論文の内容の要旨

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論文題目

Laboratory and *in-situ* Investigations of Methane Bubble Occurrence and Ebullition in Peatland
（湿原におけるメタンバブルの存在と噴出に関する室内実験および現地観測研究）

Chapter 1 Introduction

Natural wetlands, with more than half of their geographical area covered with peat-rich ecosystems, are likely to be the single largest source of atmospheric methane, a potent greenhouse gas. Northern peatlands probably contribute about one-third of the world's total wetland emissions. However, there is a considerable uncertainty in current methane emissions estimates from peatalnds. Accurate observation of methane exchange between the peat and the atmosphere is a fundamental basis for any efforts to improve our knowledge of the methane cycles in peatlands. At present, most of the flux studies are based on infrequent, temporally discontinuous ground-based measurements, assuming that the flux was stationary during the substantial non-sampling period. However, this assumption has not been explicitly verified, presumably because diffusion of dissolved methane or releases of methane through aquatic plants, which may have small temporal variability, are believed to be the main transport mechanisms to the atmosphere.

In contrast to conventional wisdom that wetland soils are saturated below the water table and methane exists in a dissolved state, presence of biogenic gas bubbles originating from anoxic methane fermentation has recently been suggested. If the occurrence of methane-containing bubbles in peat is found to be pronounced, it is plausible that a considerable portion of methane emitted from peatland might be via release of bubbles, i.e. ebullition. As ebullition seems highly variable in space and time, widely-used flux measurement scheme may not be able to capture the ebullition events. Hence, there seems an urgent need to improve our understandings regarding the volume, composition, and distributions of the bubbles as well as the factors that may cause possible ebullition.

The objectives of this study are to investigate occurrence of methane bubble in peat and its possible release to the atmosphere. This research was conducted at Bibai wetland (an ombrotrophic bog), located at Hokkaido, northern Japan (141°48'E, 43°19'N).

Chapter 2 *In situ* accumulation of methane bubbles in a natural wetland soil

Compared with numerous papers on measurements of methane emission from wetland surfaces, there are few reports on methane configuration and distribution within the soil profiles. By using a newly designed gas sampler, we succeeded in collecting free-phase gas from beneath the water table. The volumetric percentage of methane in the gas phase increased with depth and was generally more than 50% beneath the zone within which the water table fluctuates (Fig. 1). The volume of the gas phase beneath the water table was estimated to be from 0-19%. Using the volume ratio of the gas and liquid phases and methane concentration in the gas phase, as well as assuming that methane was in equilibrium between the two phases, we calculated that ~60% of the methane down to 1 m accumulates in the form of bubbles. These results suggest the importance of ebullition in methane emission. Most importantly, our results show the need to consider gaseous-phase methane for understanding the production, transport and emission mechanisms of methane in peatlands, which has largely been overlooked to date.

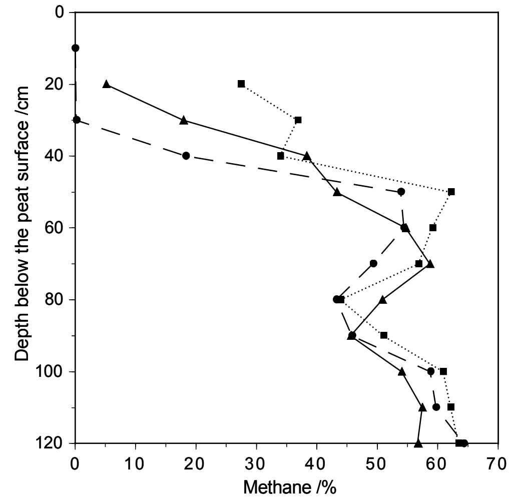

Fig. 1 Vertical profile of methane concentration at *Sphagnum*-dominated site. Methane is expressed as a percentage of gas sampled from the peat beneath the water table. ■, 23 August 2002; ●, 18 October 2002; ▲, 26 October 2003.

Chapter 3 Ebullition of methane from peat with falling atmospheric pressure

Among various potential parameters, which may trigger methane ebullition, barometric pressure is possibly one of the most important factors because a drop in atmospheric pressure may lead to gas generation from solution and the enlargement of the volume of the gas phase. We have investigated the quantitative relationship between the amount of methane emitted via ebullition and changes in the atmospheric pressure through a laboratory experiment. During flux measurement periods, ebullitions were recorded almost exclusively in air-pressure-declining phases (Fig. 2). The increased volume of the gas bubbles due to reduction in atmospheric pressure and the amount of released gas bubbles revealed a strong linear relation, suggesting that *in situ* methane emissions via ebullition can be estimated using this correlation. Our results clearly showed that atmospheric pressure can be one of the most important factors to control methane emissions from peatlands and that ebullition can be the main transport mechanism during the pressure-falling phase.

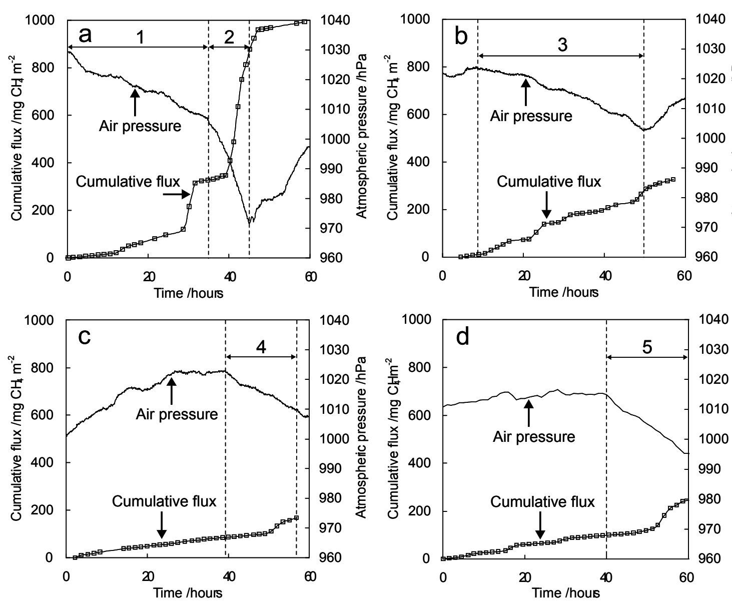

Fig. 2 Cumulative methane flux and change in atmospheric pressure starting at days 366 (a), 378 (b), 399(c), and 408(d) of the experiment. The timing of the flux measurement by a closed chamber is indicated by open squares.

Chapter 4 Falling atmospheric pressure as a trigger for methane ebullition from peatland

90-hour of field study was carried out in high-summer season to determine whether or not methane ebullition into the atmosphere from peat soil occurs in field condition, and if does, to identify factors that control it. We measured the air pressure, water table, and peat temperature as potential factors to control the ebullition. We found that the methane flux can change by two orders of magnitude within a matter of tens of minutes due to the release of free-phase methane and the contribution of the ebullition to the total methane flux during the measurements was significant (50-64%) (Fig. 3). Episodic ebullitions were always associated to the reductions in air pressure, indicating that increased buoyancy forces due to enlargement of the trapped gas caused the upward migration of the bubbles (Fig. 4). These results clearly revealed that field campaigns must be designed to cover this rapid temporal variability caused by ebullition, which may be especially important in intemperate weather. Process-based methane emission models should also be modified to include air pressure as a key factor for the control of ebullient methane release from peatland.

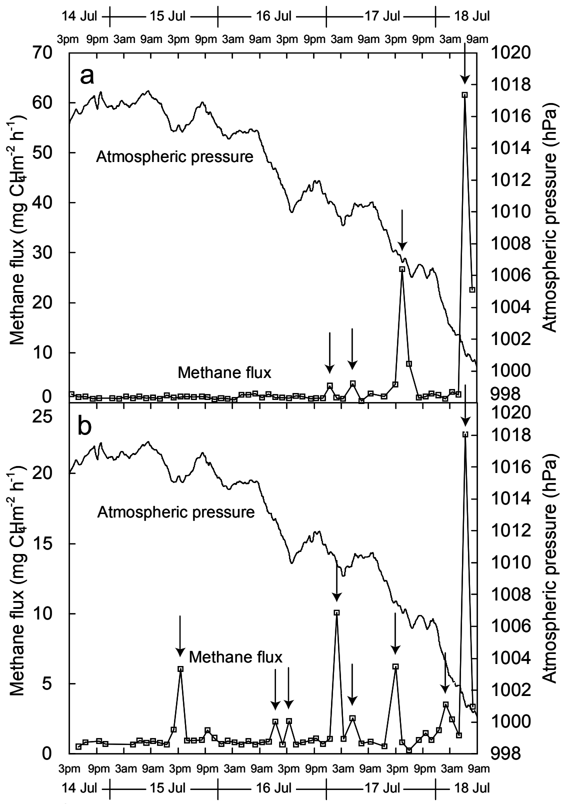

Fig. 3 Time series of methane flux and atmospheric pressure at two plots A (a) and B (b) (31.2 m apart). Both plots were located in *Sphagnum*-dominated site. The timing of the flux measurement is indicated by open squares. The vertical arrows indicate episodic fluxes, which are significantly greater ($P < 0.05$, by one-tailed t-test) than the other emission rates. The difference in the scale of methane flux between a and b is noteworthy.

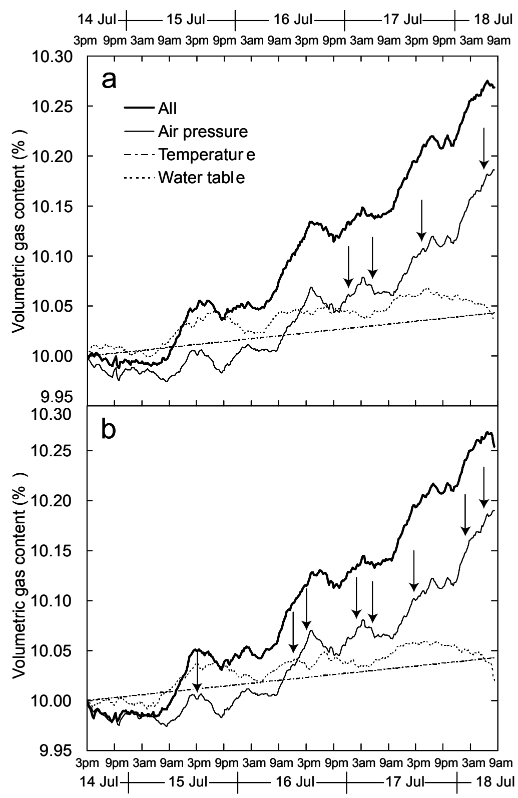

Fig. 4 Effect of changing atmospheric pressure, peat temperature, water table level, and all the variables on the volumetric gas content from the water table level to a depth of 80 cm at plot A (a) and plot B (b). The timing of episodic emissions is indicated by vertical arrows.

Chapter 5 Episodic release of methane bubbles from peatland during spring thaw

In most northern peatlands, it is clear that drastic changes in physical environments such as melting of snow and near-surface frozen peat take place during the spring thaw in relatively short periods of time, invoking a sudden change in methane emission rates. We have conducted

165 hours of intensive flux measurements and found a large methane flush at the very moment the surface ice cover thawed (Fig. 5). Bubbles were found to be trapped in the ice layer (Fig. 6) and very high concentration of methane (~20%) was detected in them. The abundance of the bubble-form methane was likely to be sufficient to explain the observed episodic release during the thaw. Omission of the episodic release of stored methane during the spring thaw results in underestimation of annual methane emissions as well as misunderstanding of seasonal methane dynamics. The results also imply the gas-phase methane may play an important role also in cold season methane dynamics in northern peatlands.

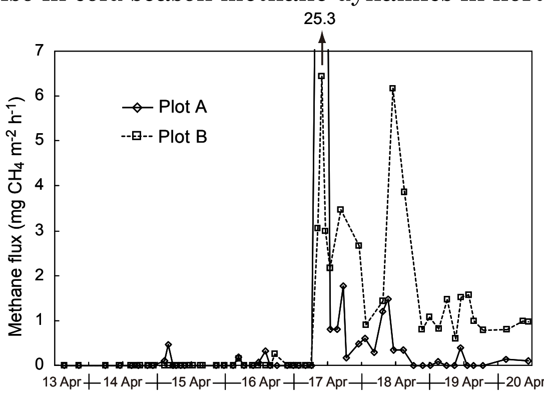

Fig. 5 Methane flux from either snow cover, standing water or peat surface from 13 April to 20 April, 2006 at plots A and B. Symbols indicate the timing of the chamber measurements.

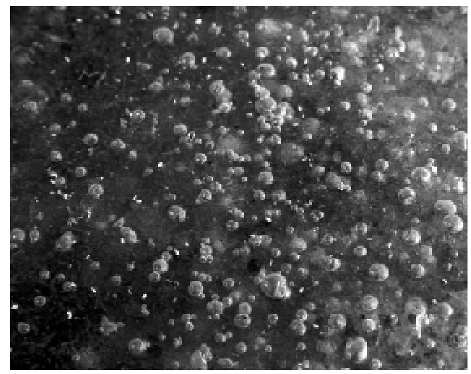

Fig. 6 A picture of bubbles stored in the ice between the snow and the peat layers. The diameters of the bubbles ranged from several mm to 1 cm.

Conclusions

The main conclusions of the study are:
1. *In situ* volumetric gas profiles and methane concentrations in the gas phase beneath the water table level were quantified for the first time, showing the occurrence of methane bubbles in waterlogged peat soil.
2. Approximately 60-70% of methane stored in water-logged peat from the surface to a depth of 1 m was found to exist as gas-phase gas (30-40% were in dissolved state).
3. The very frequent sampling regime adopted in the field study provided a clear evidence that ebullition represents an important mechanism of methane emission from peatland and that it occurs as episodic events.
4. Theoretical calculations followed by numerical computations confirmed our hypothesis that fluctuations in the atmospheric pressure play a dominant role in determining the timing and magnitude of the ebullition events.
5. Field campaigns must be designed to cover the rapid temporal variability caused by ebullition, which may be especially important under intemperate weather.
6. Because existing methane flux data may not capture ebullition events, widely-accepted process-based models might have been tested against erroneous data. Also, our findings may reveal the need to revise the model itself, i.e., air pressure should be included as a key factor for the control of methane release via ebullition.
7. Large methane emissions associated with melting of the surface ice layer occurred as a result of release of entrapped bubbles found in the ice layer, suggesting that the gas-phase methane is play an important role not only during the growing season but also in the cold season methane dynamics in northern peatlands.