



## The fine scale variability of dissolved methane in surface peat cores

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### ABSTRACT

Peat forming wetlands are globally important sources of the greenhouse gas CH<sub>4</sub>. The variability of flux recordings from peatlands is however considerable and the distribution of CH<sub>4</sub> below the water table poorly described. Surface peat (0–500 mm below the water table) is responsible for the bulk of emissions and a localised region of intense CH<sub>4</sub> concentration may exist within this region but the structure of peat and presence of gas bubbles make the determination of *in situ* gas distributions problematic. We report on the *in situ* distribution and concentrations of CH<sub>4</sub>, CO<sub>2</sub> and O<sub>2</sub> in surface bog peat cores using Quadrupole Mass Spectrometry and relate this to peat physical structure. Replicate cores collected in spring and autumn from both hollows and hummocks are used ( $n = 10$ ). CH<sub>4</sub> recorded in almost every profile was localised in intense peaks reaching concentrations up to 350 μM at depths where O<sub>2</sub> was absent. Each CH<sub>4</sub> peak had a coincident CO<sub>2</sub> peak with a minimum mean ratio of ~20:1 (CO<sub>2</sub>:CH<sub>4</sub>) and we found more CH<sub>4</sub> beneath hollows than hummocks. In statistical comparisons CH<sub>4</sub> concentration and distribution differed significantly between profiles for each depth. We demonstrate that variability found within a single core is at least as great as that between cores collected across the bog. The distribution of CH<sub>4</sub> was negatively correlated with bulk density and in some cases the location of roots matched those of intense CH<sub>4</sub> concentration where bubbles had formed and been trapped. Our comparisons suggest variability in gas distribution is caused by a heterogenous peat structure that controls the movement of gas bubbles and contains localised hotspots of gas production. The small and fine root systems of vascular plants on the peatland surface may cause high levels of methanogenic activity in their vicinity and also represent a physical barrier capable of trapping CH<sub>4</sub> bubbles.

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### 1. Introduction

Organic matter has accumulated as peat in bogs since the last glacial retreat and its anaerobic decay by microbial communities produces methane (CH<sub>4</sub>). CH<sub>4</sub> released from northern Boreal peat currently represents a significant proportion of the global CH<sub>4</sub> budget but their contribution under future climates remains uncertain. Post-Holocene increases in atmospheric CH<sub>4</sub> concentrations as well as pre-industrial reductions in CO<sub>2</sub> concentrations have been attributed to an explosive peatland expansion 12–8 thousand years ago (MacDonald et al., 2006). Understanding carbon cycling in bogs is therefore crucial in accounting for its contribution to future climate forcing but large uncertainties limit the value of studies aiming to quantify CH<sub>4</sub> release from these natural sources (Houghton et al., 2001).

Biogenic production of CH<sub>4</sub> can only proceed under anoxic conditions and in bogs oxygen is absent from just 20 mm beneath

the water table (Benstead and Lloyd, 1996; Lloyd et al., 1998), the depth that marks the oxic/anoxic boundary. Around this zone, where the water table fluctuates, decomposition is more rapid than anywhere else in the vertical peat profile (Clymo, 1987; Belyea, 1996). CH<sub>4</sub> is produced as a result of this decomposition of organic material with the upper 500 mm of peat being responsible for 98% of CH<sub>4</sub> flux (Clymo and Pearce, 1995). It has been suggested that a single zone of peak CH<sub>4</sub> concentration exists 70–150 mm below the peatland water table (Benstead and Lloyd, 1994, 1996; Daulat and Clymo, 1998). To date however, this representation of gas distribution in peat has not been rigorously tested. The vertical resolution of reported *in situ* profiles ranges from 5 mm (Benstead and Lloyd, 1994, 1996; Daulat and Clymo, 1998; Lloyd et al., 1998) to 10 mm (Sheppard et al., 2007) and only 'typical' profiles are reported. However, typical profiles tell us little of the processes responsible for peak CH<sub>4</sub> concentrations and variability in their distribution. They may also poorly represent within-site variability. Mass balance approaches utilise the same types of 'typical' profiles to determine *in situ* rates of carbon turnover. However, potential CH<sub>4</sub> production rates recorded using peat slurries have demonstrated between-topography variability below the peatland water

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table (Bergman et al., 2000; Yavitt and Seidmann-Zager, 2006) and a highly heterogeneous distribution of bubbles has been recorded *in situ* which can lead to substantial variability in the dissolved CH<sub>4</sub> distribution (Laing et al., 2008). A comprehensive data set of the *in situ* distribution of decomposition gases in both dissolved and gaseous phases accounting for between-topography differences could increase the accuracy of carbon turnover rate calculations and go some way to understanding recorded variability in fluxes at the water/air interface.

The peat physical structure plays an important role in determining gas distribution variability but it is rarely considered. The breakdown or utilisation of the constituents of peat is responsible for carbon gas production and the decomposability of litter has been shown to vary with microtopography (Belyea and Clymo, 2001) and depth (Belyea, 1996). Pore water CH<sub>4</sub> concentrations have also been demonstrated negatively correlated with *Carex* remains in peat (Nilsson and Bohlin, 1993). The movement and formation of bubbles may also be related to peat structure (Baird et al., 2004; Strack et al., 2005): low ebullitive fluxes have been recorded from peat cores with surfaces dominated by vascular plants relative to those more dominated by *Sphagnum* mosses (Beckmann et al., 2004). Trapped bubbles can represent a prominent gas storage phase in peatlands at some sites (Fechner-Levy and Hemond, 1996; Baird et al., 2004) and it is possible that peat physical structure may influence ebullition and bubble storage. Surface bog peat (the acrotelm) is also the rooting zone for vascular plants and the rhizosphere has been shown to act as a site for CH<sub>4</sub> oxidation (Whalen, 2005), to serve as a conduit for direct gaseous CH<sub>4</sub> diffusion to the atmosphere (Shannon et al., 1996; Hornibrook et al., 1997; Strom et al., 2005; Bowes and Hornibrook, 2006) and be a delivery system for fresh organic matter (Bellisario et al., 1999; Christensen et al., 2003; Strom et al., 2005) which can rapidly be turned over to CH<sub>4</sub>. So far, the location of root surfaces has not been linked to *in situ* gas measurements and this may offer important information on the cause of peak CH<sub>4</sub> distribution and bubble formation.

Here, we report on the fine scale variability in CH<sub>4</sub>, CO<sub>2</sub>, O<sub>2</sub> and Ar distribution in the upper 300 mm of peat from an oligotrophic bog using high-resolution Quadrupole Mass Spectrometry (QMS). Dissolved concentrations and distributions were studied in replicate peat cores from spring and autumn and from hollows and hummocks with the aim of quantifying vertical and horizontal variability. We also use a recently published technique (Laing et al., 2008) to identify the location of bubbles within these cores. We relate this to level of humification, bulk density and the location of roots and obstructions on a 10 mm vertical scale to study the effect peat structure may have on gas transport (such as the movement of bubbles) and determining the location of peak CH<sub>4</sub> concentrations by the release of root exudates. We also rigorously explore the possibility that the majority of dissolved CH<sub>4</sub> in surface peat can be found concentrated in small 'hotspots' and discuss the variability and causes of the recorded distributions.

## 2. Methods

### 2.1. Peat collection and maintenance

Peat was collected from the Silver Flowe Bog, New Galloway, Scotland (latitude 55°07' N, longitude 04°24' W). This Ramsar site is the least disturbed example of acid peatland in South Scotland. It is an oligotrophic site receiving around 1600 mm rain a year with a pH between 3 and 4 (JNCC, 1981). The surface of the Silver Flowe Bog has a prominent vascular component with sedge species such as *Eriophorum vaginatum* and *Carex* spp. growing

within a *Sphagnum* matrix with a pool, hollow and hummock complex. Six cores, 300 mm diameter by 400 mm deep were cut with the surface vegetation intact, as described by Daulat and Clymo (1998) in May 2006 and a further four in November 2007. All were analysed within six weeks of collection. In each season, half of the cores were from *Sphagnum* hollows and half from hummocks. These two topographical features of Northern peat bogs differ in the amount of surface vegetation above the water table and species composition. They also differed in seasonally dominant vegetation. Hummock cores were dominated by *Sphagnum capillifolium* with *Calluna vulgaris* (less than 8 cm tall), and other growing vascular species including *Carex* spp., and *Drosera* spp. Hollows were dominated by *Sphagnum papillosum* carpets, with growing *E. vaginatum*, *Menyanthes trifoliata* and *Narthecium ossifragum* in spring.

Once collected, cores were kept in open buckets. The cutting process retained most of the peat water in the core but each was topped up to its original water level with bog water prior to leaving the site. The buckets were transported to the roof of the Life Sciences building at Oxford Brookes University, where they were rain fed. Supplementary maintenance of the water table (~50–100 mm below the hummock surfaces and at, or just below, the surface of hollows) at its original level was performed when necessary using deionised water. Deionised water was also used to maintain these levels throughout all experimental procedures in the laboratory. Cores were brought into the laboratory from the roof 48 h prior to sampling to equilibrate to 15 °C (±0.25 °C) in a large, custom made water bath which maintained the core temperature. The central core temperature was checked for stability with an Omni Instruments (USA) XR440 data logger throughout the experiments.

### 2.2. Measurement of gases

Quadrupole Mass Spectrometry (QMS; VG Micromass, UK) with a silicone membrane inlet was used to record CH<sub>4</sub>, CO<sub>2</sub>, O<sub>2</sub>, and Ar vertical distributions within individual profiles. The *m/z* ratios used were 15, 44, 32 and 40 respectively. The water table marked the starting point for each profile. The positions of eight profiles within a core were determined by using a random number table to select points from a 44-point, 30 mm × 30 mm, grid laid over the core surface. Five hollow cores (3 spring, 2 autumn) and five hummock cores (3 spring, 2 autumn) were analysed. The QMS inlet consisted of a stainless steel capillary tube 200 cm long, 7 mm in diameter with a 5 mm internal diameter covered by a 100 μm thick silicone membrane. The inlet was positioned and moved down through the peat by slowly raising the core and its surrounding water bath upwards using an automated stepping hoist (see Daulat and Clymo, 1998). The QMS was custom built with an ion source under vacuum at 10<sup>-3</sup> mbar giving a response time of less than 2 s. Measurements were taken every 15 s in 0.6 mm vertical steps. A 300 mm profile comprises 534 measurements, recorded over a 2 h period. Wet calibration was carried out at the start of each QMS run as described in Laing et al. (2008). In the measurement of a few profiles the silicone membrane was torn by physical obstructions encountered during profiling. Only data from successful profiles are used in the analysis. After completion, the position of each profile was marked with a wooden skewer.

### 2.3. Distinction between dissolved and gaseous phases

We have demonstrated previously that bubbles are recorded *in situ* in peat cores and that measuring this phase shift (dissolved to gaseous) with a wet-calibrated QMS inlet can result in an over-estimation of concentrations (Laing et al., 2008). Regarding the

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