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Mitigation of methane emissions in a pilot-scale biocover system at the AV Miljø Landfill, Denmark: 2. Methane oxidation

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ABSTRACT

Greenhouse gas mitigation at landfills by methane (CH_4) oxidation in engineered biocover systems is believed to be a cost effective technology but so far a full quantitative evaluation of the efficiency of the technology in full scale has only been carried out in a few cases. A third generation semi-passive biocover system was constructed at the AV Miljø Landfill, Denmark. The biocover was fed by landfill gas pumped out of three leachate collection wells. An innovative gas distribution system was used to overcome the often observed uneven gas distribution to the active CH_4 oxidation layer resulting in overloaded areas causing CH_4 emission hot spot areas in the biocover surface. The whole biocover CH_4 oxidation efficiency was determined by measuring the CH_4 inlet load and CH_4 surface fluxes. In addition, CH_4 oxidation was determined for single points in the biocover using two different methods; the carbon mass balance method (based on CH_4 and carbon dioxide (CO_2) concentrations in the deeper part of the cover and CH_4 and CO_2 surface flux measurements) and a new-developed tracer gas mass balance method (based on CH_4 and tracer inlet fluxes and CH_4 and tracer surface flux measurements). Overall, the CH_4 oxidation efficiency of the whole biocover varied between 81 and 100% and showed that the pilot plant biocover system installed at AV Miljø landfill was very efficient in oxidizing the landfill CH_4. The average CH_4 oxidation rate measured at nine campaigns was approximately 13 g m^{-2} d^{-1}. Extrapolating laboratory measured CH_4 oxidation rates to the field showed that the biocover system had a much larger CH_4 oxidation potential in comparison to the tested CH_4 load. The carbon mass balance approach compared reasonably well with the tracer gas mass balance approach when applied for quantification of CH_4 oxidation in single points at the biofilter giving CH_4 oxidation efficiencies in the range of 84 to a 100%. CH_4 oxidation rates where however much higher using the tracer gas balance method giving CH_4 oxidation rates between 7 and 124 g m^2 d^{-1} compared to the

carbon mass balance, which gave CH_4 oxidation rates -0.06 and 40 g m^2 d^{-1}. The study also revealed that the compost respiration contributed significantly to the measured CO_2 surface emission, and that the contribution of the compost respiration decreased significantly with time probably due to further maturation of the compost material.

1. Introduction

Landfills contain organic waste, which leads to production of biogas containing methane (CH_4) and carbon dioxide (CO_2). Landfills are important sources of atmospheric CH_4, which contributes to climate change (Bogner et al., 2008). At some landfills utilization of landfill gas (LFG) is not or cannot be carried out, and the gas is either flared with risk of producing toxic combustion products or just emitted to atmosphere. As an alternative to mitigation by gas utilization systems, mitigation systems based on CH_4 oxidation processes may be implemented. Such systems, here called biocover systems are based on microbial CH_4 oxidation in full surface biological biocovers, biowindow systems or open or closed bed biofilter systems. Experiments have documented that very high CH_4 oxidation rates can be obtained in bio-covers, high enough to significantly reduce CH_4 emissions from landfills (Scheutz et al., 2009). Documentation of the efficiency of bio-covers has so far only been carried out in full scale in a few cases for example in a research project carried out at Fakse Landfill where a new-developed protocol for biocover establishment and monitoring were presented (Scheutz et al., 2011a, b) and in a project where a second generation biocover system was established at the Klintholm Landfill (Scheutz et al., 2014, Kjeldsen et al., 2009). Both of the mentioned biocover systems were passively loaded with landfill gas from the landfilled waste.

This study was carried out at AV Miljø, which is a modern landfill situated outside Copenhagen, Denmark. The landfill receives non-combustible waste with a low organic content. CH_4 is produced but in too little quantities for energy utilization. The LFG produced at the landfill is mainly emitting through the leachate collection wells. More details of the landfill are given in Scheutz et al. (2011c). A semi-passive pilot-scale compost based biocover system designed for CH_4 oxidation was installed in September 2012 at the AV Miljø

Landfill with the aim to mitigate CH_4 emissions from the landfill. The biocover system (12 m x 42 m) consisted of a gas distribution layer (30 – 50 cm) overlain by a compost layer (80 – 90 cm). The CH_4 fed to the biocover was collected from three leachate wells. Inlet pumping rates and CH_4 contents were continuously monitored. Other gases (oxygen (O_2), carbon dioxide (CO_2) and nitrogen (N_2)) were monitored manually on a monthly basis. A pipe system consisting of a combination of un-slotted and slotted pipes was used to feed and homogenously distribute the LFG to the compost layer. The upper surface of the gravel gas distribution layer was shaped in a series of valley and ridges (in a "zig zag" shape) to minimize continuous water accumulation in the compost layer and subsequent locking due to capillary effects. The biocover system was equipped with a water draining system including a water lock to avoid gas by-pass. A section of the gas distribution layer and the compost layer was equipped with horizontal multi-port gas monitoring units to study gas distribution and CH_4 oxidation processes. Two transects each having 50 gas ports were constructed at two different distances from the gas inlet. More details of the biocover system design are given in Cassini et al. (2017).

The objective of this study was to quantify the efficiency of CH_4 oxidation in the biocover system using several approaches. The CH_4 oxidation efficiency was determined for single points in the biocover using two methods: a) Carbon mass balances (based on CH_4 and CO_2 concentrations in the deeper part of the cover and CH_4 and CO_2 surface flux measurements), and b) Tracer gas mass balances (based on CH_4 and tracer inlet fluxes and CH_4 and tracer surface flux measurements). A potential inhibitory effect from the presence of the tracer to the CH_4 oxidation process was evaluated in laboratory batch experiments. The CH_4 oxidation efficiency of the whole biocover was evaluated by establishment of a CH_4 mass balance for the biocover system (based on CH_4 inlet load and CH_4 surface flux measurements). Due to elevated temperatures in the biocover as a result of CH_4 oxidation and compost respiration processes, the field based CH_4 oxidation efficiency of the whole biocover was supported by laboratory batch experiments to study the temperature dependency of the CH_4 oxidation and compost respiration processes.

This paper is the second in a series of two. The first paper presents the design of the biocover system and investigations on the gas distribution in the biocover system (Cassini et al., 2017).

2. Materials & Methods

2.1 Laboratory experiments: Methane oxidation and respiration

Two batch laboratory experiments were carried out during the biocover study to evaluate compost respiration, CH_4 oxidation capacity, and the potential inhibitory effect from the added tracer gas (1,1,1,2-Tetrafluoroethane ($C_2H_2F_4$) commercially referred to as HFC-134a) toward the microbial CH_4 oxidation process (see next section). Biocover compost was sampled at two different locations of the biocover in November 2012 and April 2013, respectively. The two locations were chosen randomly to represent different parts of the biocover. At both occasions, samples were taken at different depths of the biocover using an auger and immediately sealed in tight plastic bags to avoid moisture evaporation. Sample locations are shown in Fig. 1. The first compost sample was taken close to T2 and used for the compost CH_4 oxidation and respiration test, while the later sample was taken at 5.2 and used for the tracer inhibition test. Two types of compost samples were used for the study: compost from a specific depth (-40 cm depth) and composite compost sample, obtained by manually mixing compost samples from different depths. The first composite sample consisted of a mix of compost from five depths (-20 cm, -40 cm, -60 cm, -80 cm, -90 cm), whereas the second composite sample consisted of compost from three depths (-20 cm, -40 cm, -60 cm). The sampling depth of 40 cm was chosen as gas concentration profiles and temperature measurements in the biocover indicated optimal conditions for CH_4 oxidation at this depth (see section 3.3 and Cassini et al., 2017).

For all batch experiments a standard experimental setup was used. Standard glass bottles (500 mL) were filled with 50 g of moist compost, previously sieved to avoid inhomogeneities. The bottles were sealed with gas tight butyl rubber septa (Norsorex DC 97 rubber stoppers) and aluminum caps. Bottles intended for CH_4 oxidation capacity evaluation were pre-treated overnight by injecting 80 mL of pure CH_4 to activate methanotrophic bacteria naturally present in the compost. This normally reduces lag phases and thus maximal CH_4 oxidation rates can be calculated using the initial CH_4 concentration and time. Bottles were re-opened and flushed with air to remove remaining CH_4 and CO_2. After the final sealing, gas was extracted from the bottles (120 mL for respiration bottles and 200 mL for CH_4 oxidation bottles) and replaced with pure gases to create suitable conditions for microbial reactions. A starting O_2 concentration of about 30 %vol. (120 mL) was chosen for compost respiration evaluation. Starting concentrations in bottles for CH_4 oxidation evaluation were 12-14 %vol. (80 mL) for CH_4 and 32-35 %vol. (120 mL) for O_2. The initial high O_2 content should assure sufficient O_2 for both CH_4 oxidation and compost respiration. Gas samples were transferred

every 12 hours from the bottles to standard vacuum 5 mL Exetainers© (Vial 819 W, 6 mL, Labco Ltd., UK) by using gas tight syringes. Gas samples composition (O_2, CO_2 and CH_4 concentrations) were analyzed by using a gas chromatograph (Agilent Technologies MicroGC 490). Calibration was performed using laboratory standard gas mixtures ranging from 1 to 40 %vol. As the Exetainers© are not completely vacuumed (5 mL vacuumed volume out of 6 mL total volume), the standard gas mixtures were injected (5 mL) into Exetainers© prior to analysis. Measured concentrations were plotted with time and generation or consumption rates of gases calculated (O_2 and CO_2 for respiration test, CH_4 and CO_2 for CH_4 oxidation). Maximum generation or consumption rates were determined by applying a zero order kinetic to the data for the initial phase of the test period. Compost moisture contents were separately measured and generation or consumption rates normalized to dry mass (DM). For all experiments duplicates and blanks (bottles with injected gases but without compost addition) were incubated to check for potential gas loss during the experimental run.

The CH_4 oxidation batch test series was started in November 2012 and had a duration of about 100 hours, while the respiration test was started in December 2012 and had a duration of 350 hours. In both experiments a composite compost sample (obtained by mixing equal amounts of compost from all sampled depths; -20 cm, -40 cm, -60 cm, -80 cm, -90 cm) were incubated at eight different temperatures (4°C, 10°C, 15°C, 20°C, 30°C, 40°C, 55°C, 70°C) with the purpose of analyzing the temperature dependence of CH_4 oxidation and respiration of a standard compost sample. The tested temperature range was comparable to the temperature range measured in the biocover (3 to 60°C) (Cassini et al., 2017). The moisture content on the incubated compost sample was 0.4 g g DM^{-1}, thus the samples were sufficiently moist, so moisture was not limiting CH_4 oxidation and temperature was the main controlling factor.

2.2 Laboratory experiments for tracer influence evaluation on methanotrophic oxidation

A tracer gas (HFC-134a) was injected during several measuring campaigns performed at AV Miljø biocover to calculate the CH_4 oxidation efficiency of the bacterial community (see section 2.4) and to study the gas distribution quality of the system (confer Cassini et al., 2017 for more details on the tracer experiment and gas distribution quality). In order to study the possible inhibitory effect of the tracer gas on the microbial methane oxidation activity, two laboratory experiments were carried out. The compost was sampled in April 2013 at location 5.2 (see Figure 4.1). For both experiments compost were sampled from the biocover at three depths (-20 cm, -40 cm and –60 cm). Two compost samples were then prepared; a

compost sampled from -40 cm depth and a composite compost sample consisting of an equal mix of compost from -20 cm, -40 cm and -60 cm depth. For both experiments, standard bottles (500 mL) were filled with 50 g sieved compost and sealed with butyl rubber septa and aluminum caps as in the batch tests described in the previous section (section 2.2). The compost in the bottles was pre-treated for 12 hours with 80 mL of CH_4 to shorten bacterial lag phase; bottles were then flushed with air before the experimental procedure started. The same experimental procedure as described in the previous section was adopted.

The first tracer batch test had a duration of about 130 hours. The purpose of this experiment was to evaluate the eventual potential inhibitory effect of HFC-134a gas on the microbial methane oxidation activity as a function of HFC-134a concentration. The compost sample from 40 cm depth was chosen to represent compost where significant CH_4 oxidation had been observed. The compost was incubated at 50°C, which was a typical temperature observed in the biocover at 40 cm depth (see Cassini et al., 2017). Air was extracted from the sealed bottle (200 mL) and replaced with 80 mL of CH_4 and 120 mL of O_2 as previously done for the CH_4 oxidation batch tests. Five different quantities of tracer (HFC-134a) were then injected into the bottles creating five increasing mixing ratios (0 ppmv, 8 ppmv, 40 ppmv, 90 ppmv and 140 ppmv). The highest tracer mixing ratio was well above the maximum field tracer gas mixing ratio of about 80 ppmv.

To confirm the findings of the first tracer gas inhibition test for other compost depths and temperatures, the second tracer gas batch test was carried out on both the compost sample from 40 cm depth and on the mixed compost sample. The compost was incubated at two temperatures (30°C and 50°C). The initial CH_4 concentration was between 14 and 18 %vol., while the initial tracer gas concentration was 160 ppmv. To conclude on any potential inhibitory effect on the methane oxidation activity, only half of the batches received tracer gas. For both tests, duplicates and blanks were prepared for most combinations. The moisture content on the incubated compost sample was 0.4 g g DM^{-1}, thus the samples were sufficiently moist, so moisture was not limiting CH_4 oxidation.

2.3 Determination of in-situ single point CH_4 oxidation by the carbon mass balance method

The CH_4 oxidation in a single point of the biocover can be determined by establishment of a carbon mass balance for that specific point. The carbon mass balance method is described in Christophersen et al. (2001), Einola et al. (2008), Philopoulos et al. (2008), Einola et al. (2009) and more recently in Pedersen et al. (2011). The method builds on the conservation of mass for carbon and assumes that the relationship

between the fluxes of CH_4 and CO_2 and the concentrations of CH_4 and CO_2 is equal in the bottom of the compost cover and that the vertical gas flow is uniform in the region of the sampling point. The method is based on the assumption of steady state and therefore assimilation of carbon by the methanotrophic bacteria is not accounted for (it is thus assumed that growth equals decay). Additionally, the dilution of CO_2 in percolating rainwater was assumed negligible, as was found in Christophersen et al., (2001). Finally, we did not consider CO_2 production from compost respiration. Combining Eq. 1 and 2 results in Eq. 3, which was used to calculate the load of CH_4 entering the biocover in a specific point. Eq. 4 was used to calculate the CH_4 oxidation rate, $R_{CH4,oxidation}$. Finally, the CH_4 oxidation efficiency, $MO_{Efficiency}$ (Eq. 5) was calculated.

$$J_{LFG,bottom} = J_{CO_2,surface} + J_{CH_4,surface} = J_{CO_2,bottom} + J_{CH_4,bottom} \qquad \text{(Eq. 1)}$$

$$\frac{J_{CH_4,bottom}}{J_{CH_4,bottom} + J_{CO_2,bottom}} = \frac{C_{CH_4,bottom}}{C_{CH_4,bottom} + C_{CO_2,bottom}} \qquad \text{(Eq. 2)}$$

$$J_{CH_4,bottom} = \frac{C_{CH_4,bottom}}{C_{CH_4,bottom} - C_{CO_2,bottom}} \cdot \left(J_{CO_2,surface} + J_{CH_4,surface} \right) \qquad \text{(Eq. 3)}$$

$$R_{CH_4 oxidation} = J_{CH_4,bottom} - J_{CH_4,surface} \qquad \text{(Eq. 4)}$$

$$MO_{Efficiency} = 100 \cdot \frac{J_{CH_4,bottom} - J_{CH_4,surface}}{J_{CH_4,bottom}} \qquad \text{(Eq. 5)}$$

$J_{LFG,bottom}$ is the load of landfill gas coming into the biocover, $J_{CO2,\ surface}$ is the surface CO_2 emission, and $J_{CH4,surface}$ is the surface CH_4 emission. $C_{CH4,bottom}$ and $C_{CO2,bottom}$ are the concentrations of CH_4 and CO_2 in the bottom of the biocover. $J_{CH4,bottom}$ is the load of CH_4 into the biocover and $R_{CH4oxidation}$ is the CH_4 oxidation rate.

To quantify the CH_4 oxidation efficiency at specific points in the biocover, gas concentrations of CH_4 and CO_2 at -95 cm depth in the biocover were determined in parallel with measuring surface fluxes of CH_4 and CO_2. Two field campaigns were carried out between November 2012 and January 2013 and included eight measuring points in the cover (S1, S7-8, S10, N1, N7-8, and N10). The surface flux chambers as well as the horizontal multi-port gas probe sampling system is described in details in Cassini et al. (2017). Each flux chamber was placed on the surface covering the same area as where the probes for gas concentration sampling were situated (the 8 sampling locations are shown in Fig. 1 as S1, S7-8, S10, N1, N7-8, and N10).

2.4 Determination of single point in-situ CH$_4$ oxidation by the tracer mass balance method

A new and innovative method for quantifying CH$_4$ oxidation was tested using the injection of an inert gas inside the biocover body. The inert gas, called tracer gas, was mixed into the inlet flow of LFG. Tracer gas inlet flow and surface fluxes in single points were measured (see Cassini et al., 2017). The tracer gas mass balance is based on the assumptions that the tracer gas is not subjected to production or consumption and that steady state of the tracer gas movement in the biocover has been reached. The CH$_4$ oxidation efficiency, MO$_{Efficiency}$ can be determined by measuring the CH$_4$ to trace gas flux ratio in the inlet and at the biocover surface using equation 6. It is noticed that the assumption of vertical 1-D flow set up for the carbon mass balance method is not needed for the tracer mass balance method.

$$MO_{Efficiency} = \frac{\left(\dfrac{J_{CH_4}}{J_{tracegas}}\right)_{Inlet} - \left(\dfrac{J_{CH_4}}{J_{tracegas}}\right)_{Surface}}{\left(\dfrac{J_{CH_4}}{J_{tracegas}}\right)_{Inlet}} \qquad \text{(Eq. 6)}$$

The tracer gas was continuously pumped at a controlled rate into the inlet mixing chamber and further distributed to the gas distribution layer and the CH$_4$ oxidation layer (confer Cassini et al. (2017) for more details on the performance of the tracer test). Tracer gas breakthrough curves in selected points of the horizontal multi-port gas probe sampling units were established by sampling and analyzing the tracer gas concentration over time in the duration of the tracer gas experiment. At the end of the tracer gas experiment, tracer gas emissions through the surface of the biocover was measured in 50 locations by use of a mobile static flux chamber (confer Cassini et al., 2017). A complete tracer gas mass balance (total influx of tracer gas equaled total surface flux of tracer gas) showed that steady state conditions were obtained at this point in time (Cassini et al., 2017). The CH$_4$ oxidation of the biocover was then determined by calculating the ratio of CH$_4$ to tracer gas surface fluxes in selected points and comparing these to the ratio of the inlet CH$_4$ to tracer gas flux (in accordance to Eq. 6).

2.5 Determination of whole biocover CH_4 oxidation based on CH_4 load and CH_4 surface emission

The whole biocover CH_4 oxidation efficiency was determined by comparing the measured CH_4 inlet load to the biocover with the integrated CH_4 surface emission from the biocover. The CH_4 load to the biocover was continuously monitored and surface emissions of CH_4 and CO_2 were measured using stationary flux chambers (both activities are described in Cassini et al. (2017)). The total CH_4 emission from the biocover was determined by kriging interpolation of the measured surface fluxes (see Cassini et al. 2017). In total, nine campaigns were conducted in the period from January 2013 to March 2014. At each campaign, the surface fluxes were measured in 50 locations in the biocover.

A tracer gas test was performed in the period May 16th-27th, 2013 where tracer gas was mixed into the inlet gas fed the biocover. At the end of the tracer gas test (on May 24th, 2013) tracer gas and CH_4 surface fluxes were measured in 50 locations in the biocover (Cassini et al. 2017). The CH_4 oxidation of the biocover was then determined following the approach explained in section 2.4 using Eq. 6 comparing the ratio of CH_4 to tracer gas in the inlet to the surface flux ratio. However, instead of determining the CH_4 oxidation in individual points, the surface flux CH_4 to tracer gas ratio was obtained by kriging interpolation of the measured surface fluxes of CH_4 and tracer gas (in the 50 locations) and thus the CH_4 oxidation of the whole cover was determined.

3. Results & Discussion

3.1 Methane oxidation and respiration of compost material

Table 1 shows the results of methane oxidation and respiration experiments performed at different temperatures on the composite compost sample from November 2012. The highest CH_4 oxidation rate (73 µg CH_4 gDM^{-1} h^{-1}) was obtained at 55°C. At 70°C, the CH_4 oxidation was significantly reduced (to 2.0 µg CH_4 gDM^{-1} h^{-1}) indicating an optimum temperature of 55°C or higher but below 70°C, which is higher than what was found at a previous study of compost sampled from the Klintholm Biocover (Scheutz et al., 2011d) where an optimal temperature of 45°C was observed. The high optimum temperature indicates presence of thermotolerant and/or thermophilic methanotrophs, which could be elucidated by performance of microbial analysis. Observed optimum temperature at 45-55°C is much higher than what is often reported for landfill cover soils (Scheutz et al., 2009).

Data from the compost respiration experiments showed a substantial O_2 consumption and simultaneous CO_2 generation activity in the biocover compost. Both the O_2 consumption and the CO_2 generation clearly increased when the temperature was increased. In the temperature range of 4 to 70°C, the O_2 consumption ranged between 1.1 and 65 µg O_2 gDM^{-1} h^{-1}, whereas the CO_2 production ranged between 0.5 and 73 µg CO_2 gDM^{-1} h^{-1}, with maximum rates obtained at a temperature of 70°C (Table 1). The respiration experiment thus showed that an optimal temperature was not reached as the highest O_2 consumption and CO_2 production was seen in the batches with the highest applied incubation temperature (70°C). This is probably not surprising since high temperatures (>70°C) are observed in composting windrows (e.g. Andersen et al., 2010). Composting is a self-driven process as it is exothermic and it will thus perform better at higher temperatures.

Table 2 shows additional results regarding CH_4 oxidation rates from batch experiments, but achieved on compost samples collected in the biocover six months later (April 2013) and at another location (5.2). CH_4 oxidation rates for experiments performed at 50°C were in the same range (57 to 79 µg CH_4 gDM^{-1} h^{-1}) as the first incubation experiments. However, for experiments carried out at 30°C consistently higher CH_4 oxidation rates were observed for the April sample compared to the November sample, (131 µg CH_4 gDM^{-1} h^{-1} compared to about 25 µg CH_4 gDM^{-1} h^{-1}). Thus there seems to be different temperature optimums depending on sample location and/or time of sampling. Factors that could explain the different CH_4 oxidation rate observed at the two sampling locations (T2 and 5.2) include temperature and moisture differences in the biocover, and CH_4 exposure differences in terms of both CH_4 load to the oxidation layer and pre-exposure time (compost was sampled with about 5 months apart). No considerable difference could be observed between the CH_4 oxidation capacity of compost from a particular depth (-40 cm) and the composite compost sample made out of compost from different depths (Table 2) indicating a broader zone (from -20 to -60 cm) in the biocover with methanotrophic activity.

A wide range of CH_4 oxidation rates have been reported in a previous study (Scheutz et al., 2011e). Measured CH_4 oxidation rate at 22°C ranged between 14 and 168 µg CH_4 gDM^{-1} h^{-1} demonstrating a wide variability of the methanotrophic activity. These results are widely comparable with the ones obtained in the study presented here with a CH_4 oxidation rate at 30°C ranging between 25 and 131 µg CH_4 gDM^{-1} h^{-1}.

3.2 Potential inhibitory effect of tracer addition on methane oxidation activity

The result of the first experiment evaluating the potential inhibitory effect of the presence of HFC-134a to the CH_4 oxidation activity is displayed in Fig. 2. The CH_4 oxidation rates for all the different HFC-134a concentrations (0 to 140 ppmv) were almost equal and ranged between 20 and 25 µg CH_4 gDM^{-1} h^{-1} (Fig. 2). Thus no noticeable influence was found on the performance of CH_4 oxidation due to the presence of HFC-134a. These results are also corroborated by the second tracer experiment performed in May 2013 and presented in Table 2. CH_4 oxidation rates measured on mixed compost or depth-specific compost at both 30 and 50°C did not show any change depending on tracer presence, validating the results achieved in the first test. This means that the performance of the tracer test in full scale in the biocover will not have any influence on the CH_4 oxidation performance of the biocover during the tracer release.

3.3 Single point CH_4 oxidation – comparison of the carbon balance and the tracer gas method

Fig. 3 shows representative vertical gas composition profiles measured at two locations in the Southern and Northern sampling transects (S8 and N8). Gas composition profiles in the biocover indicated CH_4 oxidation especially in the lower part of the biocover (100 to 50 cm below the surface). It is evident that also the lower part contained significant concentrations of oxygen (often 3-5% or more) from the atmosphere as well as from the fed gas. The presence of O_2 at all depths of the cover indicated that CH_4 oxidation in the field was not limited by available O_2.

CH_4 oxidation in the biocover was also demonstrated by the vertical distribution of the ratio between the concentrations of CH_4 and CO_2 with the concentration of HFC-134a ($C_2H_2F_4$) as shown in Fig. 4 (locations S1, S4, S8 and S10 in the Southern transect). The profiles were measured after tracer breakthrough had been reached. Gas concentration profiles represented in Fig. 4 clearly show how CH_4 and CO_2 concentrations change with depth in the biocover due to methanotrophic activity. The $CH_4/C_2H_2F_4$ ratio clearly shows a decreasing trend from the bottom to the upper surface of the biocover demonstrating that CH_4 is being oxidized during its flow through the biocover pores. At the same time the $CO_2/C_2H_2F_4$ ratio describe an increasing trend during its flow through the biocover. In this case, the CO_2 production from CH_4 oxidation is however not the only reason of the ratio variation because compost respiration is also active in the biocover. The latter is clearly shown by the higher mixing ratio of $CO_2/C_2H_2F_4$ (up to 6000 in the upper

part of the biocover) in comparison to the $CH_4/C_2H_2F_4$ ratio (starting from 400 at the lower part of the biocover).

Table 3 compares CH_4 oxidation efficiencies and rates obtained by using the carbon mass balance method (as described in section 2.3) and the trace gas method (see section 2.4) obtained from eight single points during two campaigns. Using the carbon mass balance approach, a CH_4 oxidation efficiency in the range of 84 to a 100% was obtained whereas using the tracer gas approach resulted in a slightly higher CH_4 oxidation efficiency ranging from 97 to 100% (Table 3). One of the uncertainties related to the carbon mass balance method is the assumption that the measured CO_2 emission is caused solely by CH_4 oxidation. However, in reality a part of the emitted CO_2 is due to compost respiration. Considering CO_2 from compost respiration in the carbon mass balance (Eq. 1-5) will however reduce the CH_4 oxidation efficiency further thus not explaining the observed difference between the two methods. Subtracting a surface CO_2 flux caused by respiration from the measured CO_2 surface flux (Eq. 1) will result in a lower CH_4 flux to the bottom of the biocover (Eq. 3) and thus a lower oxidation rate (Eq. 4) and oxidation efficiency (Eq. 5). The CH_4 oxidation rates were much higher using the tracer gas balance method giving CH_4 oxidation rates between 7 and 124 g m^2 d^{-1} in comparison to the using carbon mass balance, which gave oxidation rates between -0.06 and 40 g m^2 d^{-1}. The higher CH_4 oxidation rates obtained using the tracer gas mass balance approach is most likely due to fact that CH_4 oxidized in the gas distribution layer is included in the oxidation, since it is the ratio between inlet tracer and surface emitted tracer which is calculated (Eq. 6). In the carbon mass balance it is the gas composition in the bottom of the biocover that is the starting point (Eq. 1-5) and in this case, the CH_4 that is oxidized in the gas distribution layer (before it enters the compost layer) is not included. Another difference between the two methods is that the assumption of 1D gas flow in the carbon mass balance method, is not necessary for the tracer method. The flow in the methane oxidation layer may not be complete vertical making some additional uncertainty in the carbon mass balance method.

The correlation between the two methods were relatively weak (R^2 <0.6), which most likely is caused by inhomogeneities in the biocover system – both in the gas distribution system as well as in the compost layer. The results showed significant spatial variations in CH_4 load and CH_4 oxidation and also showed a much higher CH_4 oxidation capacity in some single points in comparison to the average whole biocover CH_4 oxidation rates of 13 g m^2 d^{-1} (see next section). In general, higher CH_4 oxidation rates were obtained for single points located in the Southern multi port sampling transect (Fig. 1; S1-S10), which most likely was due

to the fact that this part of the biocover received a higher CH_4 load as it was closer to the un-slotted inlet gas load pipe. In addition to this, a significant part of the CH_4 might be oxidized in the gas distribution layer once the gas enters the slotted gas distribution pipes, why the CH_4 load to CH_4 oxidation layer in the Northern part is lower in comparison to the Southern part. This was supported by the results of a tracer gas experiment, which showed equal distribution of tracer gas to different parts of the biocover by the un-slotted gas distribution pipe and even distributed emissions of tracer gas over the biocover area (Cassini et al., 2017). Combined the results show that both CH_4 and tracer gas was efficiently distributed throughout the Southern section of the biocover but that CH_4 most likely was oxidized before reaching the Northern part of the biocover, whereas the tracer gas was equally distributed throughout the biocover (both the Southern and the Northern section). In all parts of the biocover, the CH_4 oxidation capacity was however higher than the CH_4 load resulting in very low surface CH_4 emissions.

3.4 Whole biocover CH_4 oxidation based on CH_4 load and CH_4 surface emission

As shown in the presentation of the performed surface fluxes (confer Cassini et al., 2017) very low CH_4 fluxes were recorded in general varying between negative emissions (-0.006 mol m^{-2} d^{-1}) indicating atmospheric CH_4 uptake to maximum CH_4 surface fluxes of about 0.12 mol m^{-2} d^{-1}. In comparison to CH_4 fluxes, much higher CO_2 fluxes were recorded indicating significant CH_4 oxidation and compost respiration. A few areas with higher CH_4 emissions were observed indicating some spatial emission variation. Table 4 presents CH_4 oxidation efficiencies and CH_4 oxidation rates for the whole biocover system determined based on the CH_4 load (inlet CH_4 concentrations and gas flow recording) and surface CH_4 emissions measurements. Overall, the CH_4 oxidation efficiency of the biocover varied between 81 and almost 100% - in most cases over 94% - and showed that the pilot plant biocover system installed at AV Miljø Landfill was very efficient in oxidizing the landfill CH_4. The CH_4 oxidation rate varied between 8 and 18 g m^{-2} d^{-1} (Table 4). The average biocover performance was estimated to be around 13 g m^2 d^{-1}, which is relatively low in comparison to previous biocover studies (Scheutz et al., 2009). Considering the high CH_4 oxidation efficiency it is likely that the biocover was over-dimensioned and had the potential to oxidize a much higher CH_4 load if needed (see further below).

The laboratory-measured CH_4 oxidation potential as function of temperatures can be used to estimate the whole biocover CH_4 oxidation potential (assuming that the CH_4 oxidation, which is occurring in the field is not limited by O_2 and/or by moisture content). The methane oxidizing layer of the biocover was divided in five

horizontal layers (20 cm each) and temperature homogeneity assumed for each stratum. Reference

temperatures were chosen based on data from probes installed in the biocover soil (as described in Cassini

et al., 2017). The maximal CH_4 oxidation potential of the biocover was then calculated by considering the

methanotrophic oxidation rate measured during the laboratory incubation studies for each reference

temperature (results reported in section 3.1 and data shown in Table 1). The specific CH_4 oxidation potential

was calculated in this way to be around 300 g CH_4 m^{-2} d^{-1} (assuming no O_2 or moisture limitation). If the

higher CH_4 oxidation rates obtained in the laboratory experiments testing the potential inhibition of HFC-134a

(Table 2) were used instead, a specific CH_4 oxidation potential higher than 300 g CH_4 m^{-2} d^{-1} would be

obtained. The results of the field measurement campaigns showed significantly different results. The

average specific biocover performance was as previously mentioned calculated to be between 8 and 18 g

CH_4 m^{-2} d^{-1}, but peaks of noticeably higher values where measured in different points of the biocover from

time to time (up to124 g CH_4 m^2 d^{-1} as reported in the previous section). It is reasonable to conclude that the

maximal CH_4 oxidation potential of the biocover is several times higher than the average performance

calculated based on the whole-biocover measuring campaigns. This is both shown from theoretical

calculations (based on lab experiments) and from field (point flux chambers) results. The reason for the low

specific biocover performance of 8-18 gCH_4 m^{-2} d^{-1} is that the load of CH_4 injected in the biocover was much

lower than the biocover CH_4 oxidation potential, which could be realized with a higher CH_4 load. We do not

find it likely that the CH_4 oxidation occurring in the biocover was limited by O_2 or by compost moisture as O_2

was present at all depths of the biocover and as the volumetric compost moisture content ($m^3 \cdot m^{-3}$) varied

between about 0.2 and 0.5 (Cassini et al., 2017), which is a sufficient moisture content for CH_4 oxidation

(neither too low nor too high) (Scheutz et al., 2009). Mor et al. (2006) obtained maximum CH_4 oxidation rates

in compost at moisture contents ranging from 45% to 110% (dry weight basis). A volumetric compost

moisture content of 0.2 to 0.5 corresponds to moisture contents of 40 to 100 % (dry weight basis) (when

assuming a compost bulk density of 500 kg m^{-3}), which is within the moisture range for optimal CH_4 oxidation

reported by Mor et al. (2006) and thus indicating that moisture was most likely not a limiting factor for CH_4

oxidation in the biocover.

However, high CH_4 oxidation efficiencies in the range of 81-99% were measured and the biocover was

demonstrated to be efficient during the whole study period. Reasons for the substantial efficiency of the

biocover were the optimal distribution of O_2 at every depth of the biocover, the relatively low CH_4 load to the

biocover, the efficient and homogeneous gas distribution throughout the Southern section of the biocover perpendicular to the gas inlet, and significant CH_4 oxidation in the gas distribution layer. High CH_4 oxidation efficiencies were even found during winter time with low ambient temperatures, which in previous studies brought the methanotrophic activity to stagnation (Stern at al., 2007). During winter time when the ambient air temperature was between -5 and +5°C, the temperature in the biocover remained high and closed to the optimum temperature (around 55°C) as measured in the laboratory incubation experiment. This obviously had a positive effect on the methanotrophs resulting in high CH_4 oxidation rates and efficiencies. Previous studies reported optimal temperature for methanotrophic bacteria ranging between 25-30°C (Scheutz et al., 2009) and 30°-40°C (Streese and Stegmann, 2003). Gas profiles (Fig. 3) showed that the maximal CH_4 oxidation activity was measured between -40 cm and -90 cm, which was also where optimal temperatures were recorded and where also O_2 was present.

3.5 Evaluation of compost respiration by biocover carbon balance

The carbon mass balance application as shown in Table 4 proved that the total emitted carbon (sum of C-CH_4 and C-CO_2) from the biocover surface was considerably higher than the carbon load into the system. Considering the CO_2 load in the inlet (about 4-6 kg C d^{-1}) and the CO_2 generated from methanotrophic activity (about 4-8 kg C d^{-1}), the additional CO_2 must be produced from respiration of the compost (about 0-76 kg C d^{-1}, when subtracting the carbon in the CO_2 in the inlet gas and the carbon in the CO_2 generated from CH_4 oxidation from the carbon in the total CO_2 emission) (Table 4). The average estimated carbon flux caused by compost respiration activity based on field measurements for the year 2013 steadily decreased from a value of 76 kg C d^{-1} in March to a value of 14 kg C d^{-1} in October (Table 4). For the last campaigns in 2014, the estimated carbon flux caused by compost respiration activity had further decreased to a very low level (the estimated values are actually below zero, which probably is due to uncertainties in the measured inlet load and surface emissions). Similar decrease of the respiration with time was observed at the Klintholm Biocover (Scheutz et al., 2014).

The respiration potential of the biocover compost for the early period (January - May 2013) was estimated using the laboratory data previously presented. The CH_4 oxidation layer was divided into 5 horizontal parallel layers and for each layer a representative temperature was assumed. Data collected from temperature probes (confer Cassini et al., 2017) installed at different depths in the biocover CH_4 oxidation

layer were analysed. A flexible temperature range was adopted based on the corresponding probe data to simulate the changing seasonal temperature trend. Respiration rates determined by the laboratory experiments were used (as presented in section 3.1). A maximum and minimum compost respiration potential was calculated by associating a respiration rate range to the temperature range. The calculated CO_2 generation rates showed plausible results. The calculated respiration range was 10 - 33 kg C d^{-1}, which is in the same range as the previously reported average estimated carbon flux contributed by compost respiration activity based on field measurements; 12 - 26 kg C d^{-1} - omitting the exceptional high values estimated for the 2nd and the 3rd measuring campaigns (see Table 4). For these two campaigns the field results showed double of the maximal load calculated with the laboratory studies. The use of kriging interpolation in case of considerably higher emission hot spots (6-8 times the average value) can give over estimation of the integrated carbon flux and may be the reason for the deviation between field measurements and laboratory result based estimations for the 2nd and the 3rd measuring campaign.

4. Conclusion and Perspectives

Laboratory batch incubation experiments performed by use of compost sampled from the CH_4 oxidation layer of the biocover system showed a high CH_4 oxidation activity in the whole range of temperatures (4 to 70 °C) observed in the biocover system. The experiment showed a temperature optimum for the CH_4 oxidation process of 55°C, which is much higher than temperature optimum measured for landfill cover soils. Respiration tests performed in the same temperature range showed the highest respiration activity for the highest temperature tested (70°C). The test revealed that the biocover will emit considerable amounts of CO_2 as a result of compost respiration.

Laboratory experiments performed with and without the addition of the used tracer gas, HFC-134a, showed no inhibitory effect from the tracer gas in a relevant concentration range (up to 160 ppmv) on the CH_4 oxidation activity in the compost layer. This means that the tracer gas can safely be added to the inlet gas without affecting the ongoing CH_4 oxidation process.

CH_4 balances of the biocover established by measuring the CH_4 load to the biocover system and the emitted CH_4 through the biocover surface (through integration of flux measurements performed in over fifty points on the biocover surface) showed that the system had a very high CH_4 oxidation efficiency independent

of the time of the year – in most cases close to 100% efficiency. The active CH_4 oxidation process was further documented by point measurements of the CH_4 oxidation capacity using a carbon balance approach and a new-developed method using information about CH_4 and tracer gas fluxes. The tracer-based method gave more realistic CH_4 oxidation efficiencies in comparison to the carbon balance method, and was proven to be an efficient method for quantifying the CH_4 oxidation efficiency of a pipe-loaded biocover system, where the tracer gas easily can be added to the gas inlet.

The CH_4 balances of the biocover showed that the CH_4 load to the biocover system was in general lower than anticipated and resulted in relative low methane oxidation rates – on an absolute scale between 8 and 18 g CH_4 m^{-2} d^{-1}. Extrapolating the laboratory determined CH_4 oxidation rates assuming no O_2 limitation and using oxidation rates corresponding to the temperatures measured in the biocover, a CH_4 oxidation potential of 300 g CH_4 m^{-2} d^{-1} was calculated; much higher than the actual CH_4 oxidation rate, which most likely was limited by the low CH_4 load to the system.

At many landfills a significant part of the generated landfill gas may emit through leachate wells. At such landfills, the CH_4 emission may efficiently be mitigated by a biocover system similar to the pilot-scale system implemented at the AV Miljø Landfill. However, additional studies need to be performed to construct the optimal gas distribution system – the distance between the gas distribution pipes can maybe be increased to reduce costs. The CH_4 load to the biocover system could be significantly increased to evaluate the in-field capacity of the biocover system.

Acknowledgments

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Tables

Table 1

Results of methane oxidation and respiration experiments performed at different temperatures on the composite compost sample from November 2012 at location T2. The compost moisture content was 0.4 g g DM^{-1} and the initial CH_4 concentration was 14 %vol. All rates are in the unit µg g DM^{-1} h^{-1}.

Temperature (°C)	Methane oxidation experiment			Respiration experiment	
	CH_4 oxidation	O_2 consumption	CO_2 production	O_2 consumption	CO_2 production
4	0.13[a]	4.7[a]	2.1[a]	1.1 ± 0.23	0.52 ± 0.05
10	0.04[a]	3.4[a]	2.8[a]	1.4 ± 0.28	1.0 ± 0.04
15	1.8 ± 0.02	14.2 ± 0.1	4.5 ± 0.7	2.8 ± 0.29	2.8 ± 0.38
20	6.0 ± 0.5	27 ± 2.0	15 ± 1.1	4.5 ± 0.72	4.2 ± 0.30
30	25 ± 1.5	76 ± 19	51 ± 8.6	5.4 ± 0.26	5.6 ± 0.19
40	65[a]	210[a]	170[a]	22 ± 8.0	26 ± 9.0
55	73 ± 4.6	290 ± 8.7	230 ± 18	39 ± 0.57	43 ± 1.1
70	2.0 ± 0.11	84 ± 8.7	91 ± 17	65 ± 0.16	73 ± 1.3

a: The batches for the methane oxidation experiments at 4, 10 and 40 °C were not duplicated.

Table 2

Methane oxidation rates determined in batch tests for depth specific compost samples (sampled at -40cm depth) and for composite compost samples (obtained by mixing equal amounts of compost sampled at -20 cm, -40 cm and -60 cm depth). First experimental round made on depth specific sample (-40cm) at HFC-134° concentration in the range of 0-140 ppmv at 50°C. Second experimental round made both on composite and depth specific sample with either no addition of HFC-134° or HFC-134° addition at fixed concentration (160 ppmv) and at two temperatures (30°C and 50°C). The compost moisture content was 0.4 g g DM^{-1}. All rates are in the unit $\mu g\ CH_4\ g\ DM^{-1}\ h^{-1}$.

Compost	Initial CH_4 conc. (%)	CH_4 oxidation rate: No HFC-134a addition		CH_4 oxidation rate: HFC-134a addition		Initial HFC mixing ratio (ppmv)
		30°C	50°C	30°C	50°C	
First experimental round						
Depth specific (-40cm)	14				26.0 ± 0.6	0
	14				22.2 ± 1.7	8
	14				23.1 ± 3.0	40
	14				22.7 ± 1.2	90
	14				22.5 ± 0.2	140
Second experimental round						
Composite sample	14	123.8 ± 3.9	57.5 ± 0.1	123.8[a]	54.8[a]	160
	18	131.6 ± 3.8	79.2 ± 0.6	130.7[a]	72.5[a]	160
Depth specific (-40cm)	14	126.2 ± 5.5	57.4 ± 5.9	125.9[a]	58.1[a]	160
	18	124.7 ± 6.1	71.6 ± 8.5	128.8[a]	68.5[a]	160

a: The batches for the methane oxidation experiments with HFC-134° addition in the second experimental round were not duplicated.

Table 3
Comparison of CH_4 oxidation efficiencies and CH_4 oxidation rates obtained using two different methods; the carbon mass balance (CMB) method and the tracer mass balance (TMB) method.

Measure-ment point	Date	CH_4 Oxidation Efficiency (%)		CH_4 Oxidation Rate (g $CH_4 \cdot m^{-2} \cdot d^{-1}$)	
		CMB	TMB	CMB	TMB
S1	08.12.12	94.4	98.7	27.38	124.18
	24.05.13	97.3	98.6	39.91	85.04
S7-S8	08.12.12	91.1	98.9	8.71	75.19
	24.05.13	97.2	98.3	14.43	37.07
S10	08.12.12	96.2	98.7	23.55	68.86
	24.05.13	97.7	98.7	4.29	7.41
N1	08.12.12	80.5	-	0.15	-
	24.05.13	65.0	97.4	1.37	54.46
N7-N8	08.12.12	100.0*	-	-0.06	-
	24.05.13	67.8	98.2	1.55	47.02
N10	08.12.12	100.0*	-	-0.04	-
	24.05.13	56.3	98.0	0.37	22.57

- not analyzed; *: negative CH_4 emissions were measured indicating that the biocover at these points take up atmospheric CH_4 in addition to the landfill gas CH_4. As a result the CH_4 oxidation efficiency has been set to 100%.

Table 4

Overview of whole biocover CH_4 oxidation efficiencies and CH_4 oxidation rates obtained during nine measuring campaigns performed from January 2013 to March 2014.

Date	Inlet load		Total surface emission		CH_4 oxidation rate		CH_4 oxidation efficiency	CO_2 from compost respiration*
	g C-CH_4 d^{-1}	g C-CO_2 d^{-1}	g C-CH_4 d^{-1}	g C-CO_2 d^{-1}	g C-CH_4 d^{-1}	g CH_4 m^2 d^{-1}	%	g C-CO_2 d^{-1}
25.01.13	6,930	6,121	19	25,007	6,911	18.3	>99	11,975
08.03.13	7,732	5,370	332	88,386	7,516	15.0	96	75,500
11.04.13	8,066	4,537	1,541	77,623	6,526	13.0	81	66,560
21.04.13	7,020	6,265	35	36,277	6,985	14.0	>99	23,027
02.05.13	6,396	7,242	30	39,489	6,366	12.7	>99	25,881
24.05.13	4,046	3,760	246	28,219	3,800	7.6	94	20,659
25.10.13	4,327	3,856	42	22,050	4,285	8.6	>99	13,909
02.02.14	5,917	5,238	315	11,024	5,602	11.2	95	0,184
29.03.14	6,841	6,081	17	8,266	6,824	13.6	>99	-4,639

* CO_2 from compost respiration = Total CO_2 surface emission – Inlet CO_2 flux – CO_2 generated from CH_4 oxidized
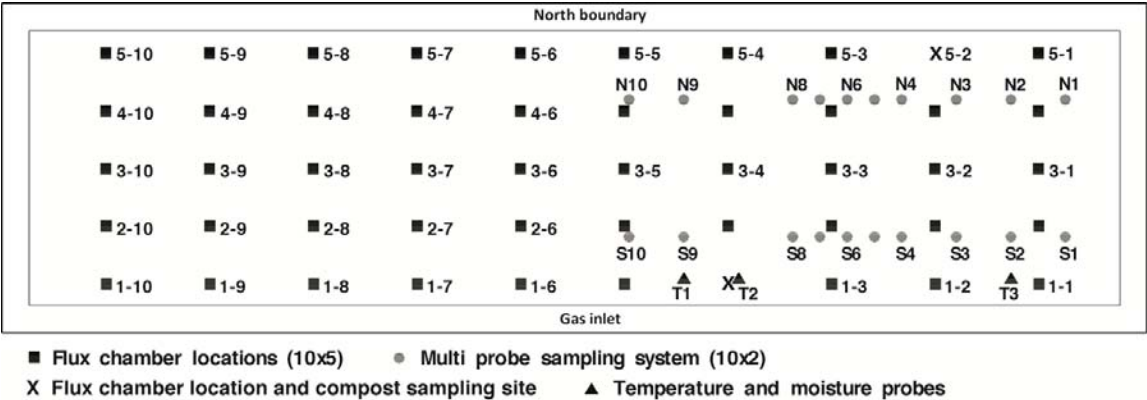
Figures

Fig. 1. Map of the biocover with indications of locations of flux chamber measurements, the multi probe sampling locations, the temperature/moisture probes and compost sampling sites.
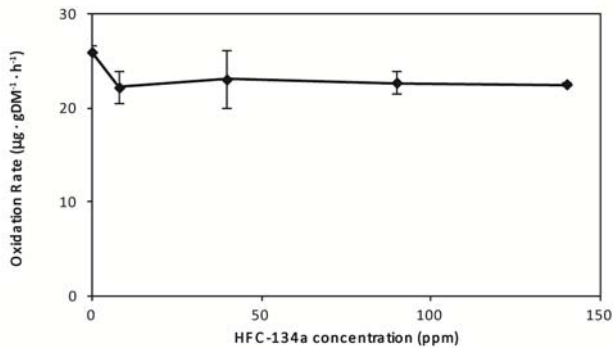

Fig. 2. Methane oxidation rates determined in batch incubation tests shown as a function of initial tracer gas (HFC-134a) concentrations in the batches (0 to 140 ppmv).
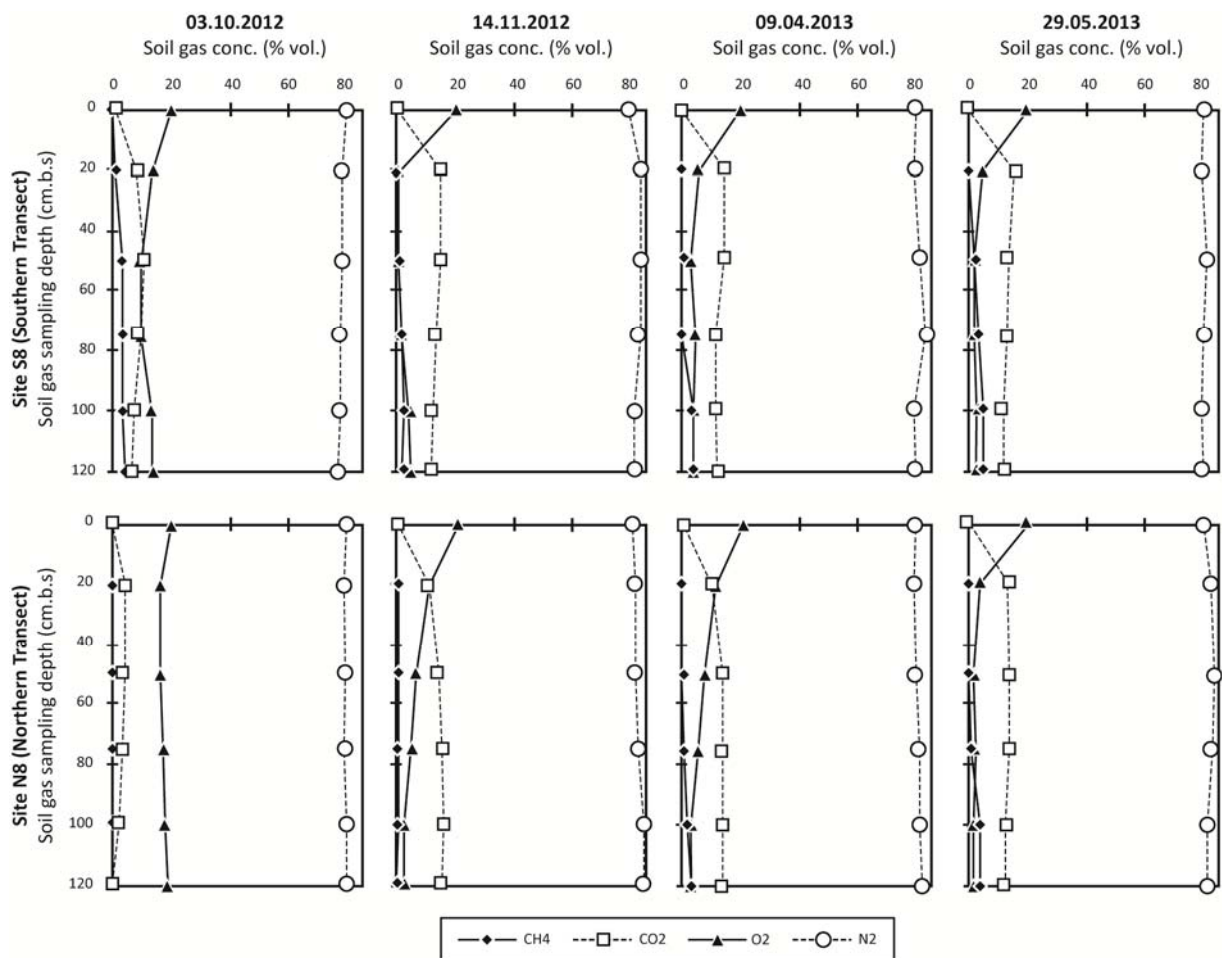
Fig. 3. Vertical gas concentration profiles for CH_4, CO_2, O_2 and N_2 at two selected locations (S8 and N8) and times (October 3th, 2012, November 14th, 2012, April 9th, 2013 and May 29th, 2013).
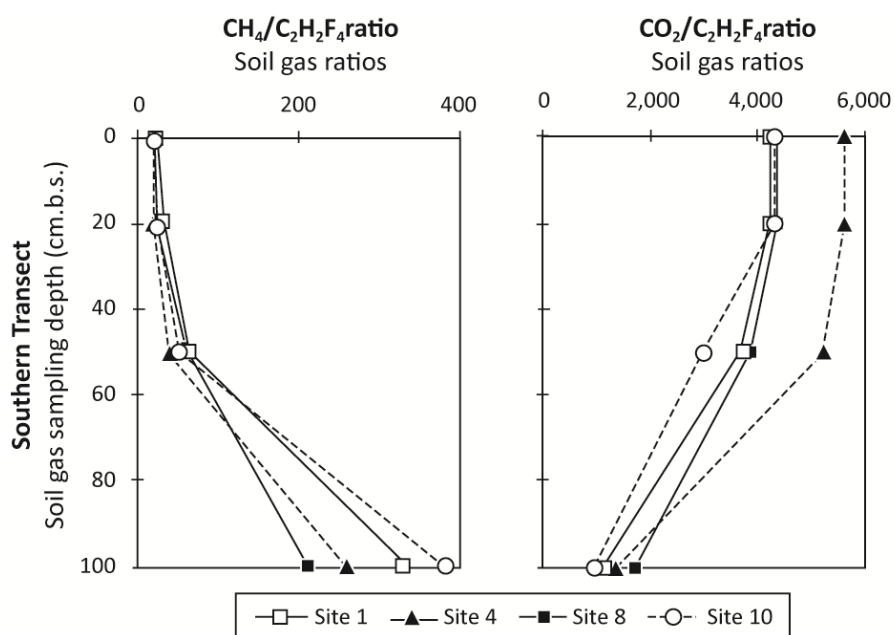
Fig. 4. Vertical gas concentration profiles of the ratios $CH_4/C_2H_2F_4$ and $CO_2/C_2H_2F_4$ at four selected locations in the Southern transect (S1, S4, S8, and S10) as measured on December 8th, 2012. Ratios were calculated based on mixing ratios given in ppmv of the individual gases.