Effect of summer throughfall exclusion, summer drought, and winter snow cover on methane fluxes in a temperate forest soil

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Abstract

Soil moisture strongly controls the uptake of atmospheric methane by limiting the diffusion of methane into the soil, resulting in a negative correlation between soil moisture and methane uptake rates under most non-drought conditions. However, little is known about the effect of water stress on methane uptake in temperate forests during severe droughts. We simulated extreme summer droughts by exclusion of 168 mm (2001) and 344 mm (2002) throughfall using three translucent roofs in a mixed deciduous forest at the Harvard Forest, Massachusetts, USA. The treatment significantly increased CH_4 uptake during the first weeks of throughfall exclusion in 2001 and during most of the 2002 treatment period. Low summertime CH_4 uptake rates were found only briefly in both control and exclusion plots during a natural late summer drought, when water contents below 0.15 g cm^{-3} may have caused water stress of methanotrophs in the A horizon. Because these soils are well drained, the exclusion treatment had little effect on A horizon water content between wetting events, and the effect of water stress was smaller and more brief than was the overall treatment effect on methane diffusion. Methane consumption rates were highest in the A horizon and showed a parabolic relationship between gravimetric water content and CH_4 consumption, with maximum rate at 0.23 g H_2O g^{-1} soil. On average, about 74% of atmospheric CH_4 was consumed in the top 4–5 cm of the mineral soil. By contrast, little or no CH_4 consumption occurred in the O horizon. Snow cover significantly reduced the uptake rate from December to March. Removal of snow enhanced CH_4 uptake by about 700–1000%, resulting in uptake rates similar to those measured during the growing season. Soil temperatures had little effect on CH_4 uptake as long as the mineral soil was not frozen, indicating strong substrate limitation of methanotrophs throughout the year. Our results suggest that the extension of snow periods may affect the annual rate of CH_4 oxidation and that summer droughts may increase the soil CH_4 sink of temperate forest soils.
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Keywords: CH_4 flux; Drought; Forest; Gas diffusion; Moisture; Organic horizon; Snow cover; Water stress

1. Introduction

Methane is an important greenhouse gas. Its concentration in the atmosphere increased from about 0.75 to 1.75 ppmv during the past 200 years (IPPC, 2001). This strong increase resulted from an imbalance between increasing CH_4 sources and decreasing CH_4 sinks. The most relevant anthropogenic and natural CH_4 sources (totaling about 600 Tg yr^{-1}) are fossil fuel exploration, rice production, ruminants, biomass burning, landfills, waste management, wetlands, termites, oceans, and hydrates. Major sinks for atmospheric methane are the reaction with hydroxyl radicals in the troposphere (~ 500 Tg yr^{-1}), losses to the stratosphere (~ 40 Tg yr^{-1}) and microbial oxidation in terrestrial soils (~ 30 Tg yr^{-1}) (IPPC, 2001). Although the sink strength of soils is rather low, it is susceptible to changes that could significantly alter the global methane budget, because active methanotrophic bacteria are strongly affected by soil disturbances and changes in methane concentrations.

One reason for the sensitivity of the soil methane sink to a variety of disturbances is probably the slow growth rate of methanotrophs at atmospheric methane concentrations. These bacteria have a very high affinity for methane in order to support growth and maintenance at atmospheric or even lower methane concentrations in terrestrial soils. Because the enzymatic turnover is substrate-limited, the methane concentration in the soil gas phase has a strong influence on the uptake

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rate (Bender and Conrad, 1992). Thus, a wide range of soil disturbances can inhibit methane uptake simply by restricting the chemical or physical availability of methane. In contrast, temperature has little effect on the activity of methanotrophs (King and Adamsen, 1992).

The transport of atmospheric methane into the mineral soil is largely controlled by soil porosity and soil moisture. Methane uptake rates generally increase with decreasing soil moisture (Castro et al., 1994) because molecular diffusion in water is a factor 10^4 slower than in air. Hence, shifts in the frequency and intensity of rainfall and evapotranspiration may affect the soil methane sink. In a field experiment, prolonged summer droughts increased annual methane uptake of a spruce forest in Germany by 102 and 41% (Borken et al., 2000). Extremely low water contents, however, could potentially limit the biological activity of methanotrophs as it was reported for a desert soil (Striegl et al., 1992) and an arable soil (Dobbie and Smith, 1996). It is not known if water stress may also limit the methane uptake in temperate forest ecosystems during dry summers.

Another factor that could limit the diffusion of atmospheric methane into the soil is snow cover. Most studies have not considered CH_4 fluxes during snow-covered periods, and thus, annual fluxes may have been over- or underestimated. One of a few studies showed that alpine and subalpine soils consume considerable amounts of atmospheric methane under >1 m thick snowpacks (Sommerfeld et al., 1993) and that this snow-covered period made greater contribution to the annual flux than the snow-free period.

Our objectives in this study were (1) to evaluate the effects of throughfall exclusion and summer drought on methane uptake in a temperate forest, (2) to determine the location of maximum methane uptake in the soil profile, and (3) to describe the relationship between soil moisture and methane uptake of the most active soil horizon, and (4) to measure the effect of winter snow cover on methane uptake rates.

2. Materials and methods

2.1. Site description

The experiment was conducted in a mixed deciduous forest at the Prospect Hill tract of the Harvard Forest (42 32′N, 72 11′W) at 340 m elevation, in Petersham, Massachusetts. The present forest developed after a hurricane in 1938 and is dominated by red maple (*Acer rubrum* L.) and red oak (*Quercus rubra* L.). Black birch (*Betula lenta*), white pine (*Pinus strobus*) and hemlock (*Tsuga canadensis*) are also present. The O horizon is 3–8 cm thick, stores about 7 kg m^{-2} of organic matter, and consists of Oi, Oe and Oa horizons (Table 1). As a result of 19th century agricultural use, the upper mineral soil is partly disturbed as indicated by varying thicknesses of the A horizon between 2 and 15 cm, being 4.4 cm thick on average at our study site. The thickness of the total mineral soil varies from 40 to 65 cm and has rock contents up to 40% of soil volume. The soil, a fine sandy loam on glacial till, is well aerated and has been classified as a well-drained Typic Dystrochrept.

According to the long-term records at the Harvard Forest meteorological station, mean annual air temperature is 8.5 °C and mean annual precipitation is 1050 mm, with precipitation evenly distributed throughout the year. Average air temperatures were 9.7 °C in 2001 and 8.4 °C in 2002 with means of 16.4 and 15.7 °C during the growing seasons from May 1 to October 31. The amounts of precipitation were 865 mm in 2001 and 1110 mm in 2002 with 461 and 583 mm during the growing seasons of 2001 and 2002. The soil is intermittently covered by thick layers of snow from late December to early April. The spring snowmelt often causes field capacity of soils in March and April.

2.2. Experimental design and installations

Three translucent roofs, each 5×5 m, were constructed 1.3 m above the forest floor in April 2001 in order to simulate severe soil droughts during the growing seasons of 2001 and 2002. Throughfall water was excluded over 84 days from July 2 to September 24 in 2001 and over 127 days from May 7 to September 10 in 2002. Roof panels were removed in September to enable undisturbed leaf litterfall and snowfall to the forest floor in the exclusion plots. A control plot of same size was established 2 m from each of the three exclusion plots.

In April 2001, three quantitative pits of 1×1 m were established adjacent to the control and exclusion plots to determine some soil characteristics and soil physical parameters (Table 1). The thickness, mass, and gravel and rock contents (defined as size fractions greater than 2 mm) were measured for each organic and mineral soil horizon down to the bedrock. Soil pits were enlarged to about 1.5 m width for the installation of sensors within each control and exclusion plot. One wall of each pit was adjacent to a control plot and another wall of the same pit was adjacent to an exclusion plot.

Table 1
Some soil characteristics of the study site

Horizon	Thickness (cm)	Amount of soil (kg m^{-2})	Rock content (m^3 m^{-3})	Bulk density (<2 mm fraction) (g cm^{-3})	Water holding/field capacity (mm)
Oi	2.0	0.53	–	–	1.9
Oe	2.6	4.09	–	–	10.2
Oa	2.1	2.60	–	–	4.7
A	4.4	29.1	0.02	0.71	15
B I	16.1	99.6	0.15	0.81	43
B II	34.5	229.3	0.28	0.92	73

Six temperature and four TDR probes (CS615 Water Content Reflectometer, Campbell Scientific, Logan, Utah) were installed into each of these two pit walls to record soil temperatures and volumetric water contents. Additionally, six $1/8''$ stainless steel tubes of 1.8 m length were horizontally inserted about 50 cm into the pit walls at same depths as soil temperatures probes to measure CH_4 gradients. Mean installation depths of probes and gas tubes were 4–5, 7–8, 11–14, 26–29, 37–38, and 53–54 cm, varying from plot to plot because of differences in the thickness of soil horizons and in total depth of the soil profiles. The pits were refilled with soil and rocks after the installations.

Volumetric water contents of the A and B horizons were recorded at hourly intervals with two data loggers (CR10X, Campbell Scientific, Logan, Utah) and were converted into WFPS as follows

$$WFPS = \theta/(1 - BD/PD),$$

where BD is the bulk density (g cm^{-3}) and PD is the particle density (g cm^{-3}) of the mineral soil, which were estimated as 2.59 g cm^{-3} for the A horizon and 2.63 g cm^{-3} for the B horizon, based on the organic matter content of the horizons. For more information about the experimental design, installations and measurements see Borken et al. (in press).

2.3. CH$_4$ flux rates and CH$_4$ profile concentrations

In each of the six plots, four PVC collars, 10 cm tall and 25 cm in diameter, were installed approximately 4 cm into the soil for the duration of the experiment. CH_4 uptake was measured between 9 AM and 12 noon on a weekly basis during the growing season and biweekly or monthly during the dormant season. Chambers with a volume of 5.2 L were placed over the collars for 30 min. Gas samples were taken from the headspace 0, 10, 20 and 30 min after closure using 10 ml gas tight plastic syringes which were closed with stopcocks.

From December to March two chambers were pushed 5 cm into the snow cover of each control plot to determine the CH_4 flux. Another two chambers were placed on two collars of each control plot 30–60 min after the removal of the snow cover to allow an equilibration of CH_4 concentration in the soil atmosphere. The snow was completely removed on an area of about 40×40 cm to expose the litter layer within and outside the collars to the atmosphere.

Soil CH_4 concentration profiles were determined by taking 10 ml samples with plastic syringes from the buried gas sampling tubes in both control and exclusion plots. The first 10 ml of gas were expelled to flush the stainless steel tubes with ambient soil air. Then 10 ml gas samples were drawn and the syringes were closed with stopcocks. All gas samples were analyzed within 6 h in a laboratory at the Harvard Forest with a gas chromatograph (Shimadzu GC 14A) equipped with a FID detector. Linear regressions were performed on the four samples collected from each flux chamber to calculate the CH_4 fluxes rates, using air pressure and air temperature within the chamber headspace.

2.4. Laboratory incubations

We performed two incubation studies in August and September of 2001. In the first study, we investigated the effect of soil water content on CH_4 consumption of the A horizon. Soil from the A horizon was taken from each of three pits, mixed and sieved to remove roots and gravel (> 2 mm). Gravimetric water content was determined by drying sub-samples at 105 °C over 24 h. In total, 5×9 subsamples of 35 g soil (dry weight) were placed in gas tight glass jars with a volume of 465 ml. One set of five subsamples was maintained at field-moist conditions, and multiple subsamples ($n = 5$) were adjusted to eight different water contents by air-drying and wetting at room temperature. We incubated field-moist (0.30 g H$_2$O g^{-1} soil), air-died (0.01, 0.10, 0.15, and 0.23 g H$_2$O g^{-1} soil) and wetted soil samples (0.46, 0.63, 0.77 and 0.80 g H$_2$O g^{-1} soil) after an equilibration time of about 24 h. The CH_4 consumption of the A horizon was measured at room temperature (24 ± 1 °C) over 14 days. After closure of the jars, 5 ml gas samples were taken with syringes from the headspace. The syringes were closed with stopcocks and then the gas samples were immediately analyzed with the gas chromatograph (see above). The incubation time of each jar (12–72 h) and the number of CH_4 analysis during that period ($n = 5$–7) varied with the consumption rate of the samples. Water contents were determined by weighing the jars before each incubation.

In the second study, two soil samples were quantitatively removed by horizon from within a frame of 100 cm^2 in each control and exclusion plot 1 week before the roof panels were removed. These samples were separated into the Oi, Oe/Oa, A and B horizons to evaluate the potential of each horizon for atmospheric CH_4 consumption. After coarse roots and stones were picked out by hand, 20–40 g of field-moist samples were separately incubated using same jars as described above. The samples were sieved (2 mm) and gravimetric water contents were determined after the incubations were complete.

2.5. Data analysis

CH_4 flux rates and CH_4 concentrations of soil air, soil temperatures, volumetric water contents were calculated as the means of the three replicated control and exclusion plots. CH_4 fluxes were normally distributed with a skewness of -0.089 after removal of low winter CH_4 fluxes (snow cover) from the data set. Analysis of variance for repeated measures was performed to test significant differences of mean CH_4 fluxes between the control and exclusion plots for different periods. We assumed that CH_4 uptake rates were not different between the control and exclusion plots from December 2001 and March 2002 and used therefore the same values to calculate cumulative CH_4 fluxes. Linear regressions were performed to evaluate the relationships between the differences of the exclusion and control plots in methane uptake and water contents of the Oi, Oe/Oa, and A horizons.

3. Results

3.1. Effect of throughfall exclusion on CH_4 uptake and water-filled pore space

CH_4 uptake followed a strong seasonal trend in both the control and exclusions plots with highest fluxes in July/August and lowest fluxes from December to March when a snow cover was present (Fig. 1a). Mean CH_4 uptake rates were not different in the control and exclusion plots during the pre-treatment periods from May to July 2001 and from April to May 2002 (Table 2). The exclusion of throughfall significantly ($P < 0.05$) increased CH_4 uptake rates by 0.9 mg m^{-2} d^{-1} from July to August 2001 (first 7 weeks of the treatment) and from May to September 2002. However, throughfall exclusion had no significant effect in 2001, considering the full treatment period from July to September (Table 2). Overall, the changes in CH_4 uptake were relatively small considering that large portions of annual throughfall (168 mm, or 19% of total precipitation in 2001; and 344 mm, or 31% of total precipitation in 2002) were excluded by the roofs. On an annual scale (May 2001–2002), cumulative CH_4 uptake rates increased from 1225 to 1305 mg m^{-2} due to rainfall exclusion.
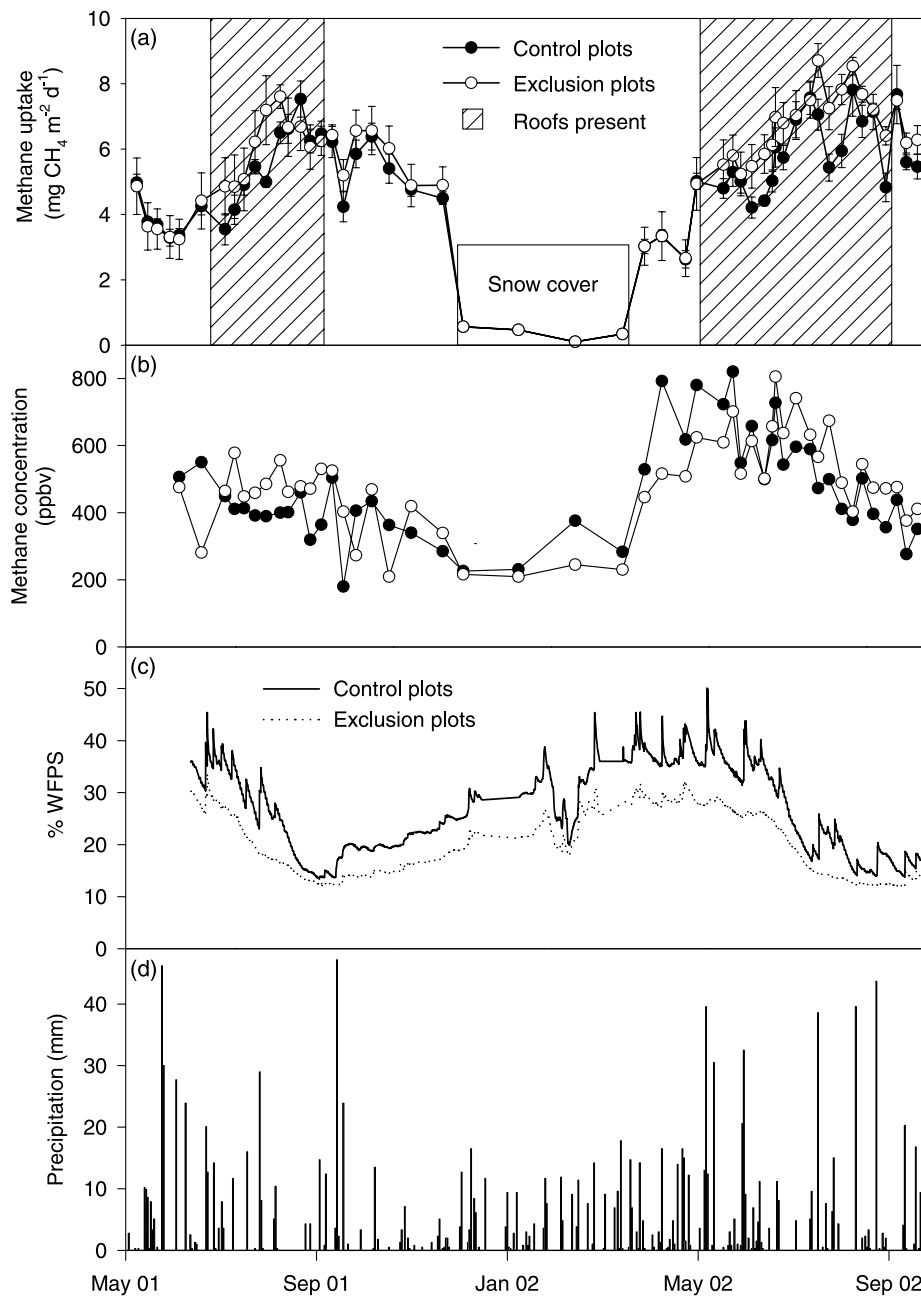

Fig. 1. (a) CH_4 uptake rates, (b) CH_4 concentration in the A horizon (4–5 cm depth), (c) WFPS in the A horizon and (d) daily precipitation in the control and exclusion plots of a mixed deciduous forest at the Harvard Forest from June 2001 to September 2002. Error bars represent the standard error of the means ($n = 3$ plots).

Table 2
Mean CH_4 uptake rates ($\pm$SE) and differences in cumulative CH_4 fluxes (exclusion plots–control plots) by treatment and season using manual chambers from 2001 to 2002

Period	No. of days	Control plots (mg CH_4 m^{-2} d^{-1})	Exclusion plots (mg CH_4 m^{-2} d^{-1})	Difference (mg CH_4 m^{-2})
Pre-treatment 17 May–1 July 2001	54	3.89 (0.19)	3.84 (0.72)	−2
Throughfall exclusion 2 July–24 Sep 2001	84	5.69 (0.33)	6.17 (0.72)	36[a]
No treatment 25 Sep 2001–7 May 2002	227	3.33 (0.18)	3.54 (0.44)	33
Throughfall exclusion 8 May–10 Sep 2002	127	5.89 (0.43)	6.82 (0.42)	99*
Annual flux May 2001–May 2002	365	4.27 (0.24)	4.55 (0.59)	80*

Significant differences are indicated by * at $\alpha = 0.05$.

[a] Throughfall exclusion had a significant effect ($P = 0.017$) on CH_4 uptake in the period from July 2 to August 21, 2001.

Although the exclusion of throughfall significantly lowered the water content of the O horizon (Borken et al., in press), the treatment effect on WFPS in the A horizon (Fig. 1c) and in the B horizon (not shown) was less clear. A difference in WFPS of the mineral soil between the control and exclusion plots existed before the installation of the roofs and may be partly explained by the large heterogeneity of the gravel and rock contents in the soil. This pre-existing difference did not change significantly following initiation of the throughfall exclusion. The lowest WFPS of about 12–13% was reached in mid-August to mid-September, when CH_4 uptake rates decreased in both the exclusion and control plots. Some strong rain events (Fig. 1d) caused short-term spikes in the WFPS, which reduced CH_4 uptake in the control plots.

In the control plots, CH_4 uptake was linearly correlated ($y = -12.3x + 8.7$, $r^2 = 0.71$) with WFPS in the A horizon (Fig. 2). A quadratic function revealed a better fit for CH_4 uptake and WFPS in the exclusion plots ($y = -169x^2 + 54x + 2.73$, $r^2 = 0.69$) than a linear function ($y = -15.8x + 9.1$, $r^2 = 0.57$). The quadratic fit indicates maximum CH_4 uptake at WFPS of 0.16 cm^3 cm^{-3} and decreasing uptakes rate at WFPS < 0.16 cm^3 cm^{-3}. However, the variation in CH_4 uptake was particularly high at low soil moisture, suggesting that methanotrophs were only occasionally limited by water stress. Similar relationships were found for CH_4 uptake and WFPS in the B horizon of the control and exclusion plots (not shown).
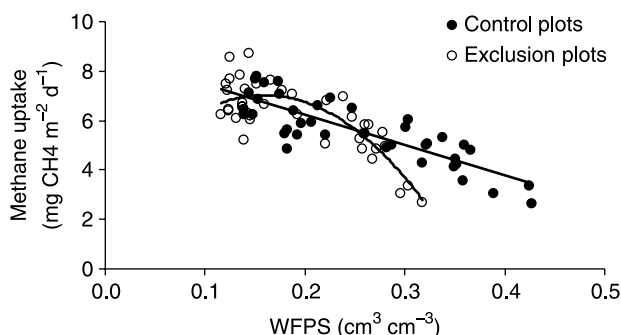
3.2. Effect of snow cover on CH_4 uptake and CH_4 concentration in soil profiles

After snowfall the uptake of atmospheric CH_4 dropped from 4.5 to 0.6 mg CH_4 m^{-2} d^{-1} in the control plots and remained below this level from December 2001 to late March 2002 (Fig. 1a). CH_4 uptake increased to 3.0 mg CH_4 m^{-2} d^{-1} immediately after snow melt in early April 2001. The removal of snow strongly increased CH_4 uptake by a factor of 7–10 in four experiments (Fig. 3). The lowest uptake rate occurred in February 2002 when the O horizon and the uppermost A horizon were frozen and the soil was partly covered by ice lenses. The water in the mineral horizon, however, was generally not frozen during the winter, indicating that the snow cover, when present, insulates the soil against frost. CH_4 concentration in the A horizon ranged between 209 and 376 ppbv during the snow period which is far below the average concentration of about 500 ppbv during the growing seasons. Assuming a winter period without snow cover as simulated by the removal of snow, annual cumulative CH_4 uptake rate would have increased from 1225 to 1485 mg m^{-2} in the period from May 2001 to May 2002.

3.3. CH_4 concentration in soil profiles

Considering the entire study period, mean CH_4 concentrations dropped in the soil profile from 1750 ppb in the

Fig. 2. Relationship between mean CH_4 uptake rates and water filled pore space for the control and exclusion plots during growing seasons of 2001 and 2002.
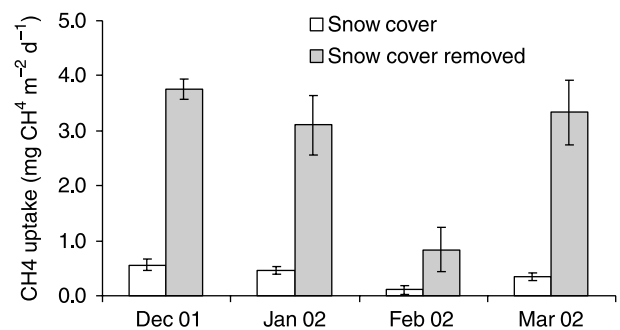

Fig. 3. Effect of snow cover and removal of snow cover on methane uptake of the control plots at four occasions during the winter of 2001/2002. Error bars represent the standard error of the means ($n = 3$ plots).
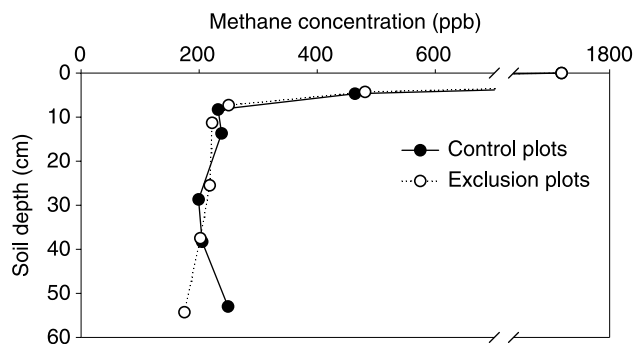
Fig. 4. Mean methane concentrations in the soil profile of the control and exclusion plots from June 2001 to September 2002.

Fig. 5. Relationship between methane consumption and gravimetric soil water content of incubated A horizon.

atmosphere at the soil surface to 460 ppb at a depth of 4–5 cm in the A horizon, indicating that about 74% of available atmospheric CH_4 was oxidized in the O horizon and top 4–5 cm of the mineral soil (Fig. 1b and Fig. 4). Rainfall exclusion had a clear effect on CH_4 concentration in the A horizon. CH_4 concentration in the exclusion plots were lower during the pre-treatment periods of 2001 and 2002, but were on average 98 ppbv (12 July–18 September 2001) and 78 ppbv (19 June–18 September 2002) higher during throughfall exclusion as compared with the control plots. This reversal suggests either a limitation of CH_4 oxidation in the top mineral soil during throughfall exclusion, or, alternatively, a faster transport of atmospheric CH_4 into the mineral soil as a result of decreasing moisture in the O horizon.

In the exclusion plots, CH_4 concentration continued to decrease to 175 ppb at a depth of 54 cm whereas CH_4 concentration moderately increased from 205 to 249 ppbv between 38 and 53 cm depth in the control plots. Production of CH_4 may explain this increase as we observed this phenomenon in all control plots. Moreover, an increase of CH_4 concentration was also observed between 38 and 53 cm depth in the exclusion plots during the pretreatment period of 2002 (not shown).

3.4. Laboratory experiments

Soil moisture had a strong effect on CH_4 consumption of sieved samples from the A horizon (Fig. 5). The maximum rates of 16.4–16.6 µg CH_4 kg^{-1} h^{-1} were determined at gravimetric water contents between 0.23 and 0.30 g H_2O g^{-1} soil. Increasing water contents reduced the consumption rate to 5.1 µg CH_4 kg^{-1} h^{-1} at 0.77 g H_2O g^{-1} soil [1]. At maximum water holding capacity (0.80 g g^{-1}) the soil still consumed 0.5 µg CH_4 kg^{-1} h^{-1}. A strong decrease in CH_4 consumption was observed when the water content was lower than 0.15 g H_2O g^{-1} soil, suggesting water stress of methanotrophs. In situ gravimetric water contents (calculated from volumetric water contents and bulk density of the A horizon) ranged between 0.14 and 0.51 g H_2O g^{-1} soil in the control plots and between 0.12 and 0.34 g H_2O g^{-1} in the exclusion plots, indicating that soil moisture fell below the optimum range (Fig. 5) only briefly during late summer drought.
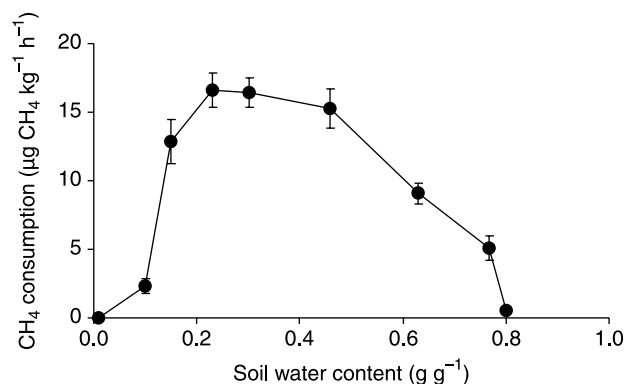
The incubated A horizon samples from both the control and exclusion plots showed the highest consumption rates of 23.3 ± 2.7 and 21.1 ± 2.4 µg CH_4 kg^{-1} h^{-1} at ambient water contents of 0.24 and 0.22 g H_2O g^{-1}, respectively. Soil samples from the B horizon consumed about two-third less CH_4 (8.2 ± 1.5 and 5.9 ± 0.9 µg CH_4 kg^{-1} h^{-1} in the control and exclusion plots) than the A horizon samples at a ambient water content of 0.17 g H_2O g^{-1}. Previous throughfall exclusion in the field had no significant effect on subsequent laboratory CH_4 consumption on this sampling date (17 September 2001).

The Oe/Oa horizon showed a small consumption rate of 0.4–0.5 µg CH_4 kg^{-1} h^{-1} in both treatments and had mean water contents of 0.84 g H_2O g^{-1} in the control plots and 0.64 g H_2O g^{-1} in the exclusion plots. We excluded two samples of the Oe/Oa horizon with higher rates (3.3. and 3.9 µg CH_4 kg^{-1} h^{-1}) in the calculation because these samples were 'contaminated' with mineral soil probably through the digging activity of chipmunks. The Oi horizon did not consume or produce CH_4 during incubation (not shown).

4. Discussion

The exclusion of throughfall of 168 mm in 2001 and 344 mm in 2002 had little effect on CH_4 uptake at our study site, although the seasonal dynamic of CH_4 uptake was strongly correlated with WFPS in the mineral soil. On an annual scale (May 2001–2002) the uptake rate increased only by 0.3 mg CH_4 m^{-2} d^{-1} or 7% as a result of the throughfall exclusion treatment.

In a temperate spruce forest in Germany, annual CH_4 uptake rates increased by 102 and 41% following 172 days and 108 days of simulated summer droughts (Borken et al., 2000). The main reason for the difference between these two field experiments is that the exclusion of throughfall at the Harvard Forest did not significantly reduce mineral soil moisture between wetting events, whereas a strong reduction in mineral soil moisture was found by the drought simulation in the spruce forest at the Solling, Germany (Borken et al., 2000). The soil at Solling, a silty loam with a 5–10 cm thick O horizon is moderately to poorly drained and is usually not as dry as the soil at the Harvard Forest during the summer. Although

the mineral soil at the Harvard Forest was wetted by rainfall events in the control plots, the soil drained very well, resulting in little or no cumulative temporal increase in mineral soil water content in the control plots relative to the exclusion plots. Seasonal changes in mineral soil water content due to natural drought were greater than differences between exclusion and control treatments. The good drainage of the Harvard Forest soil explains why annual CH_4 uptake is high (this study; Castro et al., 1995; Gulledge et al., 2004) relative to other temperate forests (Borken et al., 2003; Lessard et al., 1994; Smith et al., 2000; Yavitt et al., 1990; 1995).

The small difference in CH_4 uptake between the control and treatment plots can be attributed to temporally elevated moisture in the O and A horizon in the control plots following rain events. Throughfall exclusion had a strong effect on the water content in the O horizon (Borken et al., in press). In our laboratory experiments the O horizon showed very low CH_4 uptake, suggesting that methanotrophs are almost inactive in this horizon. The O horizon could be a diffusion barrier for atmospheric CH_4, particularly when it is wet. Several studies have shown that the removal of the O horizon may enhance CH_4 uptake of forest soils (Borken and Brumme, 1997; Dong et al., 1998). We observed at our site that chipmunks (*Tamias striatus*) spread mineral soil on the surface of the O horizon at some isolated locations by digging holes in the soil. Such disturbed organic horizons consumed considerable amounts of atmospheric CH_4 (not shown) and were not used in the laboratory study.

The A horizon is the most active horizon consuming about 74% of atmospheric CH_4 that entered the soil. In the underlying soil mean CH_4 concentrations decreased to about 175 ppbv in the exclusion plots and 200 ppbv in the control plots, indicating an efficient enzyme system with a high affinity for low CH_4 concentrations. Gulledge et al. (2004) reported an apparent half saturation constant K_m for CH_4 consumption in the 0–5 cm mineral soil depth of 42 nM CH_4, a value that is typical for high-affinity CH_4 oxidizers. Our incubation experiments suggest that the consumption of atmospheric CH_4 in these soils is limited by diffusive transport of CH_4. Based on the amount of A horizon per square meter, the potential consumption of atmospheric CH_4 based on incubations of soil cores from the A horizon was about twice as high as the CH_4 uptake rate in the field.

The incubation experiment (sieved soil) demonstrated that the consumption rate of the A horizon reached a maximum at water contents between 0.23 and 0.30 g g^{-1}. Increasing water contents diminished the diffusion of CH_4 whereas decreasing water contents may have induced water stress, which became most effective at water contents below 0.15 g g^{-1}. Gravimetric water contents of 0.12 and 0.14 g g^{-1} were reached briefly in September 2001 and 2002 in the exclusion and control plots. In agreement with our laboratory findings, CH_4 uptake was reduced at low water contents between mid-August and September in the exclusion plots and to a lesser extent in the control plots. According to the water retention curve for this soil, a gravimetric water content of 0.12 g g^{-1} is consistent with a soil matric potential of -1.5 MPa (Savage and Davidson, 2001). Hence, methanotrophs are sensitive to

water stress, as are most bacteria, but some fraction of the population was still active when the average gravimetric water content was 0.10 g g^{-1} (Fig. 5) and the average matric potential was approximately -2.3 MPa. Although moisture stress was apparently only partial and brief in this study, it has been shown to be important in a desert environment (Striegl et al., 1992).

A similar parabolic relationship between percent water-holding capacity (WHC) and CH_4 uptake was reported by Bowden et al. (1998) for other forest stands within the Harvard Forest, although there were no points between about 20 and 40% WHC to identify the optimum water content that probably fell within that range. Torn and Harte (1996) also showed a parabolic relationship using gravimetric water content for a meadow soil from Colorado, USA. The optimum range for this soil was at about 20–30% moisture by weight, whereas decreasing and increasing soil moisture diminished CH_4 consumption of incubated soil.

Compared with throughfall exclusion, snow cover had a stronger impact on annual CH_4 uptake rate. With the first snowfall of the winter 2001/2002 uptake rate was reduced from 4.5 to 0.6 mg CH_4 m^{-2} d^{-1} and remained at a low level until snow melt. Removal of snow increased CH_4 uptake by a factor of 700–1000%, indicating that snow was a strong diffusion barrier for atmospheric CH_4. The resistance of snow to gas diffusion can be highly variable as the thickness of snow cover and the quality of snow may vary spatially and temporally. CH_4 uptake rates in December, January and March were of the same magnitude after removal of snow cover as fluxes during the growing season, indicating that temperature has little apparent effect on the activity of methanotrophs. Very low CH_4 concentrations in the soil profiles support a high activity of methanotrophs at low temperatures. The fluxes were considerably lower in February when the top mineral soil was frozen and gas diffusion restricted by ice in soil pores. In agreement with our results, CH_4 uptake was present throughout the snow-covered period in alpine and subalpine regions of Wyoming, USA (Sommerfeld et al., 1993). The subalpine meadow soil consumed less CH_4 during snow-covered period as compared with the snow-free period. The results suggest that a shift in the length of the snow-covered period may affect annual CH_4 uptake in many temperate and boreal regions.

4.1. Conclusions

The exclusion of throughfall increased soil CH_4 uptake at the Harvard Forest, although the effect was rather weak. Soil water contents below 0.15 g g^{-1} appeared to cause desiccation stress that diminished CH_4 uptake in the A horizon at the Harvard Forest. However, such severe drought conditions occur only briefly in the late summer of some years. On balance, higher diffusion rates in dry soil were more important than desiccation stress, indicating that summer droughts may generally increase the soil CH_4 sink of temperate forest soils. Reduced snow cover also increased rates of CH_4 consumption. Our results demonstrate that substrate limitation, caused by processes that restrict diffusion of atmospheric CH_4 to the

mineral soil, is the most important factor limiting CH_4 consumption by these forest soils. Hence, climate change that affects hydrologic processes such as duration of snow cover and summer drought, is likely to alter rates of uptake of atmospheric CH_4 by forest soils.

Acknowledgements

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