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Key Points:

- Link between environmental forcing and CH₄ ebullition depends on peat structure
- Storage and transport processes within peat can "shred" CH₄ production signals
- Small differences in peat porosity produce large differences in CH₄ gas storage

Supporting Information:

• Texts S1 and S2 and Figures S1 and S2

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Ebullition of methane from peatlands: Does peat act as a signal shredder?

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Abstract Bubbling (ebullition) of greenhouse gases, particularly methane, from peatlands has been attributed to environmental forcings, such as changes in atmospheric pressure. However, observations from peat soils suggest that ebullition and environmental forcing may not always be correlated and that interactions between bubbles and the peat structure may be the cause of such decoupling. To investigate this possibility, we used a simple computer model (Model of Ebullition and Gas storAge) to simulate methane ebullition from a model peat. We found that lower porosity peat can store methane bubbles for lengthy periods of time, effectively buffering or moderating ebullition so that it no longer reflects bubble production signals. Our results suggest that peat structure may act as a "signal shredder" and needs to be taken into account when measuring and modeling ebullition.

1. Introduction

Methane (CH₄) is a greenhouse gas with a global warming potential much greater than carbon dioxide [Myhre et al., 2013], and a major source of naturally occurring CH₄ is peatlands [Blodau, 2002]. Ebullition in peat refers to the transport (to the ground surface) of CH₄ bubbles that form in peat pore water. As bubbles travel upward through a peat column, they can accumulate behind-existing bubbles lodged in pore necks [Baird and Waldron, 2003; Strack et al., 2005; Kellner et al., 2006] or underneath woody layers or well-decomposed layers of peat [Rosenberry et al., 2003; Glaser et al., 2004]. Where gas bubbles are not trapped and the bubbles are emitted steadily, most of the CH₄ in them will be readily consumed above the water table within the oxic layer if the water table is not at or above the peatland surface [Rosenberry et al., 2006]. However, if bubbles accumulate and are released episodically, the CH₄ reaching the water table may advect rather than diffuse through the oxic zone and bypass methanotrophic consumption [Coulthard et al., 2009]. As such, these episodic ebullition events can become major sources of CH₄ emissions from peat to the atmosphere and could be larger than emissions produced by diffusive and plant-mediated transport [Baird et al., 2004; Glaser et al., 2004; Comas et al., 2011]. Recent developments in field methods [Burrows et al., 2005; Comas and Wright, 2012] have made it possible to record CH₄ ebullition from boreal and subtropical peatlands at high temporal resolution. These advances have allowed researchers to investigate linkages between ebullition and a range of environmental factors or forcings that affect CH₄ production, consumption, and transport. For example, Goodrich et al. [2011] found cyclical—diurnal and seasonal—variations in ebullition. However, they were unable to isolate the environmental variable(s) responsible for diurnal cycles, although seasonal cycles were probably related to overall CH₄ production as mediated by peat temperature and the production of labile substrates. Ebullition may also be characterized by noncyclical "spikes" in flux (episodic ebullition) that have been linked to processes that alter bubble volume and mobility such as short-term (hourly) changes in atmospheric pressure [Tokida et al., 2007, 2009; Comas et al., 2011] or longer-term (days to weeks) variations in water table position [Glaser et al., 2004; Bon et al., 2014].

Studies have also shown that ebullition can occur in the apparent absence of environmental forcing and that relationships between ebullition and environmental factors are not always clear-cut. For example, Waddington et al. [2009] monitored the ebullition flux of CH₄ from laboratory-incubated samples of peat for 178 days. They recorded 339 pressure periods (periods during which atmospheric pressure was consistently increasing or decreasing), but only in 28% of these periods did episodes of ebullition occur. In addition, and contrary to Tokida et al. [2007] who found that falling atmospheric pressure was a trigger,

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increases in pressure can also cause ebullition [Comas and Wright, 2012, 2014; Klapstein et al., 2014]. It is also possible that ebullition from peat may be similar to ebullition from lakes [Wik et al., 2013], where changes in atmospheric pressure trigger large degassing events that exhaust the available store of gas. When this occurs, ebullition from proximate changes in atmospheric pressure will be minimal.

Some of the differences between the studies cited above may be explained by differences in the measurement and data analysis methods used by their authors. However, notwithstanding this note of caution, a common theme of these studies is that ebullition data are often "noisy" with different environmental forcings identified in different studies, which suggests that other controls on the system need to be considered, with an obvious candidate being the structure of the peat [Comas et al., 2014; Chen and Slater, 2015]. The structure of the peat and its down-profile variation will affect bubble storage and movement and therefore how production and consumption influence fluxes from the peatland surface. Structural effects are clear at large scales (e.g., tens of meters across peatlands and through the whole peat profile), where layers of woody peat may act as barriers to upward bubble migration. Bubbles accumulate below these barriers until the increase in buoyancy causes the barrier to rupture; the bubbles are then released, after which the barrier reseals [Rosenberry et al., 2003; Glaser et al., 2004]. At smaller scales (e.g., the upper ~30 cm of peat profile) and in peat in which woody layers are absent, Panikov et al. [2007] observed episodic ebullition from peat cores in which there were clear diurnal oscillations in CH₄ production at depths down to 20 cm. Although no mechanism was identified to explain why ebullition could not be linked to the forcing, Panikov et al. [2007] suggested that the capacity of peat to store gas and release it at a later time provided a possible explanation. In sediment systems, Jerolmack and Paola [2010] observed similar behavior where even in simple physical models, a one-to-one correlation could not be made between forcing and system response. They suggested that internal or autogenic processes were responsible for "shredding" any discernible response to certain-sized external forcings [Jerolmack and Paola, 2010].

The evidence from the peatland studies cited above suggests that the peat profile cannot be considered a simple entity that responds linearly to CH₄ production and consumption or to the processes that affect production and consumption. It is difficult to examine the role of peat structure experimentally because of problems imaging bubbles within peat over short time periods [Kettridge et al., 2011; Chen and Slater, 2015] and because of the difficulty in controlling the production signal within the peat profile. As an alternative approach, we explored the effects of peat structure using a computer model that provides a plausible representation of bubble dynamics in porous media [Ramirez et al., 2015]. Although not a substitute for investigating real peat profiles, our model results suggest that peat structure does, indeed, determine the degree to which CH₄ production signals affect ebullition flux at the peatland surface and helps identify ways storage effects may be investigated experimentally and thus incorporated into existing wetland methane models.

2. Method

The cellular automaton Model of Ebullition and Gas storAge (MEGA) [Ramirez et al., 2015] was used. It was chosen because (1) it is capable of simulating bubble storage and movement within porous media at large spatiotemporal scales (>1 m and >1 month), (2) it can explicitly represent the spatial heterogeneity of porous media, and (3) it produces magnitude and frequency distributions of ebullition similar to those obtained from peat [Kellner et al., 2006; Goodrich et al., 2011; Stamp et al., 2013; Yu et al., 2014] (see supporting information for details about model tests). The model conceptualizes peat as a twodimensional cellular grid. Peat solids, and therefore the pore structure, are represented as "shelves," and the number and size of these can be set according to the physical characteristics of the peat; highly porous peat requires fewer shelves than denser, lower-porosity, peat (Figures 1a and 1b). The spaces between the shelves may be occupied by free-phase gas or by water. The movement of gas bubbles within the peat is governed by a simple rule set adopted from an avalanche model [Bak et al., 1987]. In MEGA, gas accumulates under shelves in a manner akin to an inverted pile of idealized sand grains, and the steepness of the pile determines if gas will "avalanche" upward to shallower shelves or whether it remains stationary and accumulates. The avalanching process encapsulates the opposing forces of buoyancy and surface tension that act upon free-phase gas within porous media [Corapcioglu et al., 2004;

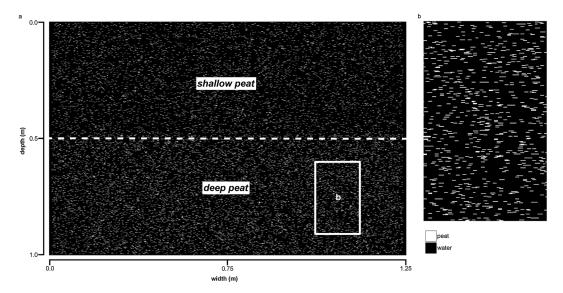


Figure 1. (a) Shelves randomly arranged to represent peat profile (1.5 m wide, 1 m deep) with higher porosity at shallower depths (0.0-0.5 m) and lower porosity at deeper depths (0.5-1.0 m). (b) Inset of peat with shelves (in white) saturated with water (in black).

Ramirez et al., 2015]. Unlike the avalanche model of Bak et al. [1987], MEGA comprises collections of bubbles under multiple shelves that may release their bubbles at different times. As bubbles are released from one shelf, they may travel directly to the water table or become trapped under another shelf; therefore, in the model, there is interaction between shelves (see supporting information for details about MEGA).

To test the idea that peat structure can shred environmental signals in ebullition, we used a series of model scenarios to represent peats of varying pore structure and introduced a range of gas production signals into the peat profiles at different depths. These production signals were chosen to represent different known environmental forcings on methane/bubble production in peat. If evidence of the production signal pattern is not present in the ebullition at the peat surface, our model peat has shredded the production signal, thus severing any link between bubble production and release.

Our modeled peat profiles represent an area 1.5 m wide and 1 m deep (thick), with a grid cell size of 1 mm × 1 mm which resulted in a model domain of 1,500,000 cells (Figure 1a). Published measurements of Sphagnum branches (average shelf length = 5.7 mm, standard deviation = 0.8 mm) within a poorly decomposed peat [Kettridge and Binley, 2008] were used to set the length of the shelves. The peat profile was partitioned into two layers of equal depth/thickness to reflect the spatial variation in decomposition often found in peats [Clymo, 1984]. The shallower layer represented less decomposed peat of greater porosity, while the deeper layer was less porous to represent peat undergoing compression and more advanced decomposition [Quinton et al., 2000, 2008]. Values of porosity used in the model were within measured ranges from shallow peats (91–98%) [Kettridge and Binley, 2008, 2011; Parsekian et al., 2012]. Three model peat profiles were created, all with a shallow layer porosity of 95% and with a deeper layer porosity of 95%, 93%, or 92%. In each layer, the shelves were positioned randomly.

Bubble production in MEGA was based on data from Stamp et al. [2013], who reported maximum, seasonally averaged bubble fluxes of 709 mL m⁻² d⁻¹ from Sphagnum lawns in a Welsh raised bog. Converting the field measurements into model, CH₄ production rates took into account that the modeled peat represents a twodimensional cross section rather than a three-dimensional volume. The smallest bubble within MEGA is 1 mm², and this bubble size was selected to be similar in size to bubbles measured within peat (0.074–2.25 mm²) [Kettridge and Binley, 2008]. We drove the model with four production signals based on the patterns observed by Panikov et al. [2007], each comprising three different subsignals, as shown in Figure 2. These subsignals represented diurnal bubble production, steady production, and spikes of production that were each added to the modeled peat profile at three depth zones to reflect the spatial variability in CH₄ production [Sundh et al., 1994; Frenzel and Karofeld, 2000; Strack and Waddington, 2008]. The diurnal signal

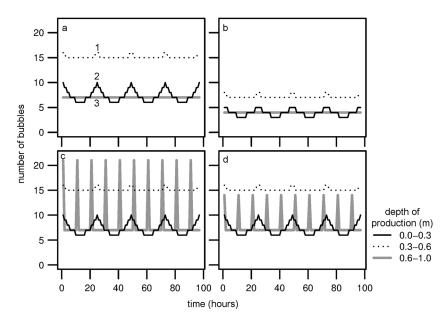


Figure 2. Methane production subsignals that are (1) weakly diurnal, (2) strongly diurnal, and (3) steady. (a) Strong diurnal (SD) CH₄ production signal decomposed into subsignals 1, 2, and 3, consisting of 50%, 25%, and 25% of the daily CH₄ production, respectively. (b) Weak diurnal (WD) CH₄ production consisting of 50% of SD subsignals. (c) Large spike (LS) signal consisting of SD plus 200% increases in steady production occurring every 10 h. (d) Small spike signal (SS) with 100% increases in steady production.

will be related to temperature changes in the peat profile affecting CH₄ production and variations in root exudation from vascular plants (exudates being a substrate for methanogens). The spikes of bubble production are related to sudden reductions in atmospheric pressure or increases in temperature that cause bubbles to come out of solution. Of the four main signals, one was strongly diurnal (SD), one weakly diurnal (WD), one had large spikes concurrent with a strongly diurnal signal (LS), and one had small spikes concurrent with a strongly diurnal signal (SS). *Jerolmack and Paola* [2010] showed that shredding is sensitive to signal frequency. Given our focus on peatlands, we fixed the frequency, and varied the amplitude of CH₄ production signals to match those known to be realistic for peat; hence, we did not consider a wider range (of possibly speculative) frequencies.

A simulation was run for every peat profile and production signal combination. With three peat profiles (low, medium, and high porosity) and four production signals (SD, WD, LS, and SS), this resulted in a total of 12 simulations. During every model hour, a quantity of bubbles corresponding to the production subsignals was added to the peat profile within the specified depth zone at random locations. To avoid edge effects, bubbles were not added within 0.25 m of the left- and right-hand edges of the profile. Using theoretical relationships between bubble size and rise velocity within a porous medium [Corapcioglu et al., 2004], it was estimated from median bubble size, calculated from a preliminary simulation, that bubble velocity would be a constant 6 mm s⁻¹. Ebullition flux was recorded at the top of the peat profile, which was also the position of the model water table, at hourly intervals. As no peat in our model setups existed above the water table, we did not simulate the consumption of CH₄ by methanotrophic bacteria. Each peat profile was driven by its production signal until the 10 day average ebullition flux stabilized, and output data after this time period were analyzed.

For each simulation, 15,000 model hours of flux output were analyzed for evidence of periodicity by estimating power spectra using a multitaper method [Thomson, 1982]. This method was chosen because it performs a harmonic F variance-ratio test (F test) for each frequency and can be used to distinguish between noise and significant peaks in spectra. Due to the large number of flux records, it is possible that random fluctuations in flux can periodically occur and produce inflated F values. To account for this artifact, Thomson [1990] suggests that significance levels for nonrandomness are set at 1-1/N, where N is the number of flux samples. Using this recommendation, peaks in spectra were significant at the 99.99%

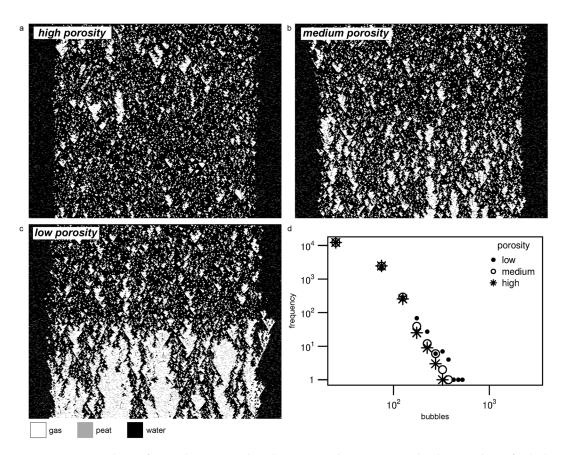


Figure 3. Gas accumulation after simulating strong diurnal (SD) CH₄ production in peat with a deep peat layer of (a) high, (b) medium, and (c) low porosity. (d) Frequency plots of corresponding hourly ebullition flux. Bin sizes are 50 bubbles.

level. Dominant peaks in spectra were located at frequencies corresponding to the production cycles every 24 h and spikes occurring every 10 h, and peaks in spectra ±1 h from those locations were also inspected to account for any lag effects in flux.

3. Results

Gas storage measured at the conclusion of the three SD simulations clearly shows that decreasing peat porosity increases gas storage (Figures 3a-3c); for peat of high, medium, and low porosity, the percentage of the profile consisting of stored gas was, respectively, 19, 25, and 35. The effect of the additional gas storage is evident in the magnitude and frequency of hourly ebullition fluxes that are strongly right skewed (Figure 3d). Although the difference in total and mean fluxes between the three simulations is minimal (<1%), which is to be expected (because, over time, model input (CH₄ production) is equivalent to model output (ebullition)), the lower-porosity peat is able to store more gas and produce extreme gas flux events that rarely, or never, occur in peat of medium or high porosity. This result demonstrates that structurally different peats can generate the same amount of ebullition but different kinds of ebullition. In our model, lower-porosity peats produced more erratic ebullition, while higher-porosity peats generated steadier ebullition.

Four out of the six diurnal simulations did not shred the diurnal production signal as can be seen in Figure 4, where the highlighted spectrum peaks signify the occurrence of diurnal ebullition. Figures 4a-4c clearly show that SD production signals are always measurable in gas flux at the peat surface (frequency~0.04, which is nearly a 24 h cycle). For WD production, a signal was detectable in high-porosity peat (Figure 4d), but no significant peaks in spectra were found for peat of medium and low porosity (Figures 4e and 4f).

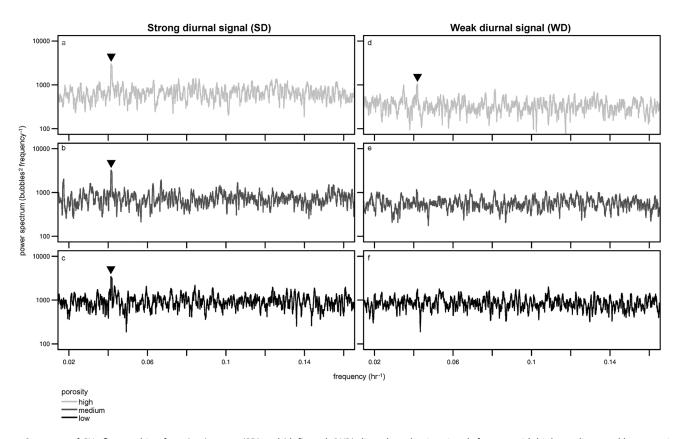


Figure 4. Spectrum of CH_4 flux resulting from (a–c) strong (SD) and (d–f) weak (WD) diurnal production signals for peat with high, medium, and low porosity. The triangles indicate significant spectrum peaks (frequency~0.04) that represent periodic flux response to diurnal input signal.

Only two of the simulations with spiked signals produced ebullition with spiked fluctuations occurring every 10 h (frequency~0.1). This signal was noticeable from high-porosity peats (Figures 5a and 5d), but no evidence of spikiness was noticeable in flux from medium- and low-porosity peats (Figures 5b, 5c, 5e, and 5f). Moreover, diurnal signals were detected in all spiked signal simulations except for the lowest-porosity peat with the weaker spiked signal (Figure 5f).

4. Analysis and Conclusions

This study suggests that the link between environmental forcing and CH_4 ebullition flux is dependant on peat pore-scale structure. If structure had no or little impact, the bubble flux at the surface would mirror the integrated production signals. Model peat with high porosity stores less gas, and fluctuations in gas production or bubble mobility at depth translate into losses at the peat surface with minimal time delay. Thus, the openness of the peat structure imparts minimal interference on the original bubble production signal, and traces of this signal exist in the flux. In contrast, lower-porosity peat can entirely decouple environmental forcing and flux response. The mechanism responsible for this decoupling is pore-scale gas accumulation, storage, and release. In lower-porosity peat, large amounts of gas are stored within the peat matrix and released at times unrelated to the original production fluctuation. This decoupling occurs in simulations with weaker signals and medium- to low-porosity peats. A secondary effect of lower porosity and greater bubble storage is the possibility of producing unsteady bubble flux containing more moderate to large bubbling events (Figure 3d). The overall effect of these events is to produce background noise within the bubble flux that further masks the presence of the bubble production signals.

Importantly, the porosity of the deep peat layers does not change dramatically (92–95%), but the resulting signal shredding is very different. For example, a 2% difference in porosity can affect whether a diurnal (Figures 4d and 4e) or spiky (Figures 5a and 5b) production signal is no longer present in the bubble flux. In the model, this difference in peat porosity contributes to greater amounts of gas storage and more

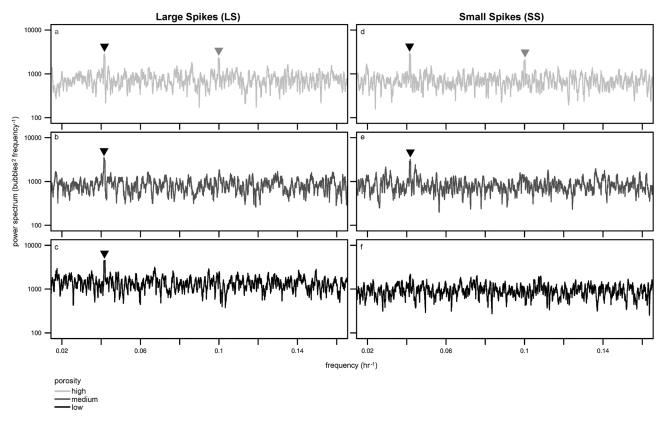


Figure 5. Spectrum of CH_4 flux resulting from a strong diurnal production with (a–c) large spikes (LS) and (d–f) small spikes (SS) for peat with high, medium, and low porosity. The gray triangles indicate significant spectrum peaks (frequency~0.1) that represent flux response to spikes in input signal occurring every 10 h. The black triangles indicate significant spectrum peaks that represent diurnal flux response.

signal shredding. Although no study to date has explicitly investigated if signal shredding occurs in peats, it has been observed that small differences between measured peat porosities (1–4%) can double the amount of gas stored [*Strack and Mierau*, 2010] and affect ebullition [*Strack et al.*, 2006]. With gas storage in peats being sensitive to small structural differences, it is likely that signal shredding will vary greatly over a peatland, and this may explain the difficulty in correlating ebullition to environmental forcings. Lastly, we find that, regardless of peat porosity, bubble flux from strong bubble signals, occurring diurnally or as spikes, can be correlated to the environmental forcing causing the change in bubble production. Given that peat structure imparts a strong influence on the timing and size of ebullition events, we suggest that peat structure should always be quantified, and in locations where peat porosity is low or relatively low, caution is taken when attempting to link ebullition to environmental forcings. We recommend that additional experimental work on peat mesocosms be performed to better understand the effects of peat structure on ebullition and environmental forcings. Evidence from our study also highlights the importance of spatially representing peat pore structure and gas storage processes in wetland CH₄ models. We suggest the inclusion of these processes in models to reduce uncertainty in CH₄ emission predictions.

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