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Sediment Characteristics and Methane Ebullition in Three Subarctic Lakes

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- Climate sensitive postglacial lake sediments can be a sustained methane source with Arctic warming

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Sediment Characteristics and Methane Ebullition in Three Subarctic Lakes

JGR

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Abstract Ebullition (bubbling) from climate-sensitive northern lakes remains an unconstrained source of atmospheric methane (CH₄). Although the focus of many recent studies, ebullition is rarely linked to the physical characteristics of lakes. In this study we analyze the sediments of subarctic postglacial lakes and investigate how sediment properties relate to the large spatial variation in CH₄ bubble flux, quantified over multiple years using bubble traps. The results show that the sediments from our lakes are rich in total organic carbon, containing 37 kg/m³ on average. This number is roughly 40% higher than the average for yedoma deposits, which have been identified as high CH₄ emitters. However, the quantity of total organic carbon is not a useful indicator of high emissions from the study lakes. Neither is the amount of CH₄ in the sediment a reliable measure of ebullition potential. Instead, our data point to coarse detritus, partly from buried submerged aquatic vegetation and redeposited peat as spatial controls on fluxes, often in combination with previously established effects of incoming solar radiation and water depth. The results once again highlight the climate sensitivity of northern lakes, indicating that biological responses to warmer waters and increased energy input and heating of organic sediments during longer ice-free seasons can substantially alter future CH₄ emissions.

1. Introduction

Lakes are important sources of atmospheric methane (CH₄) on a regional to global scale (Bastviken et al., 2004; Cole et al., 2007), and their contribution is predicted to increase in response to climate change and rapidly warming waters (O'Reilly et al., 2015; Tan & Zhuang, 2015). Lakes are common at high northern latitudes (Verpoorter et al., 2014), and their sediments store substantial amounts of carbon (C), often as a result of deposition and burial over millennia (Sobek et al., 2014; Tranvik et al., 2009). In the Arctic and subarctic, most water bodies are formed by peat degradation or thermokarst processes (Smith et al., 2007); however, postglacially formed lakes are estimated to dominate the overall distribution in terms of area (Wik, Varner, et al., 2016). Strangely enough, while northern C-rich ecosystems such as terrestrial permafrost soils have become synonymous with CH₄ emissions (Olefeldt, Turetsky, et al., 2012; Schuur et al., 2015), lakes and particularly those of postglacial origin have received little attention and only recently earned a unique place in global CH₄ budgets (e.g., Saunois et al., 2016). Perhaps one reason for the discrepancy is that CH₄ emissions from lakes are poorly understood due to complex emission pathways and a lack of robust measurements (Bastviken et al., 2004; Natchimuthu et al., 2016; Wik, Thornton, et al., 2016). Most of the CH₄ produced in lake sediments enters the atmosphere via ebullition (Bastviken et al., 2004; Walter et al., 2007), a temperature-sensitive transport mode with high heterogeneity in both space and time (DelSontro et al., 2015; Walter Anthony & Anthony, 2013). Few lake studies relate their measured fluxes to the physical characteristics of their study sites, particularly the sediments, thereby making it difficult to determine controls on the large spatial variations in measured fluxes, on both local and regional scales (Wik, Varner, et al., 2016).

Another reason why lakes are often ignored in terms of CH_4 emissions could be related to the general understanding that lake sediments are mostly allochthonous, consisting of substrates that are poor in guality or recalcitrant to anaerobic decomposition (e.g., Tranvik et al., 2009). Erosional margins and underlying



thaw, however, can supply labile organic C to countless water bodies in permafrost regions (Jones et al., 2011; Kessler et al., 2012). Labile C in shallow lakes can also be autochthonous, produced from seasonal primary productivity (phytoplankton) that contributes to sedimentation and methanogenesis (e.g., Sinke et al., 1992). In addition, strong links have been identified between CH_4 production and diagenesis of in situ submerged aquatic plants (macrophytes; Cronin et al., 2006; Marinho et al., 2015), suggesting that macroscopic benthic flora can play a role in accumulating autochthonous C, suitable for methanogenesis. Longer icefree seasons over the next decades (Dibike et al., 2011) will allow for more solar radiation into lakes, and earlier ice-out and longer heating of the sediments have been shown to positively affect CH_4 fluxes (Thornton et al., 2015a; Wik et al., 2014). Warmer lake waters are also a likely contributor to increased growth and altered communities of macrophytes (Rooney & Kalff, 2000; Zhang et al., 2015), possibly supplying more labile organic C to the CH_4 -producing sediment microbial populations.

In this paper we investigate potential links between sediment composition and CH₄ ebullition of three small postglacial lakes located in a subarctic landscape in northern Sweden. We focus on sediment properties such as total organic carbon (TOC), total nitrogen (TN), total sulfur (TS), bulk density (BD), and CH₄. Depth-stratified measurements of ice-free season ebullition and temperature have been ongoing for almost a decade in these lakes (Wik et al., 2013), allowing our sediment results to be compared with a unique data set of fluxes, and provide information that is also useful for constraining CH₄ emission from northern lakes at larger, regional scales.

2. Study Area

The study lakes (Inre Harrsjön, Mellersta Harrsjön, and Villasjön) are of postglacial origin and surround a subarctic peatland underlain by sporadic/discontinuous permafrost, the Stordalen Mire, 11 km east of Abisko in northern Sweden (Figure 1). The Stordalen area is experiencing steadily increasing temperatures (Callaghan et al., 2010). Consequently, the mire is undergoing rapid thaw, visible through collapsing palsas and vegetation shifts as the landscape becomes wetter and more fen-dominated (Johansson et al., 2006; McCalley et al., 2014). Stordalen lies adjacent to the large (330 km²) glacial lake Tornesträsk. The water level of paleo-lake Torneträsk has been ~250 m higher than its current elevation (340 m above sea level; Melander, 1977; Vogel et al., 2013) prior to the end of the deglaciation here at ~9,500 years B.P. (Berglund et al., 1996). The small postglacial lakes around the Stordalen mire exist at an elevation of 351 m above sea level, well within the extent of paleo-lake Torneträsk or younger, early Holocene lakes that formed prior to the terrestrialization of the northern Stordalen mire region, ~4,700 years B.P. (Kokfelt et al., 2010).

Inre Harrsjön and Mellersta Harrsjön are small in size (0.02 and 0.01 km²) with maximum depths of 5.5 and 6.5 m, respectively. Villasjön is larger and covers 0.17 km² but is shallower with a maximum depth of 1 m. The three lakes formed around 3,400–2,650 years B.P. (with Villasjön being the oldest) alongside the evolution of the adjacent mire (Kokfelt et al., 2010). The lakes have different dominant macrophytes in their shallow waters (0–2 m); *Sparganium angustifolium* dominates in Mellersta Harrsjön (Stilson et al., 2014) whereas *Myriophyllum alterniflorum* and *Potamogeton alpinus* are most common in Inre Harrsjön (Wik et al., 2011). None of these species are equally abundant in Villasjön, which has a lower macrophyte diversity and biomass. Inre and Mellersta Harrsjön are usually stratified with a thermocline around 3-m depth during summer, before full water column mixing occurs in the fall. Villasjön, on the other hand, is well mixed throughout the summer season due to its shallow nature and strong winds in the Stordalen valley. Further descriptions of the lakes can be found in Jammet et al. (2015) and Wik et al. (2013).

In terms of CH₄ emission, the total ice-free season flux from the Stordalen lakes of around 20 mg · m⁻² · day⁻¹ (two-third ebullition and one-third hydrodynamic flux [commonly and incorrectly called "diffusive flux"] Wik et al., 2013; Wik, Varner, et al., 2016) is similar in magnitude to those measured at many other postglacial and thermokarst lakes (e.g., Sepulveda-Jauregui et al., 2015). Villasjön is consistently most productive in terms of ebullition (average of ~23 mg CH₄ · m⁻² · day⁻¹) and Inre Harrsjön the least (~7 mg CH₄ · m⁻² · day⁻¹). The fluxes are generally higher from shallow waters compared to deep zones and increase with rising lake temperatures over the season (Wik et al., 2013, 2014).







3. Methods

To characterize the lake sediments in this study, a total of 20 sediment cores were taken in July of 2013 from a small row boat using a modified Multi Stage Soil Core Sampler (AMS, Inc). The AMS corer consists of extendable stainless steel cylinders and replaceable 1-m-long plastic liners (5-cm diameter) that were manually pushed into the sediments using a sliding weight stand above a variable number of extension rods, depending on the lake depth. Inre and Mellersta Harrsjön were cored at two locations each at different water depths, one shallow (1–1.5 m) where ebullition is frequent and one deep (5.5 and 6.5 m) where there is less bubbling. In the overall shallow Villasjön, cores were taken at six locations along a transect of bubble traps that stretched across the south-western end of the lake (Figure 1). The higher number of cores here compared to those taken in the other two lakes were aimed to investigate the controls on a steep ebullition gradient in this consistently shallow zone (Figure 3a).

Replicate cores were collected from each location. The liners were capped before being pulled out of the water to minimize loss of sediment and sediment CH_4 prior to sampling. All cores were photographed, and layering, colors, and textures were described and noted in the field. We visually distinguished

between four different types of sediment: peat, coarse organic detritus gyttja, fine organic detritus gyttja, and lithogenic deposits.

We sampled the first core at each location for TOC, TN, and TS. The procedure involved splitting the core in half, and samples of 1 cm³ were taken in 5-cm increments from top to bottom. The samples were dried, ground, and split into an untreated sample for total carbon (TC), TN, and TS, and an acidified TOC sample. Prior to the TOC analysis, inorganic carbon (IC = TC-TOC) was dissolved using 6% sulfurous acid applied to weighed samples (e.g., Verardo et al., 1990) in amounts and steps optimized for a range of carbonate contents (Phillips et al., 2011). Bulk and acidified samples were measured using a Perkin Elmer 2400 Series II CHNS/O Elemental Analyzer at the University of New Hampshire (UNH). Ratios of TOC:TN were calculated using the weight percent (wt %) TOC and TN and their respective atomic weights. Weight % of calcium carbonate (CaCO₃) was calculated by multiplying the IC wt % by 8.33 to account for the noncarbon mass fraction. The calculated bulk CaCO₃ fraction represents biogenic, authigenic, and any detrital carbonate phases; however, in this study CaCO₃ was negligible (TC \approx TOC). Repeatability error was established by analyzing replicate samples and calculating the standard deviation (SD). Duplicate samples were run approximately every 10 samples. Potential outliers were also run in duplicate.

The second, replicate core was used for quantifying total CH_4 in μ g/cm³ wet sediment and BD in g dry weight (dw) per cm³. In the field, we pulled 2-cm³ sediment plugs into cut plastic syringes through predrilled and taped holes made at 5-cm increments along the core liner. The plugs were quickly transferred to 30-ml vials containing 5 ml of 2 M NaOH, which were instantly capped and shaken (Magen et al., 2014; Pimmel & Claypool, 2001). The sample vials were stored overnight, then heated at 60 °C for 1 hr. After they cooled to room temperature, the headspaces were analyzed for CH₄ using a Shimadzu GC-2014 gas chromatograph with a flame ionizing detector (e.g., Wik et al., 2013). The CH₄ measured represents the total; that is, both the CH₄ dissolved in the water from the sediment plug and any bubbles that may have been trapped in the sediment. The remaining sediment samples in the vials were weighed and dried to constant weight to determine the dry sediment mass. Because of relatively few measurements per core (n = 8-15) and large within core variations (see below sections) we have not made statistical comparisons of sediment properties among coring locations as such tests would not be representative.

Methods for measuring ebullition and water temperature are explained in detail by Wik et al. (2013). In brief, measurements of ice-free season CH_4 bubble flux have been ongoing at the Stordalen lakes since 2009. A total of 40 bubble traps, distributed in a depth-stratified sampling scheme (Figure 1), were sampled frequently (often daily) from June to September. For this study, spatial averages in CH_4 bubble flux were calculated by binning data from traps close to the coring locations (total of 4,360 measurements). The traps chosen are shown in Figure 3a. The different means were compared statistically. In spite of skewed distributions, the large number of samples in each bin (n = 228-549) enabled the use of parametric tests, in this case analysis of variance (ANOVA) and Tukey post hoc comparisons at a confidence level of 95%. Temperature profiles throughout the water column and into the surface sediments were measured using thermistor strings (Onset HOBO U22 loggers) deployed at the deepest point of each lake, which in Villasjön was near the middle of the transect.

4. Sediment Characteristics and the Role of Submerged Vegetation

The shallow zone cores show predominantly coarse organic detritus gyttja overlying lithogenic sand and silt. The thickness of the shallow zone organic layer is relatively uniform across the lakes, ranging between 26 and 41 cm (Table 1). The western end of Villasjön (locations V1–V4) has a 15- to 20-cm-thick layer of reworked peat between the gyttja and the lithogenic sediments. This peat layer has been described previously as terrestrial vegetation, consisting of *Sphagnum fuscum*, *Ericaceae* spp., and *Carex* spp., redeposited during erosion events that occurred after 2,100 years B.P. (Kokfelt et al., 2010). In the deep zones of Inre and Mellersta Harrsjön (locations I2 and M2), the organic layer is much thicker (>65 cm; Table 1) and consists of fine organic detritus gyttja, which reflects longer transport times and longer exposures to decomposition (Sobek et al., 2009; Torres et al., 2010). Here our coring equipment was unable to penetrate deep enough to reach the underlying lithogenic layer.

The Stordalen lake sediments are TOC-rich, ranging up to almost 50 wt % in the upper 40 cm (Figure 2a), with a mean and SD of 16.1 \pm 13.1 wt % among all coring locations. Similarly high values of TOC have been found



Sampling Location and Sediment Core Characteristics							
Coring location	Date (d/m/y)	Water depth (m)	Sediment temp. (°C) ^a	Core length (cm)	Organic layer thickness (cm)	Organic sediment type	
Inre Harrsjön							
11	11 July 13	1.5	12.0	62	41	Coarse detritus gyttja	
12	12 July 13	5.5	11.6	66	>66	Fine detritus gyttja	
Mellersta Harrsjön							
M1	12 July 13	0.8	11.1	44	36	Coarse detritus gyttja	
M2	13 July 13	6.5	10.2	72	>72	Fine detritus gyttja	
Villasjön							
V1	2 July 13	0.7	15.3	44	37	Peat/coarse detritus gyttja	
V2	2 July 13	0.8	15.3	39	36	Peat/coarse detritus gyttja	
V3	2 July 13	0.8	15.3	41	41	Peat/coarse detritus gyttja	
V4	2 July 13	1.0	15.3	40	30	Peat/coarse detritus gyttja	
V5	3 July 13	1.0	15.8	41	31	Coarse detritus gyttja	
V6	3 July 13	0.8	15.8	40	26	Coarse detritus gyttja	

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Sampling	Location	and	Sediment	Core	Characteristics

Table 1

^aMeasured in the surface sediments at roughly 10- to 15-cm depth. In Villasjön, the thermistors are located near the middle of the transect, but because of well mixed waters we assume heterogeneous temperatures across this lake.

in other northern lakes of both postglacial and thermokarst origin (Dean, 2006; Fortino et al., 2016; Sepulveda-Jauregui et al., 2015). Because of increasing BDs with depth in the cores (Figure 2b), the amount of TOC (hereafter "bulk TOC" calculated as TOC wt % times the BD) is relatively uniform vertically (Figure 2c) with an overall average of 0.037 g dw/cm³ (37 kg dw/m³) in the sediments of our study lakes. This number is almost 40% greater than the average for yedoma deposits (roughly 27 kg dw/m³; Strauss et al., 2013), which are Pleistocene-aged frozen loess soils across Beringia (the landmass across northeastern Eurasia, Alaska, and northwestern Canada) that are predicted to be major C sources with Arctic warming (Schuur et al., 2015; Vonk et al., 2012). Lakes underlain by yedoma have been found to emit substantial and increasing amounts of CH_4 that are likely fueled by permafrost thaw and continued instability over centuries due to climate change (e.g., Walter et al., 2007). Carbon storage vulnerability could also be the case for postglacial lakes at high northern latitudes. For example, hypothetically, the average bulk TOC in our lake sediments (37 kg dw/m³) could potentially be enough to sustain a doubling of the current total CH_4 emission rates of 20 mg m⁻² d⁻¹ (15 mg C m⁻² d⁻¹) for about 6,300 years, assuming most of the CH₄ is produced from June to September and half of the total bulk TOC is mineralized to carbon dioxide (Corbett et al., 2012).

The allochthonous organic matter that enters the Stordalen lakes may not contribute much to sedimentation. Mellersta Harrsjön, particularly its southern part, has the potential to receive most of the terrestrial loading due to the inflow from the catchment's main stream (Figure 1). The stream transports 0.09 g \cdot m⁻² \cdot year⁻¹ of particulate organic carbon and roughly 9.7 g \cdot m⁻² \cdot year⁻¹ of dissolved organic carbon (DOC; Olefeldt, Roulet, et al., 2012). The lakes are partly spring fed (Nilsson, 2006), and they are relatively clear (secchi disk measurements in summer show ~3-m visibility). The concentration of DOC at the outlet of Mellersta Harrsjön is around 10 mg/L (Olefeldt, Roulet, et al., 2012), which is similar to that of other lakes in the area (Lundin et al., 2013). However, DOC is unlikely to settle in Mellersta Harrsjön; the short residence time of the water (days to weeks) and its low conductivity (20–60 μ S/cm) and neutral pH (6.5–7.5; Nilsson, 2006) are not ideal for aggregates to form (Tranvik & von Wachenfeldt, 2009).

Layers of intact leaves and roots from in situ macrophytes are present at various depths in the shallow zone cores. During warm summers, submerged vegetation grows densely in many parts of the Stordalen lakes. Respiration of the litter can generate bottom anoxia, allowing for substantial burial of left over plant debris (Meding & Jackson, 2003). The sediments' TOC:TN ratios are similar among the coring locations (Figure 2f and Table 2), and with a mean and SD of 17.9 ± 11.1 (n = 249), they are in the range of those found in the macrophytes of the same lakes (14.7 \pm 2.2; Horruitiner et al., 2015). However, the lakes' TOC:TN ratios are much lower than those found in the soils of the nearby fen and palsa (ranging from 32 ± 1 to 68 ± 6 ; Normand et al., 2017, see Figure 2f). Although TOC:TN can be modified by diagenesis in many environments, relative changes in the ratio through time can be a first-order indicator for the relative inputs of organic C in lakes and marine



Figure 2. Sediment profiles of (a) TOC wt %, (b) BD, (c) bulk TOC, (d) total CH_4 , (e) TS wt %, and (f) TOC:TN for cores taken from the shallow and deep zones of Inre Harrsjön (I1 and I2) and Mellersta Harrsjön (M1 and M2) and in the high and low flux zone of Villasjön (V1 and V6). The lines are color-coded by coring location. The blue and brown shaded areas in Figure 2f are for comparison and show ranges of previously measured TOC:TN in the lakes' aquatic macrophytes (Horruitiner et al., 2015) and in soils of the nearby fen and palsa sites (Normand et al., 2017).

systems (Meyers, 1994). In this case, the TOC:TN data suggest that autochthonous macrophyte material may supply much of the sedimentation and anaerobic mineralization in our study lakes.

The TS content in the Stordalen lake sediments (≤ 0.55 wt %; Figure 2e and Table 2) falls within a typical range observed in plants, freshwater peat, and freshwater lake sediments (Berner & Raiswell, 1983; Casagrande et al., 1980; Cohen et al., 1984). Measurable TS in the bulk lake sediments originates from bound sulfur (S) in organic matter and/or detrital or diagenetic iron sulfide (FeS) mineral phases. Exposure of sediment samples to oxygen (O₂) during sampling may allow some S species to oxidize (e.g., sulfide minerals); however, the TS content will still reflect the presence of inorganic S in excess of that present in organic matter. TOC:TS is lower at Inre Harrsjön and Mellersta Harrsjön (76–100) compared to Villasjön (159–195; Figure S1) suggesting a possible inorganic S phase at Inre Harrsjön and Mellersta Harrsjön. At these two lakes, the decrease in TOC: TS may be influenced by stream input and replenishment of sulfate (SO₄²⁻)-bearing waters derived from weathering of S-bearing rocks elsewhere in the watershed. The source of SO₄²⁻ may also drive anaerobic



Mean and SD of Measured Sediment Properties								
Coring location	n ^a	TOC (wt %)	TS (wt %)	BD (g dw/cm ³)	TOC:TN	$CH_4 (\mu g/cm^3)$		
Inre Harrsjön								
11	12	5.7 ± 4.8	0.2 ± 0.2	0.31 ± 0.20	14.6 ± 5.3	13.1 ± 5.1		
12	15	12.4 ± 5.7	0.3 ± 0.1	0.15 ± 0.06	16.0 ± 2.1	36.6 ± 18.4		
Mellersta Harrsjön								
M1	9	6.7 ± 3.8	0.2 ± 0.1	0.71 ± 0.40	13.9 ± 3.4	19.2 ± 4.2		
M2	14	4.1 ± 2.3	0.1 ± 0.1	0.59 ± 0.21	18.5 ± 7.8	22.9 ± 11.0		
Villasjön								
V1	9	21.6 ± 9.5	0.3 ± 0.2	0.19 ± 0.13	16.5 ± 2.3	7.4 ± 4.6		
V2	8	29.3 ± 10.1	0.4 ± 0.2	0.18 ± 0.12	17.9 ± 5.2	18.8 ± 12.8		
V3	9	27.0 ± 9.9	0.4 ± 0.2	0.19 ± 0.14	16.5 ± 3.1	13.6 ± 6.1		
V4	8	23.9 ± 11.3	0.4 ± 0.3	0.38 ± 0.41	32.4 ± 22.5	12.2 ± 5.7		
V5	9	24.8 ± 15.6	0.4 ± 0.3	0.45 ± 0.43	16.8 ± 3.8	16.3 ± 10.8		
V6	8	21.0 ± 16.9	0.3 ± 0.3	0.58 ± 0.50	22.1 ± 10.7	9.8 ± 3.4		

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Table 2

^aNumber of downcore measurements.

oxidation of CH₄ (AOM) in the sediments, potentially converting some of the CH₄ to dissolved IC (Knittel & Boetius, 2009; Smemo & Yavitt, 2011). The hydrogen sulfide produced by AOM can be retained as iron sulfide precipitates in these sediments, and contribute to measured TS, if sufficient pore water Fe or Fe-(oxyhydr) oxide minerals are present. Dissolution of magnetite and precipitation of greigite (Fe₃S₄) has been observed in other lakes of the Abisko region (Snowball, 1991, 1993), suggesting that SO₄²⁻ dependent AOM may be significant in our study area as well. Although no clear relationship exists in our data between TOC:TS and CH₄ ebullition or sediment CH₄ (discussed in sections 5 and 6), SO₄²⁻ dependent AOM may be an important sink for CH₄ in subarctic lakes. In addition, organoclastic SO₄²⁻ reduction may reduce the amount of labile organic matter available for methanogenesis.

5. Sediment CH₄ and Bubble Formation

The amount of CH₄ in our sediment cores range from 0.1 to 68.8 μ g/cm³ and increases downcore (Figure 2d). The slopes of the profiles at the surface (<10–20 cm) indicate production rates that increase with depth and CH₄ loss to the overlying water. Here in the uppermost layer, the loss is likely hydrodynamic (i.e., driven by advection and turbulent transfer) instead of ebullitive. Gas pockets that release bubbles need overlying dense sediment that allows bubbles to form but not be lost. Bubble formation is not likely close to the sediment surface because gas pockets cannot form in unsaturated sediments (Boudreau et al., 2001; Martens & Val Klump, 1980). In most of our cores, bubbles or gas pockets were observed visually below 10–20 cm. Further, aerobic CH₄ oxidation or potential AOM within the uppermost lake sediments could also decrease CH₄ concentrations near the sediment water interface; however, the O₂ dynamics and role of AOM in our lakes remain to be investigated. In addition, degassing is inevitable when cores are pulled from the sediment and adds uncertainty to the CH₄ profiles.

Sediment CH_4 is consistently low on average (7.4–19.2 µg/cm³) in the shallow zone cores, particularly at location V1, where ebullition is generally most frequent (Figure 3 and Table 2) and ongoing around the time of sampling (Figure S2). Aside from potential sampling effects, the low sediment CH_4 could imply rapid production and bubble turnover rates. Because cores are point measurements in time with narrow footprints they are likely to miss large short-lived gas accumulations. Once CH_4 saturation occurs in the sediment, which is essential for bubble formation, all excess biological production likely adds to the partial pressure of the gas phase. In thin organic layers under shallow waters, the pressure of the gas pockets can quickly exceed that of the overlying burden causing frequent ebullition (Fendinger et al., 1992). This process may explain the low amounts of sediment CH_4 in the shallow zone cores in our study.

On average, the highest sediment CH₄ (22.9–36.6 μ g/cm³) is found in cores from the deep zones (locations I2 and M2) where ebullition is rare during most of the season (Figure 3 and Table 2). The production at depth is likely slower than in the shallow zones due to overall lower temperatures, possibly in combination with lower quality sediments (see section 6). Strong relationships have been found between ebullition and temperature





Figure 3. (a) Mean CH₄ bubble flux captured by traps close to this study's coring locations. The fluxes are based on six years of data (two years are added to the four-year data set presented by Wik et al., 2013). The number of flux samples in each bin range from 228–549. The two lower panels show (b) mean bulk TOC and (c) mean sediment CH₄ at each coring location with bars that are color-coded by dominant sediment type. The error bars show 95% confidence intervals (CI) in Figure 3a and SD in Figures 3b and 3c. The reason for using CI in the former is because of highly skewed ebullition distributions for which SD would be a misleading unit about the precision of the mean.

in our study lakes, caused by thermal stratification patterns and limited or delayed solar energy transfer down the water column (Thornton et al., 2015b; Wik et al., 2014). In theory, a slower production rate affects the bubbles' growth rate, allowing them to remain in the sediments for longer. The overlying hydrostatic pressure is also greater at depth, enabling the sediments to accumulate larger gas volumes before ebullition is triggered (Scandella et al., 2011). Both these physical aspects could possibly explain the high sediment CH_4 and low bubbling rates at the deep zone sites. Although some of CH_4 produced at depth is released hydrodynamically to the hypolimnion and via ebullition to the atmosphere, it is possible that much is not released at all during the summer months, but instead stored over winter until spring overturn.



6. Sediment C and Ebullition Variation

In our study lakes, neither the sediments' bulk TOC nor their organic layer thickness appear to affect ebullition potential. Apart from in Mellersta Harrsjön, CH_4 bubble fluxes are lower from sediments with seemingly higher average bulk TOC (Figures 3a and 3b), suggesting that TOC quantity contributes little to the bubbles' spatial heterogeneity. The spatial variations in bulk TOC cannot be verified statistically in this study due to few samples per core (n = 8-15), but large within-core variations (Figure 3b) and similar profiles at most coring sites (Figure 2c) suggest relatively homogeneous bulk TOC across the lakes. In contrast, the spatial variation in ebullition appears to be more influenced by differences in the quality of the TOC, partly coupled to water depth, again also in combination with the effects of temperature and energy transfer (e.g., Wik et al., 2014).

Across the overall shallow Villasjön, CH₄ bubble fluxes are threefold higher on average (42.3 mg CH₄ · m⁻² · day⁻¹) from areas underlain by mixed, redeposited peat (locations V1–V4) compared to those underlain by more TOC-rich coarse detritus gyttja at locations V5–V6 (14.0 mg CH₄ · m⁻² day⁻¹; P < 0.001, n = 2209; Figure 3). Including the shallow zone data from all three lakes, the fluxes are twofold higher from peat than from coarse detritus gyttja (22.7 mg CH₄ · m⁻² · day⁻¹; P < 0.001, n = 3273). The peat is less decomposed than the gyttja and thus likely of higher quality and possibly a more suitable substrate for the methanogens. Studies have shown that peats of mixed vegetation are more effective in producing CH₄ than those only containing *Sphagnum spp*. (e.g., Hines et al., 2008). Many studies also report high CH₄ emissions from small ponds underlain by peat, formed by either permafrost thaw or peat degradation (e.g., Hamilton et al., 1994; Pelletier et al., 2007).

It is important to note, however, that Villasjön is generally shallower than the shallow zones of Inre and Mellersta Harrsjön and remains well mixed throughout the season. Thus, it cannot be assumed that the underlying peat is solely responsible for Villasjön's higher bubbling rates. Faster emission responses in Villasjön due to overall warmer water and more rapid sediment heating, observed by Thornton et al. (2015b), could possibly be the main driver. It is also notable that locations V3–V4 where peat is present show statistically similar CH₄ bubble fluxes as location M1 in Mellersta Harrsjön where peat is absent (P = 0.131, n = 985; Figure 3b). Further, the sediments in the southeastern part of Villasjön, although being more TOC-rich (Figure 3b), are mixed with numerous stones and small boulders. Such a sediment not only limits the area from where bubbles can be released but also decreases the likelihood for bubbles to be captured by the traps, which might explain the apparent negative gradient in ebullition moving away from the peat sediments at the west side of the lake (Figure 3a). The role of peat in fueling ebullition and the patterns found here should become more clear with additional studies and a larger number of lakes investigated.

At the other two lakes, shallow zone CH₄ bubble fluxes measured over coarse detritus gyttja are twofold higher at location M1 in Mellersta Harrsjön (40.6 mg $CH_4 \cdot m^{-2} \cdot day^{-1}$) compared to location I1 in Inre Harrsjön (18.3 mg CH₄ · m⁻² · d⁻¹; P = 0.029, n = 1064; Figure 3a). Apart from the stream inflow (see section 4), location M1 has dense growth of Sparganium angustifolium (Stilson et al., 2014) from which remnants are visible as coarse detritus in the sediment, compared to Myriophyllum alterniflorum or Potamogeton alpinus which dominate the shallow waters of Inre Harrsjön (Wik et al., 2011). Many submerged macrophyte species, possibly including those that grow in our lakes, have the ability to transport O₂ and other electron acceptors to and from the sediments, affecting redox potentials, CH₄ production, and burial (Carmichael et al., 2014; Hirota et al., 2004; Kankaala et al., 2003). The question is whether plant-induced redox processes vary depending on species composition, and if they can explain the significant difference in CH₄ bubble flux between locations I1 and M1, and why Inre Harrsjön consistently generates the least shallow zone ebullition of our three study lakes (see Wik et al., 2013). Unfortunately, because the sedimentation rate in our study lakes is low, roughly 1.5 mm/year on average (Kokfelt et al., 2009), the effect of vegetation growth on burial and emission is not measurable on these time scales using bubble traps. If we assume from Figure 2d that bubbles are not likely to form in the upper 10–20 cm of the sediment, it will, based on current sedimentation rates, take roughly 70–140 years before newly settled substrates are buried deep enough to contribute directly to ebullition, with the possibility of much longer time scales in the deeper zones.

Methane bubble fluxes from the deep zones of Inre and Mellersta Harrsjön (locations I2 and M2), underlain by fine detritus gyttja, are sixfold lower on average (4.7 mg $CH_4 \cdot m^{-2} \cdot day^{-1}$) than at the shallow coring sites of the same lakes where the gyttja is coarser (29.6 mg $CH_4 \cdot m^{-2} \cdot day^{-1}$; P < 0.001, n = 2151; Figure 3a). The



significant difference in ebullition between shallow and deep sites could partly relate to decreasing TOC quality, indicated by decreasing amounts of coarse organic detritus in the gyttja with increasing water depth. Also, the growth of macrophytes during summer as well as phytoplankton blooms contribute fresh organic material to the shallow zones, a seasonal loading that may be less significant in lakes' deeper parts, below the euphotic zone. The sedimentation at depth often relies on lateral transport and substrates that have been resuspended and partially decomposed prior to settling (Sobek et al., 2009). Although TOC quality likely contributes in regulating deep zone ebullition in our study lakes, the effects of temperature and energy transfer cannot be ruled out as the main controls on ebullition. The deep zone bubbles' response to temperature is slower than in the shallow areas due to overall colder water and delayed sediment warming (Figure S2; Wik et al., 2014), but after fall mixing when the thermocline breaks down and the temperature homogenizes, the CH₄ bubble fluxes are often similar across depths (Wik et al., 2013). This seasonal pattern would likely not be the case if differences in sediment type or quality were solely or largely responsible for the depth dependence of ebullition in our lakes. The question remains whether the deep zone sediments have potential to emit substantially more CH₄ than what they currently do if temperatures at depth were higher during most of the ice-free season.

7. Summary and Implications

In this study we investigate potential links between sediment characteristics and the spatial heterogeneity in CH_4 ebullition across three small, intensively studied subarctic lakes. Our results suggest that down-core variations in sediment CH₄ can hint at the depth of bubble formation, but overall sediment CH₄ is not a straightforward indicator of ebullition. Low amounts of CH₄ found at sites with high bubbling rates could possibly indicate rapid turnover of bubbles rather than substantial degassing during sampling. Further, we conclude that higher quantities of TOC in sediments are not synonymous with higher flux. The more or less equal bulk TOC in most cores is far from consistent with the bubbles' large spatial variation. Instead organic C quality seems to play a greater role, most likely in combination with previously established effects of water depth and temperature. The presence of macrophyte detritus as well as redeposited peats in shallow zones with significantly higher bubbling rates indicates at the importance of both in situ and terrestrial labile organic material. The sediments' TOC:TN points to a homogeneous and predominant autochthonous C source; however, additional carbon characterization is needed to confirm the C source material (s). Measurements of TOC: TS suggest a possible role of sulfate-reduction and sulfide precipitation, and future studies could illuminate the role of AOM in these lake sediments. Additional research, in both Stordalen and elsewhere, is also required to determine the influence of submerged vegetation on sediment quality and clarify whether or not submerged peat is particularly effective in sustaining ebullition in shallow subarctic water bodies.

Other studies have shown that macrophyte abundance and its related autochthonous organic loading will be affected by warmer waters and longer ice-free seasons (Mäemets et al., 2006; Rooney & Kalff, 2000; Short et al., 2016). Based on our discussion here, such feedback on aquatic vegetation could have a large effect on future CH₄ emissions, possibly to a greater degree than altered terrestrial runoff from increasing precipitation (McClelland et al., 2006) or the speculated C loading from thawing permafrost (Schuur et al., 2015; Walter Anthony et al., 2016). In addition, organic C is already plentiful and readily available in most lakes, including those that are shallow and of postglacial origin (as shown in this study) compared to often thicker permafrost soils, such as yedoma, of which many rely on seasonal active layers or thaw for decomposition. Thus, the many shallow postglacial lakes across high latitudes could be the most rapidly responding C source driven by Arctic warming.

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