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Energy input is primary controller of methane bubbling in subarctic lakes

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Key Points:

- Continuous multiyear sampling shows that ebullition is a predictable process
- Ebullition is strongly correlated to various ice-free season energy flux proxies
- Shorter ice-covered seasons will alter methane emission from high-latitude lakes

Supporting Information:

- Readme
- Figure S1
- Figure S2
- Figure S3
- Text01

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Energy input is primary controller of methane bubbling in subarctic lakes

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Abstract Emission of methane (CH_4) from surface waters is often dominated by ebullition (bubbling), a transport mode with high-spatiotemporal variability. Based on new and extensive CH_4 ebullition data, we demonstrate striking correlations (r^2 between 0.92 and 0.997) when comparing seasonal bubble CH_4 flux from three shallow subarctic lakes to four readily measurable proxies of incoming energy flux and daily flux magnitudes to surface sediment temperature (r^2 between 0.86 and 0.94). Our results after continuous multiyear sampling suggest that CH_4 ebullition is a predictable process, and that heat flux into the lakes is the dominant driver of gas production and release. Future changes in the energy received by lakes and ponds due to shorter ice-covered seasons will predictably alter the ebullitive CH_4 flux from freshwater systems across northern landscapes. This finding is critical for our understanding of the dynamics of radiatively important trace gas sources and associated climate feedback.

1. Introduction

Studies of the numerically abundant small lakes and ponds across the globe and especially at high latitudes indicate their significant contribution to regional carbon (C) budgets [*Downing et al.*, 2006; *Cole et al.*, 2007; *Downing*, 2010]. Subarctic and Arctic environments appear to be exceptionally sensitive to climate change [*Rouse et al.*, 1997; *Serreze et al.*, 2000] and have already experienced greater than global average increases in air temperatures over the past decades [*Callaghan et al.*, 2010]. An important emission from a global warming perspective is methane (CH_4), with ebullition (bubbling) typically contributing more than 50% and sometimes 90% or more of the total CH_4 flux from lakes and ponds [*Bastviken et al.*, 2004, 2011]. Freshwater emissions of CH_4 have been estimated to be equivalent in greenhouse forcing to at least 25% of the carbon uptake of all land-based ecosystems combined [*Bastviken et al.*, 2011].

Methane ebullition from lakes and ponds is challenging to quantify and model due to temporal and spatial complexities that are highly episodic and variable among depth zones [*Walter et al.*, 2006; *Wik et al.*, 2011, 2013]. Scaling the contribution of CH_4 ebullition to local and regional flux estimates has been presumed to be difficult due to multiple controls over CH_4 production (e.g., total C substrate and quality, temperature, sediment porosity, and vegetation) [*Duc et al.*, 2010] and apparently heterogeneous release from lake sediments. By inference, reliable parameterizations of CH_4 ebullition should require knowledge of multiple in situ lake parameters that vary spatially and temporally. Here we address the challenge of high-spatiotemporal variability by using frequent, spatially dispersed measurements of CH_4 ebullition that span over four ice-free seasons [*Wik et al.*, 2013], and relate those measurements to energy related parameters (solar shortwave (SW) input, number of ice-free days, and shallow and deep water sediment temperature) which are readily measured.

2. Methods

The three lakes (Mellan Harrsjön, Inre Harrsjön, and Villasjön) encircle Stordalen Mire, a subarctic peatland complex 9 km east of Abisko in northern Sweden (68°21′N, 19°02′E, 350 m asl; Figure S1 in the supporting information). Mellan Harrsjön and Inre Harrsjön are 1.1 and 2.3 ha in area, reaching maximum depths of 7 and 5 m, respectively [*Wik et al.*, 2011]. Villasjön is larger (17 ha) though shallower with a maximum depth of 1.3 m

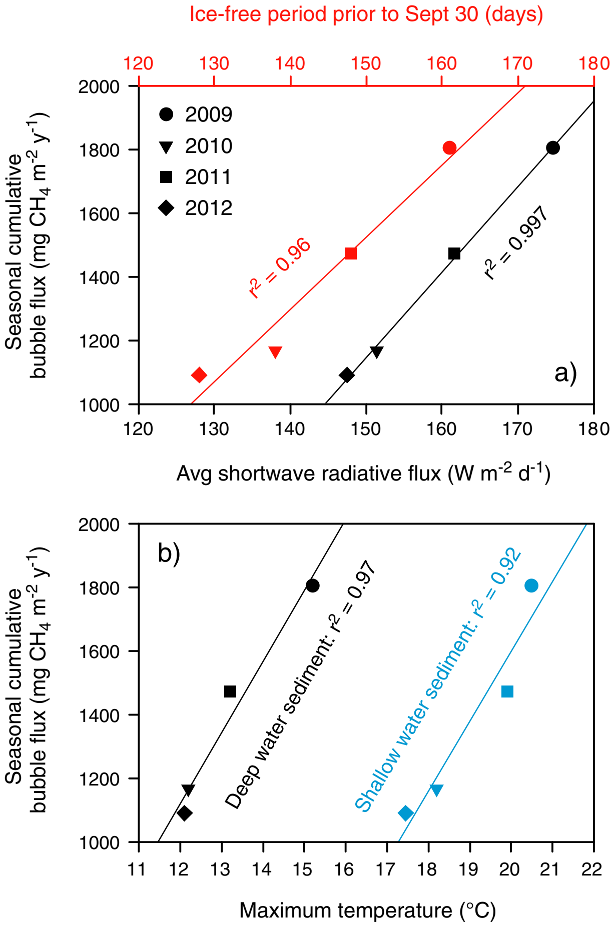

Figure 1. (a) Seasonal cumulative bubble flux of CH_4 from ice-out to 30 September versus length of ice-free period (red) and daily average SW irradiance (black) for the studied lakes until 30 September. (b) The whole-lake seasonal fluxes plotted against average maximum temperatures of surface sediments (black) in deep (Inre Harrsjön and Mellan Harrsjön) and shallow (all lakes) sites (blue) from June to September. Each seasonal cumulative flux number includes between 88 and 115 daily average fluxes (1023 to 2389 individual measurements) for all three lakes combined. See Table 1 for regression equations.

[*Jackowicz-Korczynski et al.*, 2010]. Although the study area has sporadic permafrost, the lakes are nonthermokarst features with no underlying permafrost and very limited erosion of organic material around their margins. At Stordalen Mire, warming above 0°C (the stability threshold for permafrost) during the past several decades has led to a shorter duration of ice cover [*Christensen*, 2004; *Kohler et al.*, 2006] and recent landscape alterations due to permafrost thaw [*Malmer et al.*, 2005].

The data used in this study include 6806 bubble CH_4 flux measurements made from June to September 2009–2012 using bubble traps that were systematically distributed across the various depths of the three study lakes (Figure S1). Shortwave radiation was measured over the surface of Inre Harrsjön and at the nearby Abisko Scientific Research Station (ANS) using pyranometers in net radiation sensors. Lake temperature data were collected year-round across the water column into the surface sediment at the deepest points of the lakes. Instrumentation, data collection methods, and data processing are described further in the supporting information and in *Wik et al.* [2013].

The ice-free period from ice-out until 30 September was chosen as a proxy for energy input. Ebullition was not measured in October and the vast majority of bubbles are released before the end of September (see supporting information and *Wik et al.* [2013]). We calculated the total SW radiation by summing daily ground level SW radiation averages. Daily average SW was calculated by dividing the total by the number of days between ice-out and 30 September. We report averages of the maximum deep water ($>$ 4 m) sediment temperature in Inre Harrsjön and Mellan Harrsjön, and the maximum shallow water ($<$ 2 m) sediment

Table 1. Equations for Regressions Displayed in Figures 1 and 2

Figure	Plot	*n*	r^2	*P* value	Equation
1a	Seasonal bubble CH_4 flux versus ice-free period length	4[a]	0.96	0.020	$y = 22.64x - 1871.2$
1a	Seasonal bubble CH_4 flux versus average SW radiative flux	4	0.997	0.001	$y = 26.83x - 2878$
1b	Seasonal bubble CH_4 flux versus deep water max sediment temperature	4	0.97	0.017	$y = 222.83x - 1552.6$
1b	Seasonal bubble CH_4 flux versus shallow water max sediment temperature	4	0.92	0.041	$y = 219.5x - 2790$
2a	Daily bubble CH_4 flux versus sediment temperature	16[b]	0.94	< 0.001	$y = -0.00036x^3 + 0.16x^2 - 0.94x + 1.48$
2b	ln[Bubble flux (mg CH_4 m^{-1} d^{-1})] versus 1 T/K	16	0.86	< 0.001	$y = -4 \times 10^{-5}x + 0.0036$

[a] Between 88 and 115 daily averages (1023 to 2389 individual measurements) for all three lakes combined were used to calculate each of the four seasonal cumulative bubble CH_4 flux numbers.
[b] 6806 individual flux measurements were binned into 16 temperature (1°C) intervals.

temperatures for all three lakes combined from ice-out to 30 September. The maximum temperature is reached for a short period of time but is a proxy for the total energy input required to reach that maximum.

3. Cumulative Bubble CH_4 Flux Versus Energy Input

The correlations between seasonal bubble CH_4 flux from the lakes and energy inputs during the ice-free season are robust ($r^2 \geq 0.92$; Figure 1) for any one of four easily measured energy-related parameters (regression equations are shown in Table 1): (1) number of ice-free days between spring ice-out and 30 September (Figure 1a), (2) daily average SW irradiance between ice-out and 30 September (Figure 1a), (3) maximum shallow water sediment temperature (Figure 1b), and (4) maximum deep water sediment temperature (Figure 1b).

When ice-out occurred later in the year because of cooler springs, the seasonal cumulative bubble CH_4 flux was lower ($r^2 = 0.96$; Figure 1a). Possible explanations include a delay in energy supporting primary production which leads to subsequent ebullition by contributing organic substrates [*Karlsson et al*., 2009], or, as we postulate here, simply a delay in sediment warming suitable for sufficient methanogenesis to sustain bubble formation (see Figure 2). Earlier ice-out has been recently noted at multiple sites in the Arctic and, along with an increase in the ice-free season length, it is an expected result of climate change [*Prowse et al*., 2011; *Karlsson et al*., 2013]. The earlier the ice left the lakes' surface, the sooner they began to store solar energy. Slight solar warming of the water directly under the thinning lake ice prior to ice-out was negligible compared to the amount of energy that enters the lake after ice-out. The seasonal bubble CH_4 fluxes were twice as high when ice-out occurred 30 days earlier (Figure 1a), pointing to the potential for significantly higher CH_4 fluxes with continued Arctic warming.

Daily average SW input after ice-out is observed to be the strongest single correlate with seasonal cumulative bubble CH_4 flux ($r^2 = 0.997$; Figure 1a) implying that gas formation and release depend on the heating from

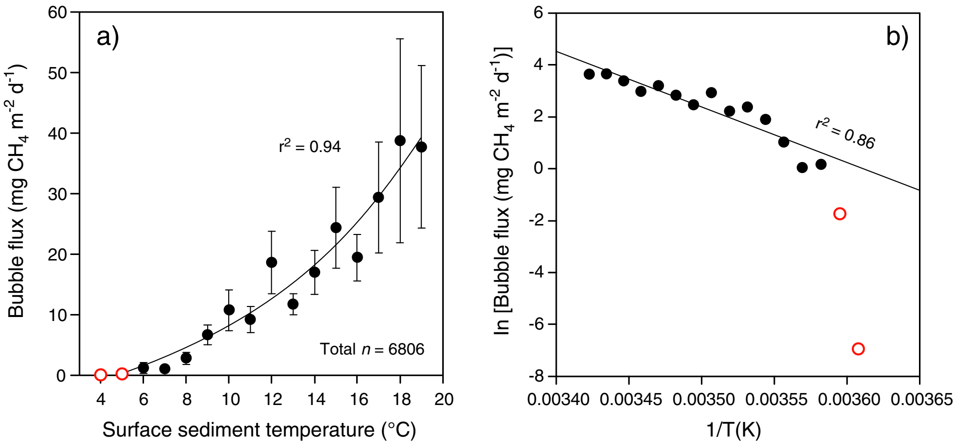

Figure 2. (a) Daily average bubble fluxes of CH_4 from start of ice-free season to 30 September versus average surface sediment temperature. All our 6806 individual bubble fluxes are binned into 1°C intervals (all lakes and depths combined). Error bars denote the 95% confidence interval. (b) Arrhenius-style plot of data from Figure 2a, with ln(bubble CH_4 flux) versus the inverse surface sediment temperature in K. Points marked in red are not encompassed by the linear correlation and correspond to the lowest surface sediment temperatures prior to the onset of ebullition; Q_{10} (7–17°C) = 14.

incoming SW. The heating in the deepest areas studied (7 m in Mellan Harrsjön) is on average 0.007°C per W m^{-2} of SW energy from the preceding day, with larger effects in shallower regions. A similarly strong correlation was observed for seasonal cumulative (as opposed to daily average) SW radiation after ice-out and seasonal cumulative bubble CH_4 flux (Figure S2).

Although CH_4 ebullition strongly correlates with maximum sediment temperature for both shallow (< 2 m) and deep (> 4 m) water sediments ($r^2 = 0.92$ and 0.97; Figure 1b), the seasonal timing of ebullition from shallow and deep sediments is different [*Wik et al.*, 2013]. Methane production is temperature dependent [*Boylen and Brock*, 1973; *Zeikus and Winfrey*, 1976]. The maximum temperature experienced by methanogenic archaea, responsible for CH_4 production in the sediments, is represented by near-surface sediment temperatures. Fluxes from shallow zone sediments, which recharge with gas first following warming in spring, was initially higher than fluxes from deep water sediments. Correspondingly, in early autumn, sediments in shallow areas lose heat sooner than sediments at depth, concurrent with a decrease in shallow zone ebullition [*Wik et al.*, 2013].

Both shallow and deep zones produced bubbles predictably depending on maximum surface sediment temperature (Figure 1b). Over the season there was an approximate threefold higher CH_4 flux in the shallow zones (Figure S3) [*Wik et al.*, 2013]. The minimum sediment temperature in the deeper lakes (Inre and Mellan Harrsjön) was reached during ice-out in spring. Consequently, cumulative bubble CH_4 flux over each season depends on the period between ice-out and the autumn decrease of sediment temperatures and incident SW radiation (similar to the number of sunny days or the inverse of cloudiness) over the same period. This dependency was reflected by the near doubling in cumulative bubble CH_4 flux from the coldest to warmest maximum sediment temperatures in both shallow and deep zones (Figure 1b). Correlations for sediment temperatures in shallow (< 2 m water depth) and deep water, analogous to Figure 1b, but with separated shallow and deep region fluxes, are shown in Figure S3.

4. Sediment Heating and Onset of Ebullition

Daily average CH_4 ebullition correlates exponentially with surface sediment temperature ($r^2 = 0.94$; Figure 2a). However, fluxes measured in early June, depicted with red points in Figures 2a and 2b, do not follow the trend. There is essentially no ebullition at temperatures below 6°C, but ebullition increases exponentially and is strongly correlated with temperatures above 6°C. Such behavior strongly suggests that the energy (as temperature) is exerting a control on the biological production of CH_4, instead of physical processes such as wintertime depletion of CH_4 in sediments [*Wik et al.*, 2011], and increased CH_4 solubility at the low temperatures [*Yamamoto et al.*, 1976]. This low temperature suppression of methanogenic activity [*Zeikus and Winfrey*, 1976] persists until sediment temperatures increase sufficiently so that temperature-controlled biological processes recharge the sediments with gas and CH_4 concentrations exceed solubility. After this onset of ebullition, its rate closely follows an Arrhenius-type curve ($r^2 = 0.86$; black points in Figure 2b) because additional biological CH_4 production can only go into the gas phase.

The strong correlations of CH_4 ebullition with the four energy flux proxies indicate that our results can be generalized. Indirect effects of changes in organic loading on ebullition are anticipated, but for our study lakes accumulation rates of organic C are low (15 g m^{-2} yr^{-1})[*Kokfelt*, 2009], and therefore, it was not necessary to invoke C loading to explain the between-year variation in ebullition. Sediment organic matter comes from both the catchment and in-lake primary production. Increases in dissolved organic C, likely with hydrological changes in northern latitudes [*Olefeldt and Roulet*, 2012; *Laudon et al.*, 2013], would darken the water and decrease light penetration causing a reduction in sediment temperatures at depth and a reduction in mixed layer thickness and thus a shallower region with warmer sediments [*Houser*, 2006]. Our data show that energy input is the primary driver for within and between-year variability of ebullition in three nearby lakes in the same catchment. At other sites, however, organic loading and quality may combine with energy inputs to influence the overall potential for CH_4 production and release.

5. Global Implications

The sporadic permafrost zone, which Stordalen Mire represents, has the highest global lake area fraction across previously glaciated northern environments [*Smith et al.*, 2007]. Hence, we believe our study lakes, although they are of nonthermokarst origin, are representative of the many shallow lakes and ponds across subarctic and Arctic regions. Using a lake area estimate for the region $> 66°$N from *Bastviken et al.* [2011], our 4 years of bubble CH_4 data, and assuming our observed range of SW energy input, the extrapolated range of annual pan-Arctic CH_4 ebullition from shallow lakes is 0.31 to 0.68 Tg yr^{-1}. Based on the relationships we present here, the high end of this range is likely to increase if Arctic warming causes longer ice-free seasons that allow for changes in the energy received by lakes and ponds. A higher value of 6.4 Tg yr^{-1} reported previously [*Bastviken et al.*, 2011] is heavily influenced by large point source fluxes observed in Siberian thermokarst lakes [*Walter et al.*, 2006]. However, not all tundra lakes and ponds emit large point source fluxes [*Repo et al.*, 2007; *Golubyatnikov and Kazantsev*, 2013]. Isotope studies indicate that different CH_4 production processes are responsible for highly persistent gas releases due to underlying C-rich taliks, hydrates, and geological faults [*Walter et al.*, 2008; *Anthony et al.*, 2012]. Given the large difference between these two extrapolations, a more accurate estimate of pan-Arctic lake emission will need both heat flux mediated CH_4 ebullition and a more accurate representation of lake morphologies that represent the variety of CH_4 production pathways and emission.

6. Conclusions

Accurate predictions of CH_4 ebullition and its controls can be obtained using near-continuous sampling over multiple years combined with local site meteorological observations. This result contrasts with earlier studies indicating that lake CH_4 ebullition is sporadic and difficult to predict. Further, we suggest that the recharge of gas in the sediment and subsequent bubble release primarily depends on the heat gained at the sediment-water interface over the summer. For our studied lakes, solar radiation and number of ice-free days are good proxies for CH_4 ebullition. The ability to correlate bubble CH_4 fluxes to energy parameters which can be obtained by meteorological networks and/or remote sensing enables improved modeling of regional and global C emissions from water bodies. While further work is needed to determine the overall ebullition potential of lakes with different characteristics, this study contributes a novel way to assess and predict temporal variability of gas venting from lakes and ponds.

Author Contributions

M.W. and B.T. contributed equally to this work. M.W. designed the ebullition sampling strategy, collected and analyzed samples, analyzed CH_4 and temperature data, and contributed to the manuscript. B.T. wrote most of the manuscript and analyzed SW and temperature data. P.C. organized possibilities for long-term measurement campaigns and contributed to ebullition sampling strategy design and data interpretation. D.B., R.V., and S.M. contributed to data analysis and interpretation. All authors assisted with preparing and editing the manuscript.

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