nature geoscience
SUPPLEMENTARY INFORMATION
DOI: 10.1038/NGEO2654

Large contribution to inland water CO_2 and CH_4 emissions from very small ponds

Meredith A. Holgerson* and Peter A. Raymond

*Corresponding author: meredith.holgerson@gmail.com

CONTENTS:

S1. SUPPLEMENTAL SENSITIVITY ANALYSIS

S1.1 Inputs to Monte Carlo Analysis

Table S1.1. The surface area, CO_2, and CH_4 fluxes used to upscale to global carbon flux in the Monte Carlo analysis. As the sample size for CO_2 flux from lakes >100 km^2 was 1, we used the mean as the standard deviation.

Size Class (km^2)	Surface Area (km^2)	CO_2 flux (SD)	CH_4 flux (SD)
<0.001	147,763 – 861,578	35.18 (36.81)	2.275 (3.571)
0.001- 0.01	406,576	21.21 (27.56)	0.653 (0.725)
0.01 – 0.1	675,234	21.57 (19.44)	0.280 (0.428)
0.1 – 1	984,651	23.87 (31.80)	0.157 (0.340)
1 – 10	782,074	22.42 (12.61)	0.116 (0.426)
10 – 100	597,789	20.90 (12.90)	0.102 (0.217)
> 100	2,024,016	11.49	0.059 (0.106)

S1.2 Discussion of Monte Carlo Analysis

The Monte Carlo sensitivity analysis indicates that variation in CO_2 and CH_4 concentrations among lakes within a size class is responsible for the large coefficients of variation (Table S1.2).

Table S1.2. Results of Monte Carlo analysis to estimate global diffusive CO_2 and CH_4 flux from lakes and ponds.

Size Class (km^2)	CO_2 flux (Pg C yr^{-1})	CO_2 CV	CH_4 flux (Pg C yr^{-1})	CH_4 CV
<0.001	0.0890	92.0%	0.00643	111.1%
0.001- 0.01	0.0425	94.2%	0.00125	87.3%
0.01 – 0.1	0.0689	74.7%	0.00101	95.6%
0.1 – 1	0.0121	93.6%	0.00102	107.8%
1 – 10	0.0802	53.2%	0.00079	124.4%
10 – 100	0.0537	60.2%	0.00037	108.6%
> 100	0.1152	77.3%	0.00067	105.0%
TOTAL	**0.571**		**0.012**	
25 – 75th quantiles	**0.439 – 0.683**		**0.006 – 0.015**	

S1.3 Additional Monte Carlo sensitivity analysis

Because our study is the first to use the most recent satellite imagery analysis of global lakes and ponds > 0.002 km^2, our analysis uses different surface areas for the global distribution of lakes and ponds than previous studies[1]. To evaluate uncertainty between the range of surface areas used in our study and previous studies, we conducted a second Monte Carlo analysis that allowed for uncertainty in surface area in addition to the uncertainty in CO_2 and CH_4 flux described above. For surface area, we randomly and iteratively (n=1,000 runs) selected a number from a uniform distribution bound by Verpoorter et al.'s (2014) recent satellite analysis and the older estimates from Downing et al. (2006) (Table S1.3).

The results were similar to the Monte Carlo analysis that just incorporated uncertainty in gas concentration, indicating that uncertainty in surface area of lakes >0.001 km^2 is minimal (Table S1.3). The proportion of diffusive CO_2 and CH_4 emissions from small ponds is also similar: 16.2% (25 – 75th percentiles: 3.8 – 25.4%) and 40.7% (25 – 75th percentiles 0 – 68.4%).

Table S1.3. The surface area estimates used to upscale to global diffusive CO_2 and CH_4 flux in this second Monte Carlo analysis.

Size Class (km^2)	Verpoorter et al. 2014: Surface Area (km^2)	Downing et al. (2006): Surface area (km^2)	CO_2 Flux (Pg C yr^{-1})	CO_2 CV	CH4 flux (Pg C yr^{-1})	CH$_4$ CV
<0.001	147,763	861,578	0.088	94.7%	0.00636	113.7%
0.001- 0.01	406,576	692,600	0.059	92.0%	0.00175	86.4%
0.01 – 0.1	675,234	602,100	0.065	76.4%	0.00098	97.0%
0.1 – 1	984,651	523,400	0.090	98.3%	0.00075	108.7%
1 – 10	782,074	455,100	0.060	56.4%	0.00065	119.4%
10 – 100	597,789	392,362	0.043	60.1%	0.00030	113.7%
> 100	2,024,016	1,573,441	0.099	80.1%	0.00060	106.8%
TOTAL			**0.501**		**0.011**	
25 – 75th Quantiles			**0.375 – 0.610**		**0.006 – 0.015**	

S2. SUPPLEMENTAL DISCUSSION

S2.1 Uncertainty in k_{600} Values

Estimating k_{600} and using the same k_{600} for all lakes in a given size class introduces some error and uncertainty into our analysis. Air-water gas exchange, and the gas transfer velocity k_{600}, is largely driven by turbulence at the air-water interface[2]. In large lakes, turbulence is primarily generated by wind, and wind speed is often used to model k_{600}[3,4]. Yet, recent studies show that there is substantial spatial and temporal heterogeneity in wind speed over lakes that complicates estimating k_{600} from a single point measurement[5]. Additionally, the relationship between wind speed and k_{600} breaks down under low-wind conditions (~3 m s^{-1}), typical of small and sheltered ponds and lakes[4,6,7]. In these small lakes and ponds, convection tends to dominate turbulence production; yet, much uncertainty remains regarding k_{600} estimates from convection[8]. Specifically, k_{600} appears higher under periods of heat loss and particularly when the water temperature exceeds air temperature and creates instability at the air-water interface[8-11]. Periods of cooling and times when the water is warmer than the air occur at night, translating to higher overnight fluxes[12,13]. As the k_{600} values used in this study for ponds < 0.01 km^2 were daily averages and may not represent short-lived but significant increases in k_{600} due to heat loss, we may underestimate gas flux.

In addition to the problems estimating k_{600} mentioned above, recent studies using eddy covariance techniques to directly measure turbulent scalar flows from water bodies typically calculate larger k_{600} estimates than those predicted from gas concentrations and wind speed[11,14-16]. This suggests that our global flux estimates are underestimated; but more work will be needed to scale up eddy covariance measurements from single lakes to regional and global scales. It will be especially fruitful to compare eddy covariance k_{600} to wind and convection models across the entire lake size gradient. Overall, while our study uses commonly accepted methods to scale up to global gas flux, estimates will only be improved as technological advances and additional studies constrain k_{600} estimates.

S2.2 Uncertainty in Global Size Distribution of Very Small Ponds (< 0.001 km^2)

We used a lower and upper bound to incorporate uncertainty in the global size distribution of very small ponds (0.0001 – 0.001 km^2), estimating that these ponds comprise between 147,763 and 861,578 km^2 of surface area globally (see Methods). The upper bound is based on the Pareto Distribution, which can accurately estimate the number of lakes in some flat regions of Earth, but overestimates the number of lakes in mountainous regions[17]. The Pareto distribution has also been shown to fit the upper tail of the size distribution of lakes in arctic regions, but the relationship broke down at different size thresholds depending on the study region (thresholds: 30 m^2, 100 m^2, and 400 m^2)[18]. As less than 25% of the Earth's surface is considered mountainous[19], the Pareto Distribution may represent lake surface area for much of the globe, but does represent an upper bound.

The midrange estimate of our range is supported by a few small-scale studies using high-resolution satellite imagery or light-detection and ranging (LiDAR) data. Multiple studies throughout the northern and eastern United States found between 1 and 15 small ponds (~100 to 3,000 m^2) per km^2 of forested landscape[20-23]. Assuming this density across the globe and a mean surface area of 0.00027 km^2 would equate to between 35,921 to 538,812 km^2 of surface area covered by ponds between 0.0001 and 0.001 km^2. In the Northern Highland Lake District in Wisconsin, all lakes > 0.0001 km^2 have been mapped[24]. When the Pareto Distribution is modeled to these lakes, the β parameter was estimated to be -0.19[24] (in comparison to -1.06 used here based on Downing et al. (2006)). Using this β at a global scale yields 334,366 km^2 of surface area covered by ponds between 0.0001 and 0.001 km^2. While the density of very small ponds is likely to vary greatly across the globe, our range – particularly mid-range estimates—seems reasonable.

S2.3 Ebullition Analysis

We conducted a separate literature review for studies where both CH_4 ebullition and diffusion were directly measured from freshwater lakes and ponds. We compiled data from 47 water bodies, which ranged in size from 0.002 to 1.449 km^2 (Supplemental Table 2). Assuming ebullition occurred 365 days a year[25], we calculated annual CH_4 ebullition. The relationship between ebullition to surface area was weak on a square meter basis (linear regression, $R^2 = 0.19$, $p = 0.02$), which was largely driven by variability in ponds and lakes between 0.001 and 0.01 km^2. We also evaluated the possibility to estimate ebullition from diffusion, but there was no significant relationship between the ratio of diffusion and ebullition and lake surface area (linear regression, $R^2 = 0.002$, $p = 0.56$). This analysis was limited by low sample size and a small range in lake surface area; perhaps with more global data we will be able to upscale ebullitive flux from inland waters.

S.3 REFERENCES FOR SUPPLEMENTAL MATERIAL

1 Raymond, P. A. *et al.* Global carbon dioxide emissions from inland waters. *Nature* **503**, 355-359 (2013).

2 Jähne, B., Heinz, G. & Dietrich, W. Measurement of the diffusion coefficients of sparingly soluble gases in water. *J. Geophys. Res.* **92**, 10767-10776 (1987).

3 Wanninkhof, R. Relationship between wind speed and gas exchange over the ocean. *J. Geophys. Res.* **97**, 7373-7382 (1992).

4 Cole, J. J. & Caraco, N. F. Atmospheric exchange of carbon dioxide in a low-wind oligotrophic lake measured by the addition of SF_6. *Limnol. Oceanogr.* **43**, 647-656 (1998).

5 Schilder, J. *et al.* Spatial heterogeneity and lake morphology affect diffusive greenhouse gas emission estimates of lakes. *Geophys. Res. Lett.*, 2013GL057669 (2013).

6 Clark, J. F. *et al.* Gas transfer velocities for SF6 and ^{3}He in a small pond at low wind speeds. *Geophys. Res. Lett.* **22**, 93-96 (1995).

7 Read, J. S. *et al.* Lake-size dependency of wind shear and convection as controls on gas exchange. *Geophys. Res. Lett.* **39**, L09405 (2012).

8 Heiskanen, J. J. *et al.* Effects of cooling and internal wave motions on gas transfer coefficients in a boreal lake. *Tellus (B Chem. Phys. Meteorol.)* **66**, 22827 (2014).

9 MacIntyre, S., Romero, J. R. & Kling, G. W. Spatial-Temporal Variability in Surface Layer Deepening and Lateral Advection in an Embayment of Lake Victoria, East Africa. *Limnol. Oceanogr.* **47**, 656-671 (2002).

10 Zappa, C., Raymond, P., Terray, E. & McGillis, W. Variation in surface turbulence and the gas transfer velocity over a tidal cycle in a macro-tidal estuary. *Estuaries* **26**, 1401-1415 (2003).

11 Jonsson, A., Åberg, J., Lindroth, A. & Jansson, M. Gas transfer rate and CO2 flux between an unproductive lake and the atmosphere in northern Sweden. *J. Geophys. Res. Biogeosci.* **113**, G04006 (2008).

12 MacIntyre, S. *et al.* Buoyancy flux, turbulence, and the gas transfer coefficient in a stratified lake. *Geophys. Res. Lett.* **37**, L24604 (2010).

13 Podgrajsek, E., Sahlée, E. & Rutgersson, A. Diel cycle of lake-air CO2 flux from a shallow lake and the impact of waterside convection on the transfer velocity. *J. Geophys. Res. Biogeosci.* **120**, 2014JG002781 (2015).

14 Eugster, W. *et al.* CO2 exchange between air and water in an Arctic Alaskan and midlatitude Swiss lake: Importance of convective mixing. *J. Geophys. Res. Atm.* **108**, 4362 (2003).

15 Laurion, I. *et al.* Variability in greenhouse gas emissions from permafrost thaw ponds. *Limnol. Oceanogr.* **55**, 115-133 (2010).

16 Polsenaere, P. *et al.* Thermal enhancement of gas transfer velocity of CO2 in an Amazon floodplain lake revealed by eddy covariance measurements. *Geophys. Res. Lett.* **40**, 1734-1740 (2013).

17 Seekell, D. A., Pace, M. L., Tranvik, L. J. & Verpoorter, C. A fractal-based approach to lake size-distributions. *Geophys. Res. Lett.* **40**, 517-521 (2013).

18 Muster, S., Heim, B., Abnizova, A. & Boike, J. Water body distributions across scales: A remote sensing based comparison of three artic tundra wetlands. *Remote Sens.* **5**, 1498-1523 (2013).

19 Meybeck, M., Green, P. & Vörösmarty, C. A New Typology for Mountains and Other Relief Classes. *Mt. Res. Dev.* **21**, 34-45 (2001).

20 Brooks, R. T., Stone, J. & Lyons, P. An Inventory of Seasonal Forest Ponds on the Quabbin Reservoir Watershed, Massachusetts. *Northeast. Nat.* **5**, 219-230 (1998).

21 Palik, B. J., Buech, R. & Egeland, L. Using an ecological land hierarchy to predict seasonal-wetland abundance in upland forests. *Ecol. Appl.* **13**, 1153-1163 (2003).

22 Capps, K. A., Berven, K. A. & Tiegs, S. D. Modelling nutrient transport and transformation by pool-breeding amphibians in forested landscapes using a 21-year dataset. *Freshwater Biol.* **60**, 500-511 (2015).

23 Wu, Q., Lane, C. & Liu, H. An effective method for detecting potential woodland vernal pools using high-resolution LiDAR data and aerial imagery. *Remote Sens.* **6**, 11444-11467 (2014).

24 Hanson, P. C., Carpenter, S. R., Cardille, J. A., Coe, M. T. & Winslow, L. A. Small lakes dominate a random sample of regional lake characteristics. *Freshwater Biol.* **52**, 814-822 (2007).

25 Bastviken, D., Cole, J., Pace, M. & Tranvik, L. Methane emissions from lakes: Dependence of lake characteristics, two regional assessments, and a global estimate. *Global Biogeochem Cycles* **18**, GB4009 (2004).

S.4 REFERENCES FOR META-ANALYSIS

The following is the comprehensive list of studies used in the meta-analysis (25 studies, including 427 lakes and ponds).

1 Barber, T. R., Burke, R. A., Jr. & Sackett, W. M. Diffusive flux of methane from warm wetlands. *Global Biogeochem Cycles* **2**, 411-425 (1988).

2 Bartlett, K. B., Crill, P. M., Sass, R. L., Harriss, R. C. & Dise, N. B. Methane emissions from tundra environments in the Yukon-Kuskokwim Delta, Alaska. *J. Geophys. Res.* **97**, 16645-16660 (1992).

3 Bastviken, D., Ejlertsson, J. & Tranvik, L. Measurement of Methane Oxidation in Lakes: A Comparison of Methods. *Environ. Sci. Technol.* **36**, 3354-3361 (2002).

4 Bastviken, D., Cole, J., Pace, M. & Tranvik, L. Methane emissions from lakes: Dependence of lake characteristics, two regional assessments, and a global estimate. *Global Biogeochem Cycles* **18**, GB4009 (2004).

5 Casper, P., Maberly, S. C., Hall, G. H. & Finlay, B. J. Fluxes of methane and carbon dioxide from a small productive lake to the atmosphere. *Biogeochemistry* **49**, 1-19 (2000).

6 Fallon, R. D., Harrits, S., Hanson, R. S. & Brock, T. D. The role of methane in internal carbon cycling in Lake Mendota during summer stratification. *Limnol. Oceanogr.* **25**, 357-360 (1980).

7 Hamilton, J. D., Kelly, C. A., Rudd, J. W. M., Hesslein, R. H. & Roulet, N. T. Flux to the atmosphere of CH4 and CO2 from wetland ponds on the Hudson Bay lowlands (HBLs). *J. Geophys. Res.* **99**, 1495-1510 (1994).

8 Holgerson, M. A. Drivers of carbon dioxide and methane supersaturation in small, temporary ponds. *Biogeochemistry* **124**, 305-318 (2015).

9 Jonsson, A., Karlsson, J. & Jansson, M. Sources of carbon dioxide supersaturation in clearwater and humic lakes in northern Sweden. *Ecosystems* **6**, 224-235 (2003).

10 Kankaala, P., Huotari, J., Tulonen, T. & Ojala, A. Lake-size dependent physical forcing drives carbon dioxide and methane effluxes from lakes in a boreal landscape. *Limnol. Oceanogr.* **58**, 1915-1930 (2013).

11 Kling, G. W., Kipphut, G. W. & Miller, M. C. Arctic lakes and streams as gas conduits to the atmosphere: Implications for tundra carbon budgets. *Science* **251**, 298-301 (1991).

12 Kortelainen, P. *et al.* Sediment respiration and lake trophic state are important predictors of large CO2 evasion from small boreal lakes. *Glob. Change Biol.* **12**, 1554-1567 (2006).

13 Juutinen, S. *et al.* Methane dynamics in different boreal lake types. *Biogeosciences* **6**, 209-223 (2009).

14 Laurion, I. *et al.* Variability in greenhouse gas emissions from permafrost thaw ponds. *Limnol. Oceanogr.* **55**, 115-133 (2010).

15 Lennon, J. T., Faiia, A. M., Feng, X. & Cottingham, K. L. Relative importance of CO2 recycling and CH4 pathways in lake food webs along a dissolved organic carbon gradient. *Limnol. Oceanogr.* **51**, 1602-1613 (2006).

16 Miyajima, T. *et al.* Distribution of greenhouse gases, nitrite, and d13C of dissolved inorganic carbon in Lake Biwa: Implications for hypolimnetic metabolism. *Biogeochemistry* **36**, 205-221 (1997).

17 Natchimuthu, S., Panneer Selvam, B. & Bastviken, D. Influence of weather variables on methane and carbon dioxide flux from a shallow pond. *Biogeochemistry* **119**, 403-413 (2014).

18 Pelletier, L., Strachan, I. B., Garneau, M. & Roulet, N. T. Carbon release from boreal peatland open water pools: Implication for the contemporary C exchange. *J. Geophys. Res. Biogeosci.* **119**, 2013JG002423 (2014).

19 Repo, M. E. *et al.* Release of CO2 and CH4 from small wetland lakes in western Siberia. *Tellus B* **59**, 788-796 (2007).

20 Riera, J. L., Schindler, J. E. & Kratz, T. K. Seasonal dynamics of carbon dioxide and methane in two clear-water lakes and two bog lakes in northern Wisconsin, U.S.A. *Can. J. Fish. Aquat. Sci.* **56**, 265-274 (1999).

21 Schulz, M., Faber, E., Hollerbach, A., Schroder, H. G. & Gude, H. The methane cycle in the epilimnion of Lake Constance. *Archiv fur Hydrobiologie* **151**, 157-176 (2001).

22 Shirokova, L. S. *et al.* Biogeochemistry of organic carbon, CO2, CH4, and trace elements in thermokarst water bodies in discontinuous permafrost zones of Western Siberia. *Biogeochemistry* **113**, 573-593 (2013).

23 Smith, L. K. & Lewis, W. M., Jr. Seasonality of methane emissions from five lakes and associated wetlands of the Colorado Rockies. *Global Biogeochem Cycles* **6**, 323-338 (1992).

24 Striegl, R. G. & Michmerhuizen, C. M. Hydrologic influence on methane and carbon dioxide dynamics at two north-central Minnesota lakes. *Limnol. Oceanogr.* **43**, 1519-1529 (1998).

25 Utsumi, M. *et al.* Oxidation of dissolved methane in a eutrophic, shallow lake: Lake Kasumigaura, Japan. *Limnol. Oceanogr.* **43**, 471-480 (1998).