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Key Points:
- A reduced-complexity model for ebullition and gas storage in peat is tested against a natural system for the first time
- Physical structure of peat is an important control on the magnitude and frequency of ebullition
- Gas bubble distribution in the peat column shows differences with depth associated with changes in peat porosity

Supporting Information:
- Supporting Information S1

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Methane Ebullition From Subtropical Peat: Testing an Ebullition Model Reveals the Importance of Pore Structure

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Abstract The role of subtropical peatlands as a source for methane gas is not well understood, partly due to uncertainties surrounding environmental controls on gas ebullition patterns. Past studies have pointed to an array of environmental factors controlling ebullition, although we have found that ebullition patterns can be replicated by a model considering only physical parameters of the peat matrix. Here we tested a computer model for gas ebullition and storage against a natural system for the first time, using a suite of field measurements in the Florida Everglades. Modeled ebullition showed patterns similar to those observed in the field in terms of frequency distribution and magnitude, specifically from areas of higher density peat fabric. These results suggest that the internal structure of the peat soil is an important control on spatial and temporal patterns of ebullition in the Everglades and should be considered when investigating environmental controls on ebullition patterns.

Plain Language Summary It is widely accepted that peatlands are major sources of global methane gas. However, their role is not well understood, partly due to the effects environmental variables have on methane release patterns. Here we present a test of a computer model for methane ebullition and storage in peatlands, and our findings point to the importance of the physical parameters of the peat matrix in controlling timings of release events. This is important, as most studies look at environmental factors as the main control on methane gas releases, and our findings suggest that physical parameters of the peat soil alone can be largely responsible for the unpredictable patterns seen in methane releases.

1. Introduction

Peatlands are one of the major sources of naturally occurring CH_4 (methane), a greenhouse gas with a global warming potential much greater than carbon dioxide (Myhre et al., 2013). CH_4 develops naturally as a metabolic by-product, forming free-phase bubbles within the peat matrix or dissolved into pore water (Whalen, 2005). CH_4 that exists as free-phase gas accumulates within pore spaces and is eventually transported upward to the peat surface and released into the atmosphere (i.e., ebullition). Studies in boreal peatlands have provided evidence that ebullition is an important pathway for CH_4 transport to the atmosphere (Baird et al., 2004; Glaser et al., 2004; Stamp et al., 2013), showing correspondence between episodic CH_4 ebullition and environmental factors, such as increases or decreases in atmospheric pressure (Comas et al., 2011; Glaser et al., 2004; Klapstein et al., 2014; Tokida et al., 2007), or depressuring effects due to changes in water table level (Bon et al., 2014; Chen & Slater, 2015; Glaser et al., 2004; Peltola et al., 2018). In subtropical and tropical peatlands, where less is known about the importance and controls of CH_4 ebullition, few studies have quantified ebullition rates (Lawson et al., 2015), though it has been shown that ebullition occurs erratically in time (Comas & Wright, 2012, 2014), is highly variable in space (Wright & Comas, 2016) and consequently difficult to measure (Nahlik & Mitsch, 2011; Shoemaker et al., 2015). It has also been shown that ebullition rates in subtropical peatland systems are comparable to boreal peats, although extremely episodic, and that increases in atmospheric pressure may trigger large ebullition events by reducing bubble volume and increasing bubble mobility within the peat matrix (Comas & Wright, 2012, 2014).

While the main driver for CH_4 gas ebullition is CH_4 gas production (which in turn requires a sufficient supply of labile carbon for methanogenic consumption in order to produce enough CH_4 to saturate pore waters with dissolved CH_4 and produce gas bubbles), this experiment assumes that these processes and resources are already in place. Since our field sites did actively produce CH_4 gas, we therefore focus on controlling factors for the transport of free-phase CH_4 gases after they are produced. Large ebullition events may be the result of gas reaching a volume threshold within the peat matrix (Baird et al., 2004; Chen & Slater, 2015; Comas et al., 2014; Klapstein et al., 2014). When a bubble's upward movement is stopped by impermeable layers or constrictions in the peat pore structure, gas accumulates until buoyancy overcomes the forces holding the gas in place. These impermeable layers of well-decomposed peat or woody materials which enhance the shear strength of peat structure act as gas traps and are commonly observed in northern systems (Comas et al., 2014; Glaser et al., 2004; Rosenberry et al., 2003). Nonetheless, the degree to which peat structure controls ebullition still remains unclear because episodic ebullition can also occur in peat soils without woody confining layers (Kellner et al., 2006; Ramirez et al., 2016; Strack et al., 2005; Yu et al., 2014).

The aim of this study was to test the performance of a simple computer model of gas dynamics in porous media against field ebullition observations, while investigating the role of peat structure in CH_4 ebullition magnitude and frequency. We used the Model of Ebullition and Gas storAge (MEGA; Ramirez et al., 2017, 2015a, 2015b) to simulate bubble accumulation, storage, and release in subtropical peat because (1) it is difficult to directly measure peat structure and gas storage at the pore scale, (2) it allows for the investigation of structural effects on ebullition in isolation from environmental forcing, and (3) it has been the focus of several recent studies but has never been tested against a natural system. While it has been demonstrated that MEGA simulates different ebullition patterns using the same production rate but different peat structures (Coulthard et al., 2009), our study is the first to test a model for ebullition that explicitly considers peat structure against observations of naturally produced ebullition at the field scale while considering a two-dimensional distribution of bubbles. Here we used physical characteristics and gas production rates measured in the Florida Everglades (see section 2) as inputs to MEGA, and compared modeled outputs to long-term, high-frequency ebullition and gas content observations in the field.

2. Methodology and Experimental Design

MEGA is a cellular automation designed to replicate two-phase flow (i.e., free-phase gas and liquid) in porous media on a two-dimensional grid (Ramirez et al., 2015a, 2015b, 2017; see supporting information for model description and supporting information data set for MEGA source code; Wright et al., 2018). MEGA is a reduced-complexity model (Larsen et al., 2014), which is computationally efficient, with low data requirements, that uses physical peat properties and estimated total free-phase CH_4 gas production as direct inputs. As implemented here, MEGA simulates a peat profile, with the pore structure represented as shelves (solid), and surrounding grid locations as either CH_4 bubbles (gas) or water (liquid). Gas storage and movement within pores are replicated using a modified rule set for avalanches in sand piles (Bak et al., 1987). Gas accumulates underneath shelves and is transported upward as bubbles *avalanche* onto shallower shelves or exit the profile as ebullition. Although MEGA does not include a detailed representation of the complex physics of two-phase flow, it does account in its rule set for the fundamental processes that control bubble accumulation and movement (Ramirez et al., 2015b).

We used porosity and peat depth to create a model domain (see supporting information for more explanation) for each of two field sites in a subtropical peatland (the Florida Everglades) and then input gas production rates for each field site. Gas bubble production rates were estimated using the mass balance approach outlined in Wright and Comas (2016). MEGA then outputs continuous values for gas storage and ebullition, which were compared to the observed ebullition values from the same field sites.

The two field sites used in this study were located in the subtropical Florida Everglades, in Water Conservation Area 3. Both sites corresponded to sloughs and therefore remained inundated during the course of this study, and typically remain inundated throughout the year. Peat soils at the sites contain mostly remnants of sawgrass (*Cladium*) and lily (*Nymphea*). Humification was estimated for each site, with von Post humification (Ekono, 1981) scores ranging from H3 to H5 (very slightly to moderately decomposed peat) at Site 1 and H2 to H4 (undecomposed to slightly decomposed) at Site 2. A platform was built at each site to take weekly measurements of CH_4 gas flux and in situ gas content of the soil, which were used to calculate an estimated overall CH_4 bubble production rate from each site using the methods outlined in Wright and Comas (2016).

Measurements of gas bubble fluxes were taken in the field using a gas trap fitted with a time-lapse camera at each site. Gas traps consisted of an inverted 20 cm plastic funnel feeding into a clear PVC graduated cylinder

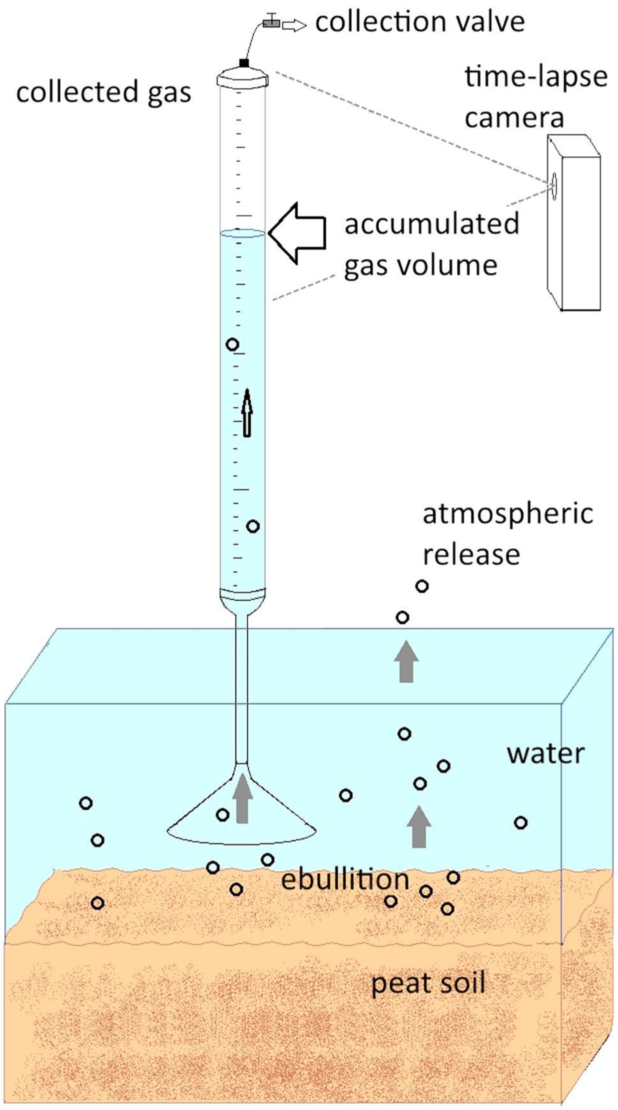

Figure 1. Schematic of improved gas trap for monitoring gas flux rates with a time-lapse camera in an inundated wetland.

extending above the water surface, where gas accumulated for weekly collection via a valve at the top of the chamber (Figure 1). The gas volume collected at weekly intervals was analyzed for CH_4 and carbon dioxide composition using gas chromatography and used to estimate a weekly free-phase gas flux. Time-lapse cameras captured gas bubble flux values with much higher frequency (i.e., 30 min) between weekly measurements, following methods outlined in Comas and Wright (2012). This improved gas trap design was unique to this study and well suited for camera monitoring, as it allows for direct visualization and measurement of gas volumes accumulating over time.

In situ gas volumes were measured in the field using time-lapse ground penetrating radar (GPR) surveys. The complex refractive index model is a three-phase mixing model, which essentially converts dielectric permittivity, measured using GPR, to gas volume in peat soils below the water table. For a more complete description of the GPR method and using complex refractive index model to find gas contents within peat soils, see Comas et al. (2007, 2011). Weekly gas content measurements were used in conjunction with weekly gas trap data to estimate a weekly average of CH_4 gas bubble production rates using the model outlined in Wright and Comas (2016). This model is based on a mass balance approach, where total free-phase CH_4 gas production is the summation of CH_4 gas lost via ebullition, diffusion, methanotrophic consumption, and net change in soil gas content between GPR measurements. The resulting free-phase CH_4 gas production rate was then used as an input to MEGA.

For all MEGA simulations, gas was added at a constant production rate. To account for uncertainty in production estimations and seasonal changes in gas production, three levels of gas production (low, medium, and high) were modeled separately per site. These three gas bubble production levels were the mean production minus one standard deviation (low), mean production (medium), and mean production plus one standard deviation (high). For Site 1, low, medium, and high production rates were 388, 488, and 588 ml $CH_4 \cdot m^{-2} \cdot day^{-1}$, and Site 2 rates were 182, 280, and 379 ml $CH_4 \cdot m^{-2} \cdot day^{-1}$. Production rates were scaled to two-dimensions and added as 1 mm^2 sized bubbles at random locations within each profile and evenly across every 24-hr period.

A total of six simulations was performed, with two sites modeled and three gas production levels per site. Each MEGA simulation was initially run for a duration of 7,800 modeled days until changes in gas content within the shelf structure stabilized. Afterward, a 200-day simulation was performed to estimate median bubble size from both sites with medium production. Bubbles within MEGA cannot accelerate, and bubble velocity is not dependent on bubble size. Therefore, all bubbles in MEGA, regardless of size, have a fixed terminal velocity. Median bubble size was used to estimate the terminal velocity of all bubbles in both simulations, using theoretical relationships between bubble size and rise velocity within a porous medium (Corapcioglu et al., 2004), resulting in a bubble velocity of 1 mm/s. In our simulations, the top of the pore structure represented the peat surface, and no changes in water table or atmospheric pressure were modeled. Following field sampling methods, bubbles reaching the top of the peat (ebullition) were collected in 30-min intervals by simulated gas traps with a width of 200 mm (matching the dimensions of gas traps used in the field). A total of five gas traps was placed side-by-side across each peat profile (Figures 2a and 2b, T1–T5). Modeled ebullition in each simulated gas trap was upscaled to a rate of milliliter per meter squared per day and compared to the observed ebullition per site using a Kolmogorov-Smirnov test (Massey, 1951) to calculate the largest vertical difference between the observed and simulated empirical cumulative distribution functions (D). For each site, modeled ebullition collected in the gas trap having both the lowest D statistic and simulated ebullition within the range of observed ebullition was selected as the best matching gas trap (see supporting information for further explanation). Although the

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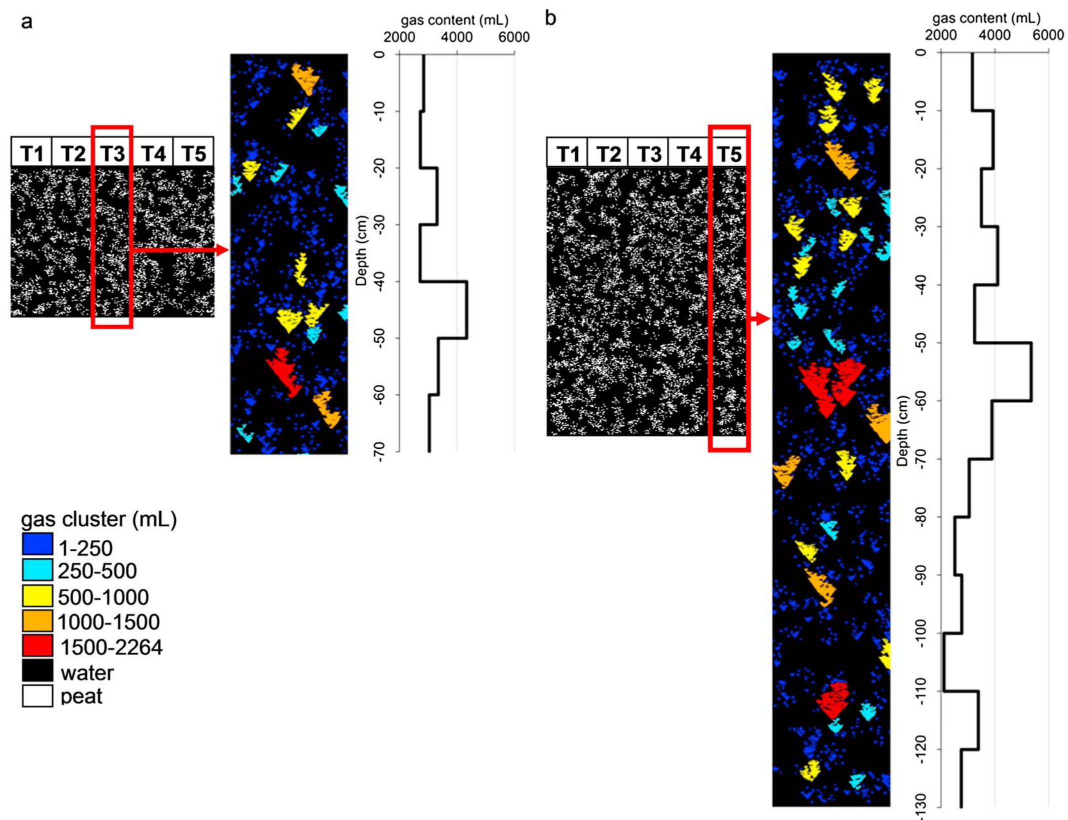

Figure 2. Modeled spaces for (a) Site 1 and (b) Site 2, with all funnel locations (T1–T5) shown. Model areas under simulated gas traps that best match observed ebullition from the field are outlined in red and shown in detail. Volume of gas clusters for each best matching gas trap are shown in color scale, with warm colors symbolizing larger clusters of gas. Finally, gas volume with depth at a sampling interval of 10 cm is shown in a 1-day plot.

spatial heterogeneity of the modeled peat underneath each gas trap is quite similar, we identify the gas trap that best replicates the observed ebullition and utilize the pore structure underneath this gas trap to suggest an explanation for the observed ebullition. Histograms of (1) simulated ebullition, with normalized frequency (and bin spacing equivalent to a 1.5-mm change in gas trap water level) from the gas trap that best replicated ebullition, and (2) observed ebullition were fitted with power law distributions. Gas content in MEGA was the proportion of gas within the shelf structure area underneath the best gas trap at the completion of simulations forced with average CH_4 production (Figure 2). Detailed gas content analysis with depth was performed by measuring the sizes of individual gas clusters and changes in gas content at 10-cm depth increments and converted to volumes by assuming a third dimension with a length of 1 m.

A large peat monolith was also collected from the field at Site 1 to investigate gas storage with depth at the lab scale. This sample, measuring 0.2 by 0.2 m laterally and 0.7 m vertically, was collected in an intact block, mounted in an acrylic tank, and allowed to equilibrate for 4 months in an environmentally controlled chamber with water level held constant at 10 cm above peat surface and temperature held at 25 °C to simulate field conditions. A series of discrete zero offset profile GPR measurements were collected vertically, yielding a one-dimensional depth profile of EM wave velocity (and thus gas content). After GPR measurements were completed, porosity was measured by comparing weights of saturated and dried volumes collected from the sample at several depths.

3. Results

Ebullition events and storage values estimated from MEGA were compared with the observed values from gas traps (ebullition) and GPR (storage) in the field. Ebullition measurements from the field sites are shown in

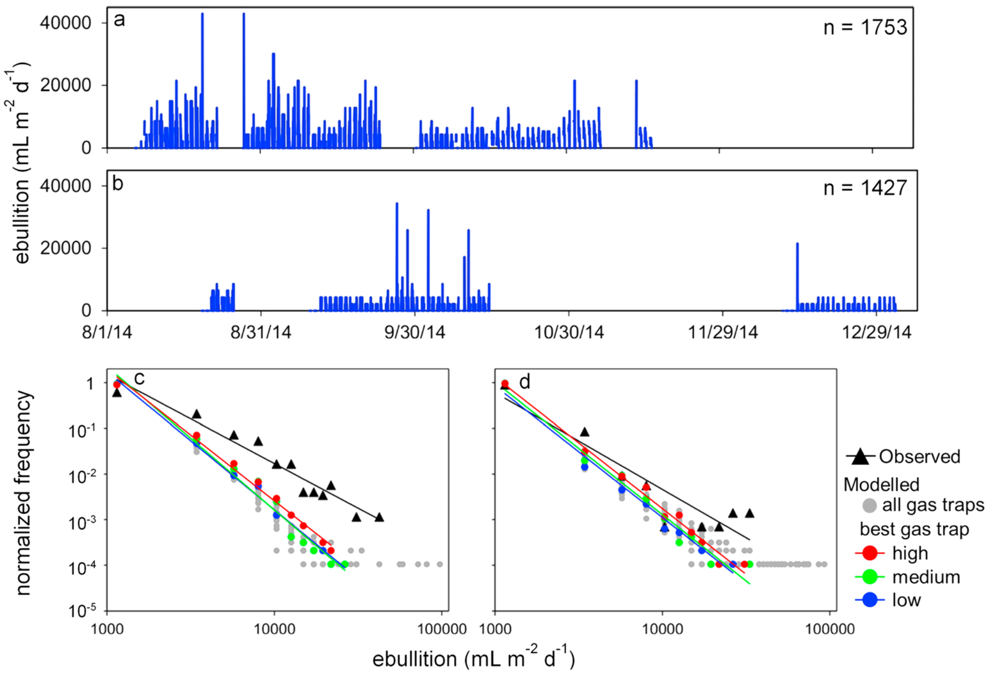

Figure 3. Observed CH_4 gas ebullition rates from the field for (a) Site 1 and (b) Site 2. Magnitude and frequency of observed and modeled ebullition from all gas traps and best gas trap with low, medium, and high gas production for (c) Site 1, and (d) Site 2. All fitted power law distribution have $p < 0.05$ and $R^2 > 0.82$.

Figure 3. There are enough data points ($n > 1,400$) for each site to lend statistically significant insights, despite gaps resulting from limited site access and equipment failures. These data show a pattern of steady background ebullition averaging around 2,933 $ml \cdot m^{-2} \cdot day^{-1}$ from Site 1 and only 992 $ml \cdot m^{-2} \cdot day^{-1}$ from Site 2, which were combined with much larger episodic releases of more than 42,000 $ml \cdot m^{-2} \cdot day^{-1}$ from Site 1 and 34,000 $ml \cdot m^{-2} \cdot day^{-1}$ from Site 2. While steady ebullition is comparable to others recorded in the Everglades, the episodic releases are higher than previously reported. For comparison, Comas and Wright (2014) reported background free-phase gas fluxes of 1,100–1,200 $ml \cdot m^{-2} \cdot day^{-1}$ and episodic ebullition rates of just above 9,400 $ml \cdot m^{-2} \cdot day^{-1}$. We attribute the disparity in episodic ebullition rates in part to an improvement in measurement resolution. While Comas and Wright (2014) was based on an hourly ebullition rate, this study used an improved gas trap design and monitoring process, with 30-min measurement intervals. These large episodic ebullition events are extremely short lived and would not realistically scale up to a daily average. Ebullition observations were arranged into histograms and compared to the ebullition outputs from MEGA (Figures 3c and 3d). Overall, variation between modeled and measured ebullition from each site is low. Both modeled and observed ebullition histograms show a pattern of frequent small ebullition events, combined with fewer but much larger episodic events. These negative power law distributions all show values of $p < 0.05$ and $R^2 > 0.82$.

In situ gas content measured in field sites using GPR averaged 14.9% by volume at Site 1 and 15.1% at Site 2 when averaged over the full length of the peat column. In MEGA, the value of gas storage along the full length of the peat column (Figure 2) showed very similar values of 16.6% in the Site 1 model and 17.5% in the Site 2 model; a difference of 1.7% and 2.4% by volume from observed values. One-dimensional profiles of gas content with depth under best modeled gas traps (Figure 2) highlight the contrast between (1) open areas (i.e., well-connected pore spaces) containing small clusters of gas (<1,000 ml assuming a 1-m-long model space) and (2) closed areas, as defined by proximity of shelves (i.e., poorly connected pore spaces), containing large gas clusters (>10,00 ml assuming a 1-m-long model space), which are located deeper within the peat profile (i.e., 40- to 50-cm depth in Site 1 and 50- to 60-cm depth in Site 2).

In situ gas content in the lab sample taken from Site 1, measured along the peat column using GPR, ranged from 8% to 21% gas content by volume (multicolored lines in Figure 4). The sample's measured porosity

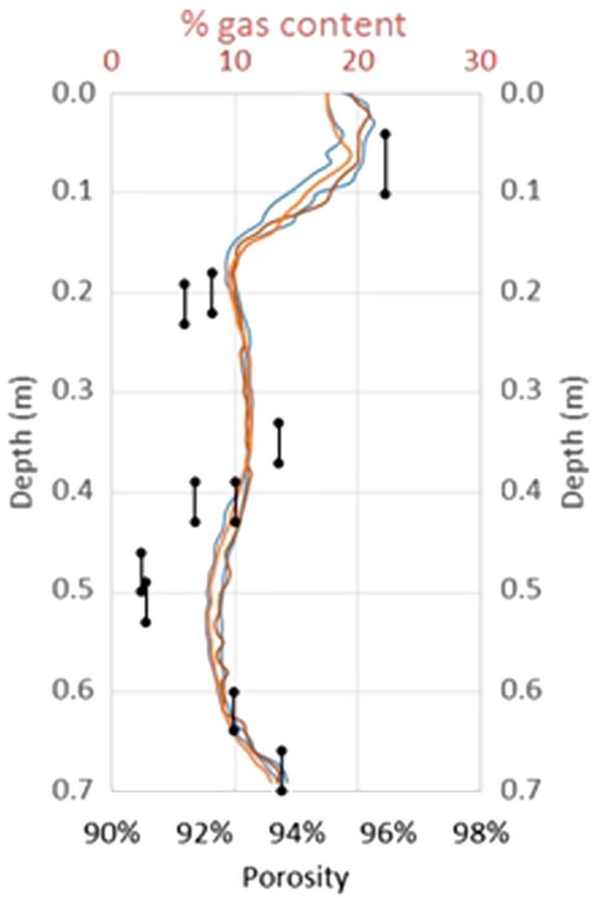

Figure 4. Gas contents in the lab sample measured using ground penetrating radar are shown as multicolored lines, where each line represents a single ground penetrating radar profile, and measured porosity values are shown as black points and lines. Depths on the *y* axis are relative to the peat surface.

values ranged from 96% at the surface to 91% at 0.6-m depth (black bars in Figure 4, where the length of lines represent the thickness of the sample taken for the porosity measurement). For Site 1, MEGA shows similar values in 5-cm depth slices, with gas contents ranging from 10.5% to 27.2% gas content by volume (Figure 2a).

4. Discussion

In this study, MEGA replicated not only the range and variability of gas storage observed in the lab sample but also ebullition patterns and gas storage measured in the field. Gas bubble production rates estimated from field sites were input to MEGA and held constant, and physical and structural properties measured in the peat (i.e., porosity and peat depth) were used to build the model domain. Most importantly, other dynamic environmental factors traditionally noted as triggers for ebullition (such as changes in atmospheric pressure, water table, or temperature) were not included in the model. MEGA's ability to replicate erratic ebullition patterns without considering environmental changes suggests that the physical peat structure alone has a randomization effect on the timing of gas ebullition events from peat soils, which may be responsible for *noisy* data, and mask correlations of the timing of ebullition events with environmental patterns (Chen & Slater, 2015; Comas & Wright, 2012, 2014; Ramirez et al., 2015a). This study therefore highlights the importance of considering peat structure when investigating environmental triggers on the timing of gas ebullition events. The MEGA simulations recorded ebullition patterns most statistically similar to those observed in the field from those portions of the model domain where peat shelves are more clustered at depth (i.e., gas trap locations T3 and T5 in Sites 1 and 2, respectively; Figure 2). The areas of higher shelf densities allowed the accumulation of larger gas clusters, which in turn produced larger ebullition events. We consider these areas of increased shelf density to be analogous to clusters of peat fibers or poorly connected pore spaces that would impede upward migration of gas in the natural system. Areas of increased density could be the result of deformation of pore structures at bubble formation or part of the initial pore geometry. Nevertheless, the buildup of gas into larger clusters under areas of higher shelf density resulted in larger, more episodic upward avalanches of gas bubbles, which we consider analogous to ebullition events in the field.

Although MEGA did replicate the general ebullition patterns, the model did not reproduce the frequency of large ebullition events observed in the field. This discrepancy may be attributed to any number of variables present in the natural system that were not accounted for in the model, such as atmospheric pressure, changes in water level, or elastic deformation of the peat matrix.

MEGA also replicated two-dimensional gas storage down the length of the peat column, showing values similar to gas contents measured using GPR in the lab sample (Figures 4 and 2a). While recent studies have shown the heterogeneous lateral distribution of gases in subtropical peat samples from the Everglades at small (submeter) laboratory scales (Mustasaar & Comas, 2017), this study is to our knowledge the first attempt to measure and model variability of gas distribution with depth in subtropical peat. Areas of increased gas content 0.15- to 0.20-m thick were observed in both the lab sample (depth of 0–0.15, Figure 4) and in model domain (depths of 0.4–0.6 m in Figure 2a and 0.5–0.7 m in Figure 2b). Additionally, MEGA showed values in gas storage along the peat column (ranging from 10.5% to 27.2% gas content by volume) similar to those measured in the lab sample (ranging from 8% to 21% gas content by volume). The observed values in the lab sample show elevated gas content levels near the peat surface which coincide with higher porosity, which is a pattern seen in other studies (Comas et al., 2005; Slater et al., 2007). Although the highest values for gas storage in MEGA do not seem to concentrate near the surface as seen in the lab sample data, this distribution pattern is not expected since MEGA shelf spacing was random throughout the full length of the peat column, with no variations in model parameters being dependent on depth.

5. Conclusion

This study was the first to directly compare MEGA against a natural system. Here we tested MEGA against several months of field observations in a subtropical peatland (the Florida Everglades). The model replicated ebullition patterns observed in the field while considering only physical and structural soil properties measured at the field sites (i.e., porosity, peat depth, and decomposition), as well as the range and variability of gas contents observed in a lab sample. Two-dimensional gas content distribution in the model also showed similar vertical patterns seen in a peat column lab sample. Since MEGA was able to replicate these natural patterns by only considering physical properties of peat, findings suggest that peat structure with areas of poorly connected pore spaces may be a primary controller of gas ebullition patterns, with environmental factors playing a secondary role. Based on this study, we attribute large sporadic ebullition events to the accumulation of gas in large pockets in areas with closed pore spaces within the peat matrix.

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