.026)	S47
Lake Valkea-Kotinen	E	0.013 – 0.022 (0.018)	S48
Lake Valkea-Kotinen	E	0.004	S49
Lake Pääjärvi	E	0.1 – 0.3 (0.2)	S50
Poyang Lake	A	-0.011 – 0.024 (0.007)	S51
Chilika Lake	A	0.025 – 0.092 (0.059)	S52
Lake Pääjärvi	E	0.014 – 0.017 (0.016)	S53
Lake Ormajärvi	E	0.006 – 0.007 (0.006)	S53
Lanca di Po	E	0.010 – 0.344 (0.177)	S54
large floodplain lake	SA	0.022 – 0.185 (0.104)	S55
Reeuwijkse plas	E	-0.002	S56
Vinkeveense plas	E	0.035	S56
Nieuwkoopse pls	E	0.012	S56
Belterwiede	E	0.024	S56
Schutsloterwiede	E	0.018	S56
Lake Neusiedl,	AU	0.00005 – 0.031 (0.016)	S57
151 Danish lakes	E	0.013 – 0.081 (0.047)	S58
Lake Merasjarvi	E	0.01	S59

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