Temperature and sediment properties drive spatiotemporal variability of methane ebullition in a small and shallow temperate lake

Leandra Stephanie Emilia Praetzel, Marcel Schmiedeskamp, Klaus-Holger Knorr

Ecohydrology and Biogeochemistry Research Group, Institute of Landscape Ecology, University of Münster, Münster, Germany

Abstract

Ebullition is a major pathway of methane (CH₄) fluxes from lakes to the atmosphere. Small and shallow lakes can have high emissions but have only recently gained more attention. We studied the quantity and spatiotemporal variability of CH₄ ebullition from a small (1.4 ha) and shallow (max 1.5 m) temperate lake in Germany during 2017 and 2018. We found a high range of fluxes (0–872 mg m⁻² d⁻¹) and >90% of the fluxes were emitted between May and August. Fluxes in early spring and late autumn were below 4 mg m⁻² d⁻¹. Also, on a spatial scale, fluxes varied distinctly and generally increased from the shore to the center of the lake. To identify drivers of observed emissions patterns, we measured temperature and air pressure, the quantity and quality of the sedimented organic matter (OM) as well as chemical and physical properties of the sediment. Generalized linear models identified temperature, sediment porosity and organic matter content as the best predictors for the observed spatiotemporal differences, whereas temperature was accountable for the observed temporal, and porosity and organic matter content for the spatial variability. We suggest that in small lakes, temperature could serve as master variable to predict site-specific CH₄ ebullition while spatial within-lake differences are determined by varying physical sediment properties more than quantity or quality of the OM.

Lakes have gained increased attention due to their significant emissions of the potent greenhouse-gas methane (CH₄) to the atmosphere. It is estimated that on a global scale, lakes, and reservoirs emit 0.94–1.7 Pg C-CO₂-eq. of CH₄ to the atmosphere per year (DelSontro et al. 2019), and emissions are suspected to increase in the future as a consequence of global warming (Wik et al. 2016b; Aben et al. 2017). However, measurements of CH₄ emissions from lakes worldwide remain insufficient as well as the understanding of the biological, chemical, and physical drivers of both the temporal and spatial variabilities of CH₄ emissions (Natchimuthu et al. 2016; Saunois et al. 2016; Wik et al. 2016a).

Small lakes (<0.1 km²) were underestimated in the past regarding their abundance, the land surface area they cover, and thus in their potential contribution to global CH₄ budgets. They cover an area of the same extent as the largest lakes (> 10,000 km²), which is equivalent to 10¹² km² or <1% of the global land surface (Downing 2010; Holgerson and Raymond 2016). Moreover, small lakes have been estimated to contribute 75% of diffusive and 72% of ebullitive CH₄ emissions from lakes worldwide (Holgerson and Raymond 2016). Small lakes are usually shallower, have a shorter water residence time and a higher perimeter to volume ratio than larger lakes. Therefore, the overall carbon turnover is more intense and they are expected to react more sensitively to changing environmental and climatic conditions (Wetzel 1992; Downing 2010). In small lakes, water column and sediments in summer months warm much faster compared to deep lakes (Jankowski et al. 2006). Yet, there exist comparably few studies on CH₄ emissions from small and shallow lakes with most studies conducted in northern regions (Bastviken et al. 2011 and references herein; Wik et al. 2016b and references herein; see Supporting Information Fig. S1).

Three pathways play a major role in the emission of CH₄ from shallow lakes: plant-mediated, diffusive, and ebullitive transport (Bastviken et al. 2004). Plant-mediated CH₄ emissions play a major role in littoral areas and their magnitude is highly dependent upon the plant species (Bergström et al. 2007). Diffusive emissions follow a concentration
The challenge in assessing CH4 ebullition is the high variability, both on a spatial and temporal scale (Varadharajan and Hemond 2012). On a temporal scale, CH4 fluxes can vary diurnally and seasonally—patterns which have been explained by changes in temperature, pressure, wind, precipitation, or water depth, as well as by thawing events in spring or convective mixing of the water column (Huttunen et al. 2003; Podgrajsek et al. 2014; Natchimuthu et al. 2016). Despite the numerous possible drivers of temporal variability, many studies have emphasized temperature and air pressure as master variables that determine seasonal CH4 ebullition (Mattson and Likens 1990; Varadharajan and Hemond 2012; Wik et al. 2013). On a spatial scale, within-lake patterns of CH4 ebullition can be considerable. Drivers for spatial patterns of CH4 ebullition are not well studied, but water depth and organic matter (OM) input might control CH4 ebullition (e.g., in areas of stream inlets) (DeSonstro et al. 2011; Natchimuthu et al. 2016). In most cases, CH4 ebullition is higher in the littoral parts of lakes and decreases with increasing water depth as (1) the availability of easily degradable OM fueling sediment methanogenesis and subsequent emission is higher, (2) the hydrostatic pressure of the shallower water column on the sediment is lower which facilitates bubble release, or (3) the residence time of ascending bubbles in the shallower water column is shorter which lowers the oxidation potential (Bastviken et al. 2008; Wik et al. 2013; Natchimuthu et al. 2016). Small lakes differ from larger lakes with regard to their relatively greater littoral zone and thereby enhanced interchange with the watershed, their greater wind-protected morphometry and their stronger benthic-pelagic coupling due to lower water volumes (Palik et al. 2001; Tessier and Woodruff 2002; Søndergaard et al. 2005). This can result in different stratification dynamics and complicates the differentiation between littoral and pelagic zones, and patterns of CH4 ebullition in small lakes likely differ from larger lakes.

CH4 in lakes is mainly produced in the anoxic lake sediment. Although several studies pointed out the importance of aerobic CH4 production in the water column of larger and deeper lakes, production rates were in the range of nanomoles to micromoles per unit of volume presumably making it a minor contribution in small and shallow waters (Bogard et al. 2014; Hartmann et al. 2020). Methane in sediments is formed as the end product of anaerobic degradation of OM and is produced in significant amounts when alternative organic and inorganic electron acceptors (NO3⁻, Mn₄⁺, Fe³⁺, humic substances, SO₄²⁻), the reduction of which is thermodynamically more efficient for microorganisms than methanogenesis, are sufficiently depleted so that they operate close to their free energy thresholds (Blodau 2011). Besides redox conditions, CH4 production is also dependent upon the activity of enzymes, the microbial community, the availability of substrates such as H₂ and acetate as its direct precursors from fermentation, and on the degradability and quality of the OM (Updegraff et al. 1995; McClatchey and Reddy 1998; Fenchel et al. 2012). Notable CH4 production in sediments is usually limited to the upper few centimeters (Zepp Falz et al. 1999; Wilkinson et al. 2015). The amount and composition of sedimented material that reaches the upper parts of the sediment therefore plays a crucial role for CH4 production rates; studies from rivers and reservoirs have shown that high sedimentation rates of easily degradable OM can cause high CH4 production and concomitantly high ebullition rates (Sobek et al. 2012; Wilkinson et al. 2015). While demonstrated for selected rivers and large lakes, we are not aware of any studies that compare sedimentation and CH4 ebullition rates in small and shallow lakes.

Also, the OM quality, that is, the abundance of labile OM compounds, determines CH4 production rates in anoxic environments. Labile compounds such as polysaccharides or proteins are preferentially decomposed by microorganisms, whereas more recalcitrant structures such as cellulose, lignin, or humic substances residually enrich in the sediment (Updegraff et al. 1995; Tfaily et al. 2014; Tittel et al. 2019). To identify such effects and to characterize OM, Fourier-transformed infrared (FTIR)—spectroscopy provides a valuable tool to identify functional moieties of OM and can therefore provide information on the decomposition state or quality of OM (Broder et al. 2012). Few studies have investigated the influence of the OM source on the spatial variability of sediment CH4 production in large reservoirs (e.g., Berberich et al. 2019), but to our knowledge, there exist no such studies from small lakes.

While biogeochemical factors determine CH4 production in sediments, for evolution and storage of bubbles, the physical sediment structure is crucial. Dependent upon the sediment texture, gas bubbles do not only have different shapes (Boudreau et al. 2005), they also form by different pathways and the intensity of ebullition differs between sediment types. Laboratory experiments revealed that in sandy sediments, bubbles form by capillary invasion whereas in fine, clay-dominated sediments, bubbles are formed by displacing the surrounding sediment, which creates effective conduits for ebullition (Liu et al. 2016, 2018). Ostrovsky and Tęgowski (2010) showed that there exists a remarkable spatial pattern of sediment grain size distribution within one lake, potentially contributing to the high variability in ebullition. It is unclear whether similar spatial patterns in sediment grain size exist in small lakes.

Despite an increasing number of measurements on CH4 ebullition from lakes worldwide, small lakes, especially in the temperate zone, are still underrepresented as the higher
quantity of studies on (small) lakes is conducted in boreal regions (e.g., Bastviken et al. 2004; Wik et al. 2013; DelSontro et al. 2016). While drivers of temporal variability have been extensively studied (e.g., Natchimuthu et al. 2016), the understanding and in situ measurements of factors that drive spatial in-lake variability remain scarce (Wik et al. 2018).

To clarify whether patterns that were observed for CH₄ ebullition, sedimentation rates, or sediment properties from larger lakes also apply to small and shallow lakes, we measured CH₄ ebullition, sedimentation rates, properties of the sedimented material and physical sediment structure in a small and shallow lake in 2017 and 2018 to determine (1) spatiotemporal variability of CH₄ ebullition and sedimentation rates and (2) drivers of CH₄ ebullition. We hypothesize that (1) despite their small surface area a distinct spatial pattern of CH₄ ebullition also exists in a small lake, (2) CH₄ ebullition closely follows temperature changes and is more sensitive to high temperatures in a small lake compared to larger lakes, (3) spatial heterogeneity of CH₄ ebullition is thereby driven by a combination of quantity and quality of sedimented OM and physical sediment structure.

**Materials and methods**

**Study site**

Lake Windsborn is a small (1.41 ha) and shallow (max. depth 1.7 m) polymeric crater lake in the Eifel uplands in Southwest Germany. It is nearly circular and surrounded by a 20-30-m-high rampart, resulting in a very small catchment area (~ 8 ha) and the absence of surficial inflows and outflows (Praetzel et al. 2020 and references herein). During 2017 and 2018, Lake Windsborn was in a mesotrophic state with average measured pH values of 6.8 ± 0.8, conductivity of 19.4 ± 1.7 μS cm⁻¹, 9.7 ± 0.9 mg L⁻¹ of oxygen, 13.7 ± 2.3 mg L⁻¹ of dissolved organic carbon, 1.0 ± 1.9 mg L⁻¹ of total nitrogen, and 0.1 ± 0.1 mg L⁻¹ of total phosphorous (Praetzel et al. 2020).

**Methane ebullition measurements and flux calculations**

Methane ebullition was measured with inverse funnel traps as described in Wik et al. (2013). We used high-density polyethylene (PE) funnels in two different sizes: smaller ones with a diameter of 28.5 cm for shallow depths and larger ones with a diameter of 38.5 cm where the water column was deeper (for more details, see Supporting Information Fig. S2; Text S1).

Funnel traps were deployed at 13 sites within the lake to cover four different depth categories (50: < 50 cm, 100: 50–100 cm, 125: 100–125 cm, and 150: 125–150 cm water depth) in three transects from the shore to the center of the lake, to account for spatial heterogeneity (see Fig. 1). Methane ebullition was measured in 2017 and 2018 from May to November and April to October respectively in a biweekly cycle as a 24 h average or as a 2-week average when fluxes were very low in early spring and late autumn. To this end, funnels were filled with lake water at the start of the measurement and deployed just below the water surface. At the end of the measurement, the volume of the trapped gas was read from the scale of the syringes/cylinders and a sample was taken for gas concentration analysis. Samples were collected in 60-mL polypropylene syringes (Omniflex®, B. Braun, Melsungen, Germany), stored at 5°C and analyzed for concentrations of CH₄ and carbon dioxide (CO₂) the following day with a gas chromatograph (8610 GC-TCD/FID, SRI Instruments, Torrance, California) equipped with a flame ionization detector and methanizer. Gas mixtures of known concentrations (10%, 25%, and 50% vol. CH₄ and CO₂) were used for daily calibration. Concentration loss due to overnight storage was determined to amount to 5% and was accounted for by adding that loss rate to the measured concentrations. Methane bubble flux \( F \) (mg m⁻² d⁻¹) was calculated as follows:

\[
F (\text{mg m}^{-2} \text{d}^{-1}) = \frac{c \times P_{\text{field}} \times 0.101325 \times V}{A \times \Delta t \times R \times T_{\text{water}}} \times 10^{-3} \times M_{\text{CH}_4},
\]

where \( c \) is the measured CH₄ concentration in ppm, \( P_{\text{field}} \) is the atmospheric pressure during sampling in atm, \( V \) is the measured gas volume in the syringes/measuring cylinders in L, \( A \) is the funnel basal area in m², \( \Delta t \) is the funnel deployment time in days, \( R \) is the universal gas constant \( 8.314 \text{ kg m}^2 \text{s}^{-2} \text{ mol}^{-1} \text{ K}^{-1} \), \( T_{\text{water}} \) is the water temperature during sampling in K, \( M_{\text{CH}_4} \) is the molar mass of CH₄ 16.0425 g mol⁻¹ and 0.101325 (dimensionless) is the conversion factor from ppm to kg m⁻² s⁻².

According to Wik et al. (2013) and Harrison et al. (2017), CH₄ redissolution from the trapped gas in the funnel into the water is negligible if samples are collected within 3 d or if the collected gas volume is > 40 mL. As 24-h measurements were not possible at few dates, our calculated flux for these occasions is probably underestimated and therefore represents a conservative estimate of CH₄ ebullition in spring and autumn.

**Sedimentation rates and properties of the sedimented material**

We determined sedimentation rates, quality, and composition of the sedimented material with sediment traps at 10 sites within the lake (see Figure 1) in 2017 and 2018. Sites with water depths < 50 cm were omitted as the water column was too shallow for proper installation of the traps. In 2017, sediment traps were emptied irregularly and therefore only used to determine sediment properties. In 2018, sediment traps were emptied every 4 weeks to additionally determine sedimentation rates. Sediment traps were composed of three 30-cm-long polyvinyl chloride (PVC) tubes (diameter 5.2 cm) arranged in a circle and were positioned 1 m above the sediment surface with the help of a buoy. To minimize the influence of the mooring on sedimentation rates, sediment traps were mounted analogously to bubble traps (see Supporting Information Fig. S2).

To determine sedimentation rates, tubes were emptied into PE bottles, transported to the laboratory at 5°C and subsequently freeze-dried (Alpha 1–4 LPplus, Christ, Osterode, Germany) and analyzed for concentrations of CH₄ and carbon dioxide (CO₂) the following day with a gas chromatograph (8610 GC-TCD/FID, SRI Instruments, Torrance, California) equipped with a flame ionization detector and methanizer. Gas mixtures of known concentrations (10%, 25%, and 50% vol. CH₄ and CO₂) were used for daily calibration. Concentration loss due to overnight storage was determined to amount to 5% and was accounted for by adding that loss rate to the measured concentrations. Methane bubble flux \( F \) (mg m⁻² d⁻¹) was calculated as follows:

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\]
Germany). Freeze-dried samples were weighed, and sedimentation rates calculated as follows:

$$\text{Sedimentation rate } \left( \text{g m}^{-2}\text{d}^{-1} \right) = \frac{m_{\text{dry}}}{\Delta t \times A},$$

(2)

where $m_{\text{dry}}$ is the dry mass of the sedimented material in g, $\Delta t$ is the deployment time of the sediment trap in days, and $A$ is the total area of the three tubes in m$^2$.

After weighing, each three replicates were homogenized, ground in a ball mill (Mixer Mill MM 400, Retsch, Haan, Germany) and analyzed for elemental composition and OM quality (for more details, see Supporting Information Text S2). Elemental and isotopic composition of carbon (C) and nitrogen (N) were determined with isotope-ratio mass spectroscopy (IRMS, Eurovector EA3000 coupled with Nu Instruments Nu Horizon, Hekatech, Wegberg, Germany). Results are reported in mass % for C and N contents and in ‰ vs. VPDB (Vienna Pee Dee Belemnite) or AIR (atmosphere of earth) for $\delta^{13}$C and $\delta^{15}$N isotopic signatures, respectively. Total phosphorous (P), sulfur (S), manganese (Mn), and iron (Fe) contents were determined by wavelength dispersive X-ray fluorescence (WD-XRF; ZSX Primus II, Rigaku, Tokyo, Japan). Abundances of major functional moieties of OM were identified by FTIR spectroscopy (Cary 670 FTIR Spectrometer, Agilent, Santa Clara, California).

Physical sediment properties

To determine OM content, water content, and porosity of the lake sediment, 13 sediment cores were collected in August 2019 from each site with a gravity corer (UWITECH, Mondsee, Austria) in 60-cm-long PVC tubes with a diameter of 5.8 cm. The upper 5 cm of each core were sampled by cutting on-site, transferred into 250-mL PE bottles, and transported cooled at $\sim 5^\circ\text{C}$ to the laboratory. Samples were freeze-dried and loss on ignition (LOI) was determined in a furnace at $550^\circ\text{C}$ for 4 h. Sediment water content ($w$) was calculated as the difference of weight before and after freeze-drying; OM content was defined as LOI:

$$w \text{(weight)} = \frac{m_{\text{wet}} - m_{\text{dry}}}{m_{\text{wet}}} \times 100,$$

(3)

where $m_{\text{wet}}$ and $m_{\text{dry}}$ are the sediment masses before and after freeze-drying in g.

$$\text{OM content (weight)} = \frac{m_{\text{dry}} - m_{\text{LOI}}}{m_{\text{dry}}} \times 100,$$

(4)

where $m_{\text{dry}}$ and $m_{\text{LOI}}$ are the sediment masses before and after LOI determination in g.

Porosity $\phi$ was then estimated as follows (Lewandowski 2002):

$$\phi = \frac{\rho_{\text{wet}}}{\rho_{\text{H}_2\text{O}}} \times w,$$

(5a)

with

$$\rho_{\text{wet}} = \frac{1}{\frac{1}{\rho_{\text{org}}} + \frac{1 - w}{\rho_{\text{dry}}}},$$

(5b)

and

$$\rho_{\text{dry}} = \frac{1}{\frac{1}{\rho_{\text{org}}} + \frac{1}{\rho_{\text{min}}}}.$$
where \( \rho_{\text{wet}} \) is the density of the wet sediment in g cm\(^{-3} \), \( \rho_{\text{H2O}} \) is the density of water at 19.25\(^\circ\)C (in situ sediment temperature during sampling, 0.998359 g cm\(^{-3} \)), \( w \) is the water content in % weight/100, \( \rho_{\text{dry}} \) is the density of the dry sediment in g cm\(^{-3} \), \( g \) is the LOI/100, \( \rho_{\text{org}} \) is the density of the organic substance 1.4 g cm\(^{-3} \), and \( \rho_{\text{min}} \) is the density of the mineral components 2.65 g cm\(^{-3} \) (Blum 2012).

Other environmental parameters

Air, water and sediment temperature and air pressure were continuously (logging interval 10 min, sampling interval 1 min) measured by a weather station (HOBO RX 3000 Remote Monitoring Station Data Logger, S-THB-M002 12-Bit Temperature/Relative Humidity Smart Sensor, S-TMB-M006 12-Bit Temperature Smart Sensor, S-BPB-CM50 Smart Barometric Pressure Sensor, Onset Computer Corporation, Bourne, Massachusetts) on a floating platform in the center of the lake with a precision of ± 0.2\(^\circ\)C and ± 3.0 mbar, respectively.

Statistics

All statistics were conducted with R Version 3.6.0. Data were tested for normal distribution and homogeneity of variance with Shapiro–Wilk and Levene test (Fox and Weisberg 2019; R Core Team 2019). Depending on that suitable tests were applied to test for differences of CH\(_4\) fluxes, sedimentation rates, and OM quality parameters between sites and sampling dates (see Supporting Information S3). Correlations between variables were calculated by nonparametric Spearman’s rank correlation. All tests were performed on a 95% confidence interval and a significance level of \( \alpha = 0.05 \).

To predict CH\(_4\) ebullition from environmental parameters, we used a binomial gamma hurdle model to deal with zero inflation of our data. With this model, we assumed a gamma distribution of the flux data set and calculated the probability of observing a flux > 0. Afterward, all nonzero flux observations were natural log transformed (ln) and modeled by a general linear mixed-effect model (GLMM) with the R package “lme4” (Bates et al. 2015) and general linear models (GLM). The GLMM was used to distinguish between temporal and spatial drivers of CH\(_4\) ebullition. In the GLM, we combined temporal and spatial drivers in one model to explain the observed variability in CH\(_4\) ebullition (for details, see Supporting Information Text S3).

Results

Environmental parameters and sediment properties

Daily average air temperature during the sampling period in 2017 and 2018 was between −9.5 and 26.4\(^\circ\)C. Sediment temperature followed air temperature but with a lower amplitude between 3.5 and 23.2\(^\circ\)C. Air pressure ranged from 929.0 to 983.7 mbar and water level measured at the lake center fluctuated between 100.7 and 149.9 cm (see Supporting Information Fig. S3).

Sediment OM content was between 8.7% and 64.7% with lowest values at site 3.50 and highest values at site 1.50. Water content ranged from 40.0% to 96.7% and porosity from 0.622 to 0.982 with the lowest and highest values found at sites 3.50 and 2.100, respectively (see Supporting Information Table S1). Sediment OM quality was defined by the FTIR peak ratio indicative of fats, waxes, and lipids normalized by the signal for carbohydrates and exhibited lowest ratios (= highest potential degradability/labile material) at site 3.50 and highest ratios (= higher abundance of moieties considered refractory) at site 3.125 as reported in detail elsewhere (Praetzel et al. 2020).

Fig. 2. Temporal variability of CH\(_4\) ebullition in 2017 and 2018. \( N = 7–13 \). Bold lines are the median, boxes show the 25 and 75 percentile, and whiskers indicate minima and maxima within 1.5 times the interquartile range.
We collected in total 283 samples during the study period (135 in 2017 and 148 in 2018). Overall, CH$_4$ ebullition fluxes showed a high-spatial and temporal variability as well as a high range of magnitude. Measured CH$_4$ fluxes in 2017 and 2018 ranged from 0 to 872 mg m$^{-2}$ d$^{-1}$ (maximum observed at site 3.150 in August 2017) with a mean of 70.31 mg m$^{-2}$ d$^{-1}$ for the whole period and no significant differences between the 2 years. Methane ebullition followed a distinct seasonal pattern with highest fluxes in July and August and differed significantly between sampling dates (median test, df = 25, p < 0.001, Fig. 2). Also, on a spatial scale, fluxes significantly differed from each other, especially fluxes at transect 3 were significantly lower than at other sites (median test, df = 12, p < 0.001, Fig. 3a). Overall, CH$_4$ ebullition was highest in the lake center and decreased toward the shore (median test, df = 3, p < 0.001, Fig. 3b). While at sites 3.50 and 3.100, CH$_4$ ebullition was hardly detectable, highest average fluxes of 162.5 mg m$^{-2}$ d$^{-1}$ were measured at site 3.150. CH$_4$ concentration in the trapped gas ranged from 0.01% to 86.4% by volume, average CO$_2$ concentration was 1.01% ± 1.84% with a range of 0.01% to 20.3%.

**Spatiotemporal patterns of CH$_4$ ebullition**

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**Sedimentation rates**

Measured sedimentation rates were between 2.4 g m$^{-2}$ d$^{-1}$ at site 3.100 in November 2018 and 91.1 g m$^{-2}$ d$^{-1}$ at site S.150 in August 2018 with an average rate of 14.6 ± 15.8 g m$^{-2}$ d$^{-1}$ (see Supporting Information Table S2). Sedimentation rates were significantly different between some sampling dates (median test, df = 7, p < 0.01, see Supporting Information Fig. S4a) and sites (Kruskal–Wallis, df = 9, p < 0.05, see Supporting Information Fig. S4b). Sedimentation rates increased from the shore to the center of the lake (see Supporting Information Fig. S4c). Similar results were obtained after normalizing (i.e., division of rates by mean values of each sampling date to remove temporal variability) sedimentation rates (sites: median test, df = 9, p < 0.001; see Supporting Information Fig. S5).

**Quality of sedimented organic matter**

Major elements in material collected with sediment traps were carbon (C), nitrogen (N), phosphorous (P), sulfur (S), manganese (Mn), and iron (Fe) with average concentrations for the whole sampling period of 314 g kg$^{-1}$ C, 28 g kg$^{-1}$ N, 4.6 g kg$^{-1}$ P, 10.5 g kg$^{-1}$ S, 0.8 g kg$^{-1}$ Mn, and 38 g kg$^{-1}$ Fe (for elemental stoichiometric ratios, see Supporting Information Table S1).
Information Table S2). Major functional moieties of OM as identified by FTIR analysis were polysaccharides (1033–1037 cm\(^{-1}\)) and proteins (1544–1558 cm\(^{-1}\)), both indicative of relatively labile material, and lignin (1375–1382 cm\(^{-1}\)) and aromatic (1646–1656 cm\(^{-1}\)) structures, humic acid-like compounds (1403–1421 cm\(^{-1}\)), fats, waxes, and lipids (2848–2852 cm\(^{-1}\)) and cellulose (3334–3394 cm\(^{-1}\)), which indicate refractory material (for average peak ratios see Supporting Information Table S2).

Even though we found significant differences between the years 2017 and 2018 (except for C/N ratio, lignin, and S) as well as between some sampling dates, we could not identify a distinct seasonal pattern in OM quality parameters (see Supporting Information Fig. S6; Table S3).

On a spatial scale, significant differences between some sites were found for C, N, C/N ratio, \(\delta^{13}C\), aromatics, P, and S. We obtained similar results with normalized values (see Supporting Information Fig. S7; Table S3). We did not find significant differences between lake depth categories except for fats, waxes, and lipids and S (Kruskal–Wallis, \(df = 2, p < 0.001\)). Although differences were not significant, we found slight positive trends of OM functional group peak ratios (except cellulose) from the shore toward the lake center. The same was true for C content, C/N ratio, P and S concentrations, whereas Mn and Fe concentrations tended to decrease with increasing lake depth (see Supporting Information Fig. S8).

Controls on CH\(_4\) ebullition

Significant correlations between CH\(_4\) ebullition and potential controls were found for sedimentation rates, C, C/S ratio, \(\delta^{13}C\), aromatics, S, Mn, Fe, lignin, phenolics, humic acid-like components, proteins, fats, waxes, and lipids, cellulose, air, water, and sediment temperature, air pressure, water depth, distance to shore, sediment water and OM content, porosity, and sediment OM quality (for test statistics, see Supporting Information Table S4). Most parameters, however, were correlated inversely to what we expected: CH\(_4\) ebullition was negatively correlated with C content, while FTIR peak ratios indicative of recalcitrant moieties (lignin, phenolics, humic acid-like components and fats, waxes, and lipids) as well as sediment OM quality, water depth, and distance of the sites to the shore were positively correlated with CH\(_4\) ebullition. Potential drivers of CH\(_4\) ebullition and its spatiotemporal variability in our study, as identified from literature, are summarized in Fig. 4.

The relationship between CH\(_4\) ebullition and sediment temperature could be described by the modified Arrhenius equation (Kadlec and Reddy 2001; Aben et al. 2017; see Supporting Information Fig. S9):

\[
\text{CH}_4 \text{ ebullition (mg m}^{-2}\text{d}^{-1}) = E_{20} \times \theta_5^{T - 20}, \tag{6}
\]

where \(E_{20}\) is the CH\(_4\) ebullition flux at 20°C, \(\theta_5\) is the system specific temperature coefficient, and \(T\) is the sediment temperature in °C. Coefficients of the modified Arrhenius equation, activation energy \(E_A\) as well as coefficients of determination \(R^2\) for nonlinear least-squares regression can be found in Table 1. Coefficients varied distinctly between sites.

The probability of observing a nonzero CH\(_4\) flux was 70%. A GLMM of CH\(_4\) ebullition \(> 0\) with temperature as fixed and site as random effect could explain 69% of the observed variability in CH\(_4\) fluxes. Among all tested parameters, temperature, porosity, and OM content together were the best predictors of CH\(_4\) ebullition in the GLM and explained 68% of the observed variability in CH\(_4\) fluxes (see Table 2). Other additional parameters did either not result in an increase of the model’s determination coefficient or were not significant (see Supporting Information Table S5).

We further observed differences between dynamic and steady-state conditions in temperature: in periods of increasing or decreasing temperature (spring and autumn) porosity alone served as good predictor for CH\(_4\) ebullition and including OM content in the model did not significantly increase \(R^2\) while in periods of constant temperature in summer, including OM content in the model could distinctly increase the explained variability of CH\(_4\) ebullition (see Supporting Information Table S6).

Discussion

Magnitude and spatiotemporal variability of CH\(_4\) ebullition

The measured average rates (70.3 mg m\(^{-2}\) d\(^{-1}\)) of CH\(_4\) ebullition at our study site were within the range of ebullitive emissions measured in small and shallow (< 1 ha, < 2 m) beaver ponds in Québec, Canada and similar to ebullition from arctic and subarctic ponds and lakes (DelSontro et al. 2016; Wik et al. 2016b). However, fluxes in this study were between 2 and 20 times higher than CH\(_4\) ebullition from oligotrophic...
and mesotrophic lakes in Southern Ontario (Canada), Wisconsin (United States), and Finland and much lower than from a temperate pond and (sub)tropical reservoirs (see Supporting Information Table S7). Some of the studies only conducted single-point measurements, which might underestimate CH4 ebullition, though. We further assume that lower fluxes can be explained due to a larger lake area, higher mean lake depth or lower annual temperatures whereas higher fluxes can be attributed to higher annual temperatures or higher nutrient concentrations.

We found a high range (0–872 mg m$^{-2}$ d$^{-1}$, daily means: 0.8–322.9) of CH4 ebullition fluxes at our study site, which confirmed the findings of previous studies that CH4 ebullition is both highly variable in space and time. For comparison and to simulate a typical sampling regime, we performed 26 (= number of sampling dates in this study) random sub-samplings of the whole dataset, which revealed that only 31% of the simulated daily averages lay within ± 20% of the measured average CH4 flux of 70.3 mg m$^{-2}$ d$^{-1}$. This result plus the unique large sampling size (n = 283), high density of sites (13 sites on 1.4 ha), high frequency of measurements (biweekly) and long measuring period (April–November) of this study point out the importance of spatiotemporal high-resolution measurements to obtain real values for CH4 ebullition fluxes from lakes (Wik et al. 2016a).

The observed seasonal variability of CH4 ebullition was expected and often observed and is to a large part driven by air and therefore sediment temperature (DelSontro et al. 2016). We could confirm that the major amount (92% and 96% of the total measured ebullitive flux in 2017 and 2018, respectively) of CH4 ebullition was emitted between May and August. Still, we could not capture CH4 fluxes after ice-off as observed from high-latitude lakes, where CH4 emissions following ice melt accounted for up to 48% of the yearly fluxes (Huttunen et al. 2003); the contribution of these events remains thus unclear in our case. Regarding the very low measured fluxes in November 2017 (3.9 mg m$^{-2}$ d$^{-1}$) and in March 2018 (0.8 mg m$^{-2}$ d$^{-1}$), approximately 3 weeks after ice melt, and considering a much shorter period of ice cover in temperate lakes (~ 2 months vs. > 6 months), we assume that fluxes after ice melt are of minor importance in the lake under study as confirmed by very low CO2 concentrations in the surface water (M. Schmiedeskamp unpubl.).

In contrast to observations at larger and/or deeper lakes (Bastviken et al. 2004; Wik et al. 2013; Natchimuthu et al. 2016), we found that CH4 ebullition in Lake Windsborn

### Table 1. Coefficients of the modified Arrhenius-equation and coefficients of determination for temperature-ebbullition relationship at every site. Activation energy (EA) was calculated as the slope of the linear regression of logarithmic (ln) CH4 ebullition and the inverse of temperature and multiplied by the universal gas constant.

| Site  | Total 1.50 | 1.100 | 1.125 | 1.150 | 2.50 | 2.100 |
|-------|------------|-------|-------|-------|------|-------|
| $E_2_0$ | 107.6      | 120.6 | 61.9  | 151.8 | 106.4| 46.5  | 84.6  |
| $\theta_S$ | 1.30       | 1.44  | 1.16  | 1.27  | 1.84 | 1.36  | 1.26  |
| $E_A$ (eV) | 2.31       | 1.73  | 2.05  | 1.78  | 2.46 | 2.04  | 2.72  |
| $R^2$ | 0.25       | 0.34  | 0.17  | 0.42  | 0.94 | 0.29  | 0.43  |

### Table 2. Model parameters, Akaike information criterion (AIC), and $R^2$ for different tested GLMs. Asterisks denote significance levels of probability (**p < 0.01, ***p < 0.001).

| Model CH4 flux ~ | Intercept | Slope 1 | Slope 2 | Slope 3 | AIC | $R^2$ |
|------------------|-----------|---------|---------|---------|-----|------|
| Temperature (T)   | 0.17      | 0.24*** | n.a.   | n.a.   | 2066.3 | 0.53 |
| Porosity (\(\phi\)) | -4.99** | 9.9***  | n.a.   | n.a.   | 2167.2 | 0.19 |
| OM content (OM)   | 2.31***  | 0.04*** | n.a.   | n.a.   | 2170.3 | 0.11 |
| Temperature + porosity | -13.13*** | 0.25*** | 13.45*** | n.a.   | 2034.9 | 0.65 |
| Temperature + OM content | -1.93*** | 0.24*** | 0.04*** | n.a.   | 2055.1 | 0.57 |
| Temperature + porosity + OM content | -13.79*** | 0.25*** | 12.39*** | 0.03 *** | 2026.3 | 0.68 |

$$\text{ln}(\text{CH}_4 - \text{ebullition}) \text{ (mg m}^{-2}\text{d}^{-1}) = -13.79 + 0.25 \ T + 12.39 \ \phi + 0.03 \ \text{OM}$$
increased from the shore toward the center of the lake. These studies from larger lakes suggest that changes in air pressure as well as wave-induced pressure changes might promote bubble release from shallow sediments (Bastviken et al. 2004), and that a deeper water column supports more oxidation during bubble rise through the oxygenated water column and therefore lower emissions (McGinnis et al. 2006). In our study, we found a reverse pattern, which indicates that other lake and/or sediment characteristics might be important drivers of CH$_4$ ebullition in a small and shallow lake.

**Controls on CH$_4$ ebullition**

**Temperature and air pressure**

In our study, we found a strong exponential relationship between temperature and CH$_4$ ebullition as shown in previous studies by, for example, Aben et al. (2017). Both CH$_4$ production in the lake sediment as well as efflux from the water surface are known to depend on temperature, as explained by the higher temperature optimum of methanogenic archaea in the sediments (Duc et al. 2010; DelSontro et al. 2016; Aben et al. 2017). A GLMM with site as random effect showed that temperature could explain 69% of the observed temporal variability in whole lake fluxes when considering each site separately. This indicated that temperature was the main factor that explained temporal variability, whereas the spatial variability in CH$_4$ emissions was caused by other differences between sites. This major effect of temperature was not expected as other factors such as the availability of oxygen, of alternative electron acceptors that suppress methanogenesis, or of substrates are considered as important drivers of the temporal variability of methanogenesis (Bastviken 2009) but were no significant parameters in our model (see Supporting Information Table S5).

The modeled average CH$_4$ flux at 20°C ($E_{20}$) of Lake Windsborn (mean: 107.6, max. 239.74 mg m$^{-2}$ d$^{-1}$) was higher than from temperate mesotrophic mesocosms (84 mg m$^{-2}$ d$^{-1}$) and standing waters in Finland and the UK (0.03 and 58 mg m$^{-2}$ d$^{-1}$) but lower than in eutrophic ponds in the Netherlands, Canada, the United States, and China (267, 141, 372, and 1309 mg m$^{-2}$ d$^{-1}$) (Yvon-Durocher et al. 2014; Aben et al. 2017). As presented in Supporting Information Fig. S3, sediment temperature followed with only little delay changes in air temperature, likely due to the direct influence of solar radiation and/or via rapid heating of the shallow water column. We expected shallow systems to be more vulnerable to increasing temperatures than larger lakes, as their smaller water column could not effectively buffer temperature changes (Jankowski et al. 2006; Bastviken et al. 2008). But comparing our data to other systems, it remains unclear whether shallow mesotrophic systems are in general more vulnerable to increasing temperatures than deeper and/or eutrophied lakes.

In contrast to earlier studies (Wik et al. 2013; Natchimuthu et al. 2016), we did neither find a strong relationship between air pressure and CH$_4$ ebullition, nor was the magnitude of CH$_4$ ebullition influenced by periods of decreasing or increasing air pressure (defined by the slope of 10-min pressure changes within the deployment period). Low air pressure events usually lead to higher CH$_4$ emissions as the overall hydrostatic pressure on the sediment decreases and bubbles are more easily released (Mattson and Likens 1990). We assumed that air pressure would have a more pronounced effect in a shallow lake as changes in air pressure would affect the hydrostatic pressure on the sediment more directly due to the shallowness of the overlying water column, but we could not confirm this with our study. It must be noted, though, that during our sampling campaigns, we only covered a comparatively low pressure-amplitude (see Supporting Information Fig. S3) and probably missed short-termed pressure decreases with our sampling method. We did carry out measurements during low-pressure periods in winter, but CH$_4$ fluxes in winter—except after ice melt—are expected to be low due to low temperatures. Our results show that on a seasonal scale, temperature is the dominant driver of temporal variability of CH$_4$ ebullition in a shallow lake whereas on a daily scale, pressure might become more important.

**Sedimentation rates**

In Lake Windsborn, we found a significant positive relationship between sedimentation rates and CH$_4$ ebullition, both on a temporal and spatial scale. In presence of high sedimentation rates, namely in summer or in the lake center, we also measured highest rates of CH$_4$ ebullition. We suggest that this positive relationship is due to the stimulation of methanogenesis in the lake sediment in response to a higher substrate supply coinciding with findings reported for temperate reservoirs and a river (Sobek et al. 2012; Wilkinson et al. 2015; Berberich et al. 2019). Although sedimentation rates increased the explained variability of observed CH$_4$ fluxes, they were not significant in our GLM. We assume that the number of measurements was not sufficient to achieve significance in the model as sedimentation rates were only measured in 2018 and only at 10 out of the 13 sites.

**Organic matter quality**

Organic matter quality is known to be a major driver of OM mineralization by microbes and therefore the production of the metabolic end products CO$_2$ and CH$_4$ (Updegraff et al. 1995; Fenchel et al. 2012). Easily degradable OM compounds like polysaccharides and proteins are favored by microorganisms over more recalcitrant compounds like aromatics, waxes or lignin-like structures; the latter therefore accumulate in the sediment, as reflected in higher absorption intensities of these moieties in sediment matter FTIR spectra (Fenchel et al. 2012; Tfaily et al. 2014). Wik et al. (2018) did not find a relationship between CH$_4$ ebullition and total organic C content in three subarctic lakes but suggested rather OM quality to be of major importance. However, we found an inverse relationship between the assumed lability of the
sedimented organic material and CH$_4$ ebullition. Along with highest rates of ebullition, highly decomposed organic material enriched in refractory moieties was found in the lake center—both in the sediment traps and in the sediment itself. We assume that the material collected there was exposed to the oxic water column for extended time periods during sedimentation and could therefore be degraded more intensively compared to sedimented OM closer to the shore where the water column was shallower (Meyers 1994). In addition, OM in the lake center could have undergone intensified decomposition during resuspension and focusing of sediment particles (Mackay et al. 2012). Although CH$_4$ production in incubations has been shown to strongly correlate with sediment OM quality in Lake Windsborn (Praetzel et al. 2020), other sediment characteristics might also be important for bubble formation, growth and subsequent ebullitive release from the sediment in situ or in intact sediments. This might be responsible for the observed discrepancy between CH$_4$ production in the sediment and ebullition.

**Physical sediment properties and organic matter quantity**

The peat matrix and physical peat structure were found to play a crucial role in the accumulation and release of CH$_4$ bubbles (Wright et al. 2018). While some studies found positive relationships between gas storage and release (Liu et al. 2020), others did not see any correlation between CH$_4$ content in the sediment and ebullition rates in lakes (Martinez and Anderson 2013). Contradictory findings were also obtained concerning gas storage and sediment properties; while high gas storage was associated with high porosity sediments and peat in some studies (Martinez and Anderson 2013; Wright et al. 2018), others found low-porosity, sandy sediment to store and retain more gas (Ramirez et al. 2015). In some cases, ebullition was found to correlate positively with sediment sand content (Amos and Mayer 2006; Martinez and Anderson 2013). In Lake Windsborn, spatial variability of CH$_4$ ebullition could be indeed modeled by a combination of sediment porosity and OM content, and such a model had a higher explanatory power than including OM content or porosity only. We suggest that both physical sediment structure and the availability of decomposable OM contribute to formation, accumulation, and release of CH$_4$ bubbles. While the quantity and quality of OM can indirectly influence CH$_4$ ebullition as it determines CH$_4$ production rates (Conrad et al. 2011; Berberich et al. 2019), pore size and distribution more directly affect bubble formation and size (Boudreau et al. 2005). Porosity sufficiently explained the dynamics of CH$_4$ bubble formation and release in a northern peatland where OM contents and porosity were similar to those in the sediments under study and might therefore act analogously (Chen and Slater 2015).

A study by Liu et al. (2016) revealed that the formation process of CH$_4$ bubbles was dependent upon the sediment type. While CH$_4$ bubbles in sand were formed by capillary invasion, bubbles in fine sediments could form by sediment displacement. The formed bubbles were larger in highly porous sediments. In both sediments, the resulting enhanced connectivity of macropores served as conduit for bubble release (Boudreau 2012; Liu et al. 2018).

If gas production is below the storage capacity of the sediment, ebullition will not occur until the gas content in the sediment exceeds the storage capacity (Liu et al. 2016). We compared the interaction of porosity and OM content under dynamic and steady-state conditions (i.e., periods of increasing/decreasing temperature vs. constant temperature) and could confirm that under dynamic conditions, porosity alone could serve as predictor for CH$_4$ ebullition while under steady-state conditions, OM content besides porosity was an important explanatory variable that significantly increased the explained variability of observed CH$_4$ fluxes (see Supporting Information Table S6).

We found higher ebullition rates in high porosity sediments. We believe this is because (1) in fine sediments, gas could be stored deeper in the sediment column and is therefore able to extend the period of gas bubbling (Liu et al. 2016) and (2) conduits for bubble release can more easily be formed in soft sediments (Boudreau 2012). Furthermore, Liu et al. (2016) found larger bubbles in fine sediments, and we do not believe that bubble dissolution during ascent through the water column plays a major role in shallow waters. McGinnis et al. (2006) confirmed that in shallow waters < 20 m, CH$_4$ loss from bubbles is negligible.

Liu et al. (2016) pointed out that the interaction of porosity, gas storage, and CH$_4$ ebullition is only valid in systems with high and episodic sedimentation rates. Whether it can be applied to small and shallow systems with gradual and comparatively (in contrast to reservoirs and rivers) low sedimentation rates, remains unclear.

We observed that ebullition and production rates in Lake Windsborn were not correlated with each other (see Supporting Information Fig. S10) and that a higher share of recalcitrant OM compounds could not explain lower ebullition rates. We therefore concluded that factors other than OM quantity and quality must be important for CH$_4$ bubble formation. Our models proved that porosity and OM quantity together could best explain the observed variability in CH$_4$ fluxes. Although positive correlations between porosity and OM content have been reported earlier for marine sediments (Bergamaschi et al. 1997), we found a positive, but not significant relationship between the two parameters. Another explanation for the observed discrepancy between CH$_4$ production and ebullition might be the lack of the influence of sedimentation rates in incubations, fuelling production. Although not significant in the model, sedimentation rates were related to CH$_4$ ebullition. Including OM content in the model hence considers the influence of (sedimented) quantity of fresh OM on production and therefore ebullition rates from the lake sediments under study.
Overall, studies on drivers of spatial $\text{CH}_4$ ebullition in lakes remain scarce, and results are partly contrary or remain vague. More research on gas bubble evolution and storage in sediments, especially under in situ conditions, is needed to further elucidate the role and interaction of physical and biogeochemical drivers of $\text{CH}_4$ ebullition in small and shallow lakes. Although OM content and quality was repeatedly shown to be major controls on $\text{CH}_4$ ebullition, our results support that considering sediment physical properties potentially improves our understanding of $\text{CH}_4$ ebullition patterns in small and shallow lakes under near-natural or in situ conditions.

**Conclusion**

Our study showed that a small and shallow temperate lake can act as significant source of $\text{CH}_4$ to the atmosphere. We could confirm our first hypothesis, namely that there exists a distinct spatiotemporal pattern of $\text{CH}_4$ ebullition also within a small lake. However, we found opposite spatial patterns than observed from most large and deep lakes: Lake Windsborn exhibited increasing $\text{CH}_4$ ebullition fluxes from the shore to the center of the lake. Moreover, temporal $\text{CH}_4$ ebullition was strongly controlled by sediment temperature, but the temperature-flux relationship was distinct between the sites. Spatial heterogeneity of $\text{CH}_4$ ebullition could be explained by sediment porosity and OM quantity of the sediment, whereas the quality of sedimented OM did not play a significant role. For future studies on spatial variability of $\text{CH}_4$ ebullition, besides focusing on biogeochemical conditions, we suggest including physical sediment characteristics, bubble evolution processes, and gas storage patterns in the sediment as important drivers.

**Data Availability Statement**

Raw data related to this article is available from PANGAEA database (Praetzel et al. 2020, https://doi.pangaea.de/10.1594/PANGAEA.917518) published under license CC-BY-4.0.

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Conflict of Interest

None declared.

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