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Methane uptake in a temperate forest soil using continuous closed-chamber measurements

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ABSTRACT

Methane (CH_4) fluxes were continuously measured in a temperate forest soil using six dynamic closed chambers with a laser-based analyzer. CH_4 uptake increased with an increase in soil temperature and a decrease in volumetric water content, where multiple linear regressions using the two variables explained approximately 80% of the seasonal variations. Based on the measurements, our forest acted as an annual CH_4 sink with a spatial variation of 25%. The calculated annual CH_4 sink significantly decreased by increasing the chamber deployment period: 8–13 mg CH_4 m^{-2} yr^{-1} min^{-1}. The annual CH_4 sink was significantly different using different calculation methods for the initial slope: 898 ± 11 mg CH_4 m^{-2} yr^{-1} using a linear regression, 924 ± 8 mg CH_4 m^{-2} yr^{-1} using a quadratic regression, and 975 ± 10 mg CH_4 m^{-2} yr^{-1} using an exponential model. These results indicate that underestimations due to disturbing the CH_4 gradient between chamber headspaces and soils were significant despite of the short deployment periods of 4–10 min. Chamber-based fluxes were consistent with canopy-scale CH_4 fluxes by the micrometeorological hyperbolic relaxed eddy accumulation method: sink of 731 ± 38 to 888 ± 87 mg CH_4 m^{-2} yr^{-1}. Continuous dynamic closed chambers with laser-based analyzers are a powerful tool to understand CH_4 fluxes in temperate forest soils, as long as a range of uncertainties are carefully evaluated.

1. Introduction

Upland forest soil has been identified as a major sink of methane (CH_4) due to oxidation by methanotrophs under unsaturated soil conditions (Conrad, 1996; Reeburgh, 2003; Kirschke et al., 2013). The estimated global CH_4 sink by unsaturated soils ranges from 9 to 42 Tg CH_4 yr^{-1} and is approximately 10–20% of the CH_4 emissions from natural wetlands (Kirschke et al., 2013). This CH_4 sink has increased in the recent decades (Kirschke et al., 2013), most likely due to increased atmospheric CH_4 concentrations (Hashimoto et al., 2011; Ito and Inatomi, 2012).

Chamber methods are one of the promising techniques for measuring upland CH_4 fluxes (e.g., Morishita et al., 2004, 2007; Smith et al., 2000). Previously, CH_4 fluxes in upland forests were quantified by periodic measurements using static closed chambers (Fang et al., 2010; Itoh et al., 2009; Morishita et al., 2004, 2007; Smith et al., 2000). This method successfully showed spatial and temporal variations in CH_4 fluxes; at the same time, several problems were noted (Davidson et al., 2002; Norman et al., 1997; Rochette and Hutchinson, 2005; Pumpanen et al., 2004; Pihlatie et al., 2013). The long chamber deployment periods associated with the use of these static closed chambers (enclosed from the start to the end of chamber measurements) induced underestimates in fluxes (Pumpanen et al., 2004; Pihlatie et al., 2013). Different calculation methods caused over- and underestimations of fluxes, from −35 to 6% (Pumpanen et al., 2004). Effective chamber volumes changed under porous soils (Jassal et al., 2012).

Recent applicability of laser-based analyzers overcomes the difficulty in continuously measuring small magnitudes of upland CH_4 fluxes using dynamic chamber techniques (Kiese et al., 2003; Savage et al., 2014; Werner et al., 2006, 2007). The high precision of laser-based analyzers allows for shorter chamber deployment periods and smaller gas concentration differences between the chamber headspace and the outside of the chamber than the common static chamber methods, resulting in smaller artifacts due

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to chamber effects (e.g., Pihlatie et al., 2013). Continuous high-frequency measurements taken by automated dynamic chambers could provide a detailed temporal picture of CH_4 exchange without missing "hot moments" of fluxes (Savage et al., 2014). A growing number of laser-based continuous chamber measurements have aided in the understanding of upland CH_4 exchange (Kiese et al., 2003; Savage et al., 2014; Werner et al., 2006, 2007), but the measurements taken in temperate forest soils have been limited.

Micrometeorological techniques have become available for measuring small CH_4 fluxes in upland forests at the canopy scale (Sakabe et al., 2012; Ueyama et al., 2013, 2014 Wang et al., 2013). These methods can quantify spatially representative fluxes, which was often difficult to accomplish with chamber methods due to the large spatial variability in CH_4 fluxes (Itoh et al., 2009). At the same time, there is uncertainty associated with micrometeorological CH_4 flux due to an analyzer's precision (Sakabe et al., 2012; Ueyama et al., 2013; Wang et al., 2013), resulting in small statistical powers in relation to environmental variables. Simultaneous studies using both micrometeorological and chamber methods could be a powerful tool to evaluate CH_4 fluxes in upland forests (Ueyama et al., 2014; Wang et al., 2013); but comparisons for each method have rarely been conducted.

This study aims to evaluate CH_4 flux in temperate forest soils and their environmental regulations, and present the uncertainties associated with the chamber method. We continuously measured CH_4 flux in temperate forest soils with six dynamic closed chambers with a laser-based analyzer; thereby temporal and spatial variations in soil CH_4 fluxes and their environmental regulations were discussed in detail. The uncertainties associated with the chamber method, including the initial slope estimation, sampling intervals, and spatial representativeness, were examined for individual and annual fluxes. Finally, we compared CH_4 fluxes by the chamber and micrometeorological methods.

2. Field measurements

2.1. Study site

The study site was located in a larch forest at the northern foothills of Mt. Fuji in Yamanashi, Japan (35°26′N, 138°45′E). The site is dominated by uniformly planted Japanese larch (*Larix kaempferi*) with interspersed evergreen (*Pinus densiflora*) and broadleaf species (*Swida controversa*, *Quercus serrata*, and *Quercus crispula*). The forest floor is covered with understory plants that include fern species (*Dryopteris crassirhizoma*), bamboo grasses (*Sasamorpha borealis* and *Oplismenus undulatifolius*), and shrubs and herbs (*Akebia quinata* and *Antenoron filiforme*) (Arase, 2012). The mean canopy height is approximately 25 m. The soil is classified as coarse volcanic ash with $0.43 \pm 0.11\ g\ cm^{-3}$ of soil bulk density (Urakawa et al., 2015). Further details about the study site are provided in our previous publications (Ueyama et al., 2012a, 2013; 2014; Mochizuki et al., 2014; Takahashi et al., 2015).

2.2. Dynamic closed chamber measurements

Six automated dynamic closed (non-steady-state through-flow) chambers (Fig. A1) were installed on the forest floor within the footprint of the micrometeorological measurement in October of 2012. The chamber bases were installed at a depth of 10 cm below the ground in July of 2012. We integrated the chamber system into a laser-based greenhouse gas analyzer (GGA-24r-EP, Los Gatos Research Inc., USA), which measured CH_4, CO_2, and water vapor concentration simultaneously. Because we used this analyzer for the micrometeorological flux measurements using the hyperbolic relaxed eddy accumulation (HREA) method from August 2011 to September 2012, the details of the precision were already known (Allan standard deviation was 0.5 ppb for 60 s integration time; Ueyama et al., 2013). The median rates of the CH_4 concentration changes in each chamber were 8.7–14.1 ppb min^{-1}, which are well above the detection limit of the analyzer. Based on the root mean square error (RMSE) of linear regressions within the measurements, minimum detection limit of CH_4 fluxes was approximately 0.12 nmol m^{-2} s^{-1} for 4 min chamber deployment period, and 0.03 nmol m^{-2} s^{-1} for 10 min chamber deployment period. The chambers were installed within 10-m distance from the analyzer to stabilize the flow rate within the closed system.

The chambers were made from polyvinyl chloride ($19 \times 19 \times 25$ cm, Length × Width × Height). A 10 cm-long vent extending to the soil surface was attached with the chamber to reduce pressure differences between the inside and outside of the chamber. Each chamber was automatically closed for 10 min every hour by flowing compressed air generated by a compressor (0.2LE-8s, Hitachi, Japan) into pneumatic cylinders (CJ2D16-125, SMC, Japan). When a chamber was closed, air within the chamber was circulated at a rate of 1 L min^{-1}. To achieve the flow rate, we constructed parallel plumbing with a diaphragm pump (GS-2EA, Enomoto Micro Pump, Japan) and a regulator (SS-6P4T-MM, Swagelok, USA) in addition to the inner diaphragm pump of the gas analyzer. To prevent the stagnation of air within the plumbing which was not used for measurements, air was ventilated using a diaphragm pump (GS-6EA, Enomoto Micro Pump, Japan). Liquid water within the compressor was removed using a drain trap (UPII-1B, Fukuhara Co. Ltd., Japan).

The system prevents dust and liquid water contamination and conducts dehydration. To prevent dust contamination, three filters were installed; membrane filters (12 μm; Millipore, USA) were glued into each chamber, a membrane filter (0.45 μm; Millipore, USA) was installed just before the gas dryer, and an inline filter (0.5 μm; SS-6P4T-MM, Swagelok, USA) was installed just before the gas analyzer. To prevent liquid water infiltration, a particle filter (SS-FCC, Swagelok, USA) was also included. Before entering the analyzer, the air sample was dehydrated using a Nafion gas dryer (MD-110-48F, Perma Pure Inc., USA). The carrier gas of the Nafion gas dryer was generated by a Peltier-cooled condenser (EFG5-10, IAC Co., Ltd., Japan).

To evaluate any change in the effective chamber volume, a standard CO_2 concentration gas of 19900 ppm was flowed at a rate of 4.3 mL min^{-1} using a mass flow controller (MQV9020, Azbil, Japan) once a day (02:00–03:00). The flow lines for evaluating the effective chamber volume were added in March 2013 and worked properly, except for July 3–October 12 of 2013, when the mass flow controller was set incorrectly.

The analyzer was automatically calibrated with two different standard gas mixtures; the first contained 1690.7 ppb CH_4 and 331.6 ppm CO_2, while the second contained 1858.4 ppb CH_4 and 381.5 ppm CO_2. The calibration was conducted every day from 23:21 to 23:29 JST (Japanese Standard Time). The first standard gas was emptied on October 11, 2013, and then a single point calibration using the second standard was conducted April 10, 2014. We found that the sensitivity drift of this analyzer was less than a few percentiles, and it was considered to be negligible (Ueyama et al., 2013).

The soil temperature and volumetric water content were continuously measured for each chamber. Soil temperatures at 5 cm below the ground were measured by a calibrated temperature sensor (TMP36GT9Z, Analog Device, USA). Air temperature within the chambers was also measured using the same temperature sensors. The sensors were calibrated using a platinum temperature sensor (109 L, Campbell Scientific Inc., USA) for temperature ranges from 0 °C to 50 °C before being installed in the field. The soil volumetric water content at the soil layer between 0 and 5 cm below the ground