Mitigation of methane emissions in a pilot-scale biocover system at the AV Miljø Landfill, Denmark

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October 2014

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Report [Nr.]
2014

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Cover photo: [Text]
Published by: Department of Environmental Engineering, Miljoevej, Building 113, DK-2800 Kgs. Lyngby, Denmark
Request report from: www.env.dtu.dk
ISSN: [0000-0000] (electronic version)
ISBN: [000-00-0000-000-0] (electronic version)

ISSN: [0000-0000] (printed version)
ISBN: [000-00-0000-000-0] (printed version)

Preface

This report is the result of a collaborative project between DTU Environment and AV Miljø carried out in the period from April 2011 until December 2013. We like to thank for the contribution by Svend Erik Christensen, Per Wellendorph and Finn R. Jensen from AV Miljø as well as Jonas Nedenskov from Amager Ressource Center, who all have actively contributed to the project and especially to the establishment of the pilot-scale biocover system at the AV Miljø Landfill.

During the project several student projects have been carried out parallel to the on-going research project. I like to thank the students Christian Pilegaard Jespersen and Stefan Emil Danielsson (contributions to the tracer experiments) and Hans Henrik Jørgensen, Monika Margrethe Skadborg and Rune Skovsø Møller (contributions to the whole biocover methane oxidation measurements) for very valuable contributions to the project.

Kgs. Lyngby, October 2014

Peter Kjeldsen

Content

Summary

Greenhouse gas mitigation at landfills by methane 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. The biocover was fed by landfill gas pumped out of three leachate wells. An innovative gas distribution system was used to overcome the often observed overloaded hot spot areas resulting from uneven gas distribution to the active methane oxidation layer consisting of garden compost. Performed screening of methane and carbon dioxide concentration at the surface of the biocover showed homogenous distributions indicating an even gas distribution. This was supported by result from performed tracer tests where the compound HFC-134a was added to the gas inlet over a period of up to12 days. Studies of the tracer movement within the biocover system showed very even gas distribution in gas probes installed in the gas distribution layer. Also the flux of tracer out of the biocover surface as measured by flux chamber technique showed a spatially even distribution.

Compost samples were taken out at several occasions from the methane oxidation layer and tested in the laboratory for methane oxidation and respiration potential performed in a temperature range of 4 to 60°C. The temperature range reflected the temperatures observed by installed temperature probes in the methane oxidation layer. The laboratory experiments showed high methane oxidation potential even at temperatures up to 60°C but also a significant respiration of the matured compost. Other performed batch experiments showed the adding concentrations of the tracer (HFC-134a) in a concentration range similar to the range observed in biocover pore gas during the tracer experiment did not have an influence on the methane oxidation process ie no inhibitory effects from the tracer was observed.

The whole biocover CH_4 oxidation efficiency was determined by measuring the CH_4 inlet load and CH_4 surface fluxes using the static flux chamber technique. In addition CH_4 oxidation was determined for single points 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 trace 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 installed at AV Miljø landfill was very efficient in oxidizing the landfill CH_4. The average CH_4 oxidation rate measured at seven campaigns was approximately 14 g m^{-2} d^{-1}. The carbon mass balance approach compared reasonable 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 oxida-

tion 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}. Extrapolation of the results from the laboratory experiments to field conditions showed that the biocover system may have a much higher methane oxidation potential and could be loaded with a larger flux of methane without losing much efficiency. A high emission of CO_2 was observed at the biocover. Analysis and calculation revealed that most of the emitted carbon dioxide originate from respiration of the compost contained in the methane oxidation layer. The carbon is however of biogenic nature and do not contribute to the greenhouse effect.

Sammendrag

Reduktion af drivhusgasser fra deponeringsanlæg ved mikrobiel metanoxidation i etablerede biocover systemer kan være en omkostningseffektiv teknologi. Der er imidlertid kun blevet gennemført kvantitative evalueringer af effektiviteten af teknologien i fuld skala i nogle få tilfælde. Et tredje generations semi-passivt biocover system er blevet etableret på lossepladsen AV Miljø. Biocoversystemet var udført i pilot skala med et samlet areal på 500 m^2 og behandlede lossepladsgas opsamlet fra tre perkolatbrønde. Et innovativ gasdistributionssystem blev brugt til at undgå metanudslip fra overbelastede hot spot områder opstået som et resultat af ujævn gasfordeling til det aktive metanoxidationlag (bestående af havekompost). Screeninger af metan- og kuldioxidkoncentrationen på overfladen af biocoveret viste homogene koncentrationsfordelinger, hvilket indikerer en jævn gasdeling til metanoxidationslaget. Dette blev yderligere understøttet af resultatet af udførte sporstofforsøg, hvor forbindelsen HFC-134a blev tilsat gastilløbet over en periode på op til 12 dage. Undersøgelser af sporstoftransporten i biocoversystemet viste at gassen forelte sig relativt jævnt i systemets gasfordelingslag. Målinger af fluxen af sporstof ud af biocoveroverfladen viste også en rumligt ensartet fordeling .

Kompostprøver blev udtaget ved flere lejligheder fra metanoxidationlaget og testet i laboratoriet for metanoxidations- og respirationspotentiale udført i en temperatur mellem 4 og 60 ° C. Temperaturområdet afspejlede temperaturer observeret via installerede temperaturfølere i metanoxidationlaget. Laboratorie-forsøgene viste høj metanoxidationspotentiale selv ved temperaturer op til 60 ° C , men også en betydelig respiration i det modnede kompost. Andre udførte batchforsøg viste at tilførelse af sporstoffet (HFC- 134a) i det i sporstofforsøget benyttede koncentrationsområde ikke havde en indflydelse på metan oxidationsprocessen i komposten, dvs. ingen inhiberende virkninger fra sporstof blev observeret.

Hele biocoverets metanoxidation blev bestemt ved måling af metantilførelsen i indløbet og metanfluxen i 50 målepunkter fordelt over biocoverets overflade målt ved hjælp af statiske fluxkamre. Desuden blev metanoxidationen bestemt for specifikke lokaliteter på biocoveret ved hjælp af to forskellige metoder, en kulstofmassebalancemetode (baseret på metan- og kuldioxidkoncentrationer i den dybere del af biocoveret og metan- og kuldioxid-overflade fluxmålinger) og en sporgasmassebalance metode (baseret på metan- og sporstof indløbsbelastning og metan- og sporstoffluxmålinger målt på biocoveroverfladen) . Den totale virkningsgrad for metanoxidation for hele biocoveret varierede mellem 81 og 100% og viste, at pilotskalaanlægget var meget effektivt til at oxidere metan indeholdt i lossepladsgas. Den gennemsnitlige CH4 oxidationsrate målt ved syv kampagner var cirka 14 g $m^{-2}d^{-1}$. Der var relativt god overensstemmelse mellem kulstofmassebalancemetoden og sporgasmassebalancemetoden, når metoderne anvendes til kvantificering af lokale metanoxidationsrater. Oxidationseffektiviteter målt med de to metoder

blev observeret i intervallet 84 til 100% . Metanoxidationrater observeret ved hjælp af sporgas-balancemetoden gav metanoxidationrater mellem 7 og 124 g m^2 d^{-1} i forhold til kulstofmassebalancemetoden, som gav metanoxidationrater mellem -0.06 og 40 g m^2 d^{-1}.

Ekstrapolation af metanoxidationspotentialer målt i laboratorieforsøg til felt viste, at biocoversystemet formentlig har en meget større evne til at reducere metanudslip og vil kunne belastes med en større flux af metan uden at effektivitet af biocoveret reduceres nævneværdigt. En høj udledning af kuldioxid fra biocoveret blev observeret. Analyser og beregninger viste, at størstedelen af den emitterede kuldioxid stammer fra respiration af komposten indeholdt i metanoxidation lag. Kulstof er dog af biogen natur og bidrager ikke til drivhuseffekten.

1. Background and scope

Landfills containing organic wastes produce biogas containing methane (CH_4). Landfills are significant sources of methane, which contributes to climate changes. 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. Landfills may be covered with biological active materials, so-called biocovers. Experiments have documented that a very high methane oxidation rate can be obtained in bio-covers, high enough to significant reduce the methane emission from the landfill. Documentation of the efficiency of bio-covers has so far only been carried out in full scale in a few cases for instance the newly finalized project at Fakse Landfill where a new-developed protocol for biocover establishment and monitoring were presented (Scheutz et al., 2011a, b) and the second generation biocover system established at the Klintholm Landfill (Kjeldsen et al., 2013). Some of the lessons learned from these two full-scale biocover systems were that avoiding point releases of methane from the leachate collection system is very difficult. Beside, a major challenge in any biocover system is to obtain an even gas distribution to the active methane oxidation layer to avoid hot spot loading, which results in significant methane emissions.

AV Miljø is a modern waste disposal site situated in Avedøre Holme, approx. 10 km south of Copenhagen, Denmark. The disposal site was established in 1989 and has a total disposal capacity of 2 mill. m^3 divided into 22 disposal cells. The landfill receives waste from approx. 1.2 mill. inhabitants and 80,000 larger and smaller enterprises. Since 1997 it has been forbidden in Denmark to use landfills for disposal of combustible waste. AV Miljø therefore mostly deals with non-combustible waste, i.e., waste with low organic content such as, e.g., shredder waste, asbestos waste, contaminated soils, construction waste, residues from street cleaning, slag, and fly ashes from waste incineration. Previous studies have shown that significant quantities of landfill gas are produced at the landfill, where a considerable amount is emitted from the leachate collection system via inspection and collection wells, (Scheutz et al., 2011c, Fredenslund et al., 2010)). The planned study and establishment of the pilot scale biocover system was focused on the western part of the landfill where several gas leaking wells previously were identified.

The scope of the project was:

- To construct and get experience with a semi-passive biocover system fed by a gas load extracted from the existing leachate collection system for reducing methane emissions from landfills
- To give special emphasis on the gas distribution system to obtain an evenly distributed gas load for avoidance of hot spot emission areas on the biocover

- To quantify the efficiency of CH_4 oxidation in the biocover system. The CH_4 oxidation efficiency was determined for single points as well as for the whole biocover

2. Pre investigation

Prior to the establishment of the biocover system two rounds of pre investigations were carried out. The first round of field investigations focused on one of the waste cells at the AV Miljø Landfill intended for hosting the pilot-scale biocover test (Pedersen et al., 2010). The second round performed gas pump tests in several potential contributing leachate wells and made preliminary tests of potentially usable compost materials for the methane oxidation layer (Pedersen et al., 2011). A short summary of the latter report is given in this chapter.

2.1 Gas flow rates from leachate wells

Pump tests were carried out in six inspection/collection wells in the western part of the landfill. Each test had duration of about three days where a gas pump rate of 47 L/min extracted gas from the well. The accumulative pumped volume was monitored by a gas meter and the content of methane was measured continuously by a Photoacoustic Gas Monitor INNOVA 1412i (LumaSenseTechnologies, 2012). The results showed that especially three out of the six wells could maintain a significant and constant methane supply. The total extracted methane from the three wells was calculated to be 26 kgCH_4/day.

2.2 Testing methane oxidation capacities of compost

Two producers of garden waste compost were identified in the vicinity of the AV Miljø landfill, (Solum and RGS90). Representative sampling of the two compost types was carried out and batch test were performed to measure the respiration activity and the methane oxidation capability of the two composts. The tests were carried out in 500 mL containers each containing 100 grams of moist compost. For the respiration tests 120 mL of the head space air was exchanged with pure oxygen (O_2) and the decrease of oxygen and increase of carbon dioxide (CO_2) contained in the head space was monitored over a period of 250 hours. For the methane oxidation similar tests were set up. Here 30 grams of moist compost was used and 200 mL of the head space air was exchanged with 120 mL oxygen and 80 mL methane followed by monitoring of the gas composition of the head space for 100 hours. More details of material and methods are given in Kjeldsen et al. (2013).

The results of the batch incubation tests showed that the compost from RGS90 had the lowest respiration (xx-xx g O_2/g/d) and also the highest methane oxidation rate (xx-xx g CH_4/g/d), so the compost from RGS90 was chosen as the active methane oxidation medium for the biocover. Besides, the compost was also produced very close to the AV Miljø Landfill. Based on previous experiences from the Fakse and Klintholm biocover systems (Scheutz et al., 2011a, Kjeldsen et al., 2014), where both batch incubations, column experiments and in situ determination of me-

thane oxidation capacities of compost layers were carried out, a conservative expected in situ capacity of the RGS90 compost was set to 50g CH_4/(m^2 and day). With an expected methane load from extracting gas from the three most productive wells of 26 kgCH_4/day, the area of the pilot scale biocover system was set to 500m^2.

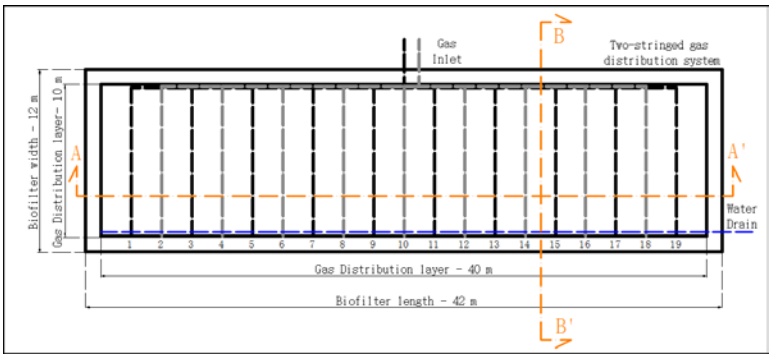
3. Overview of the biocover system

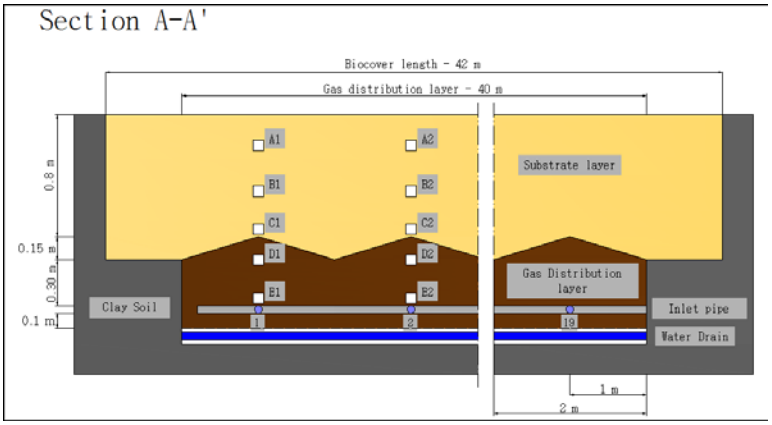
The biocover system (12 m x 42 m) consisted of a gas distribution layer (30 – 50 cm) overlain by a compost layer (80 – 90 cm). The biocover system was designed with the three supply wells mentioned above connected with underground piping to a mixing chamber where gas from the three wells was efficiently mixed.

The three landfill gas emitting leachate wells were enclosed in air tight sheds to improve gas extraction. To obtain a controlled loading to the biocover the landfill gas was pumped from the enclosed wells to the inlet mixing chamber of the biocover. The pumped gas, reached the inlet mixing chamber trough 3 PVC pipes(120 mm diameter). The cylindrical mixing chamber had an external and internal diameter respectively of 115 and 100 cm, respectively and was made of HDPE material. Pumping rates from each of the three sheds were continuously monitored and the content of methane in the mixing chamber was continuously monitored with a CH_4-sensor and data logger. Knowing the gas flow and the CH_4 content in the mixing chamber, the CH_4 load to the biocover could be determined. Other gases (O_2, CO_2 and nitrogen (N_2)) were monitored manually in gas tubes connected to the interior of the mixing chamber.

The interface between the methane oxidation layer (MOL) (consisting of compost) and the coarse gravel gas distribution layer (GDL) was ”zig-zag-shaped” to minimize continuous water locking due to capillary effects (which has been identified as a major problem in other biocover systems) – see Figure 3.1a. An unslotted gas pipe - distributing gas to 20 slotted gas pipes (see Figure 3.1b and c) - was equipped with small outlet holes to obtain an even gas distribution. A section of the GDL and the MOL was equipped with horizontal multi-port gas monitoring units to study gas distribution and MO processes (see section 4.7). Two transects each having 50 gas ports were constructed at two different distances from the gas inlet – see Figure 3.1b, c. The biogas system was equipped with a water draining system including a water lock to avoid gas bypass.

Figure 3.2 shows pictures taken during establishment of the biocover system. The system was constructed in the summer of 2012 with the gas load start up in September 2012. During the experimental period the biocover was gradually vegetated by different weeds, which at the end of the experimental period reached a height up to 150 cm. Figure 3.2 shows the biocover surface at different occasions during the experimental period. The vegetation was dominated by one single species of Chenopodium (in Danish Gåsefod), (Trapp, 2013).





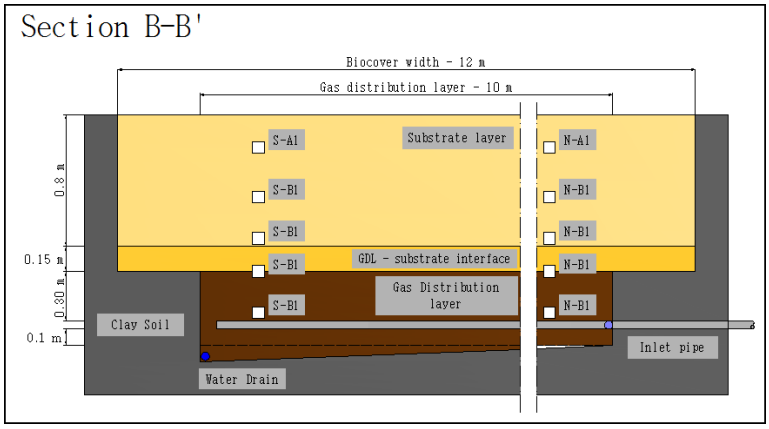


Figure 3.1. Plan view of biocover showing the gas distribution system with 19 slotted gas pipes connected to a delivery pipe. b) Sectional view of biocover showing the zig-zag shaped interface between MOL and GDL. c) Sectonal view of biocover showing water draining and gas distribution pipes. Locations of selected gas probes in the two transects are indicated in sections A-A' and B-B'.

Air tight shed on top of leachate well

Pump and control unit at well point

Biocover area during GCL construction

GDL with pipes and zig-zag shaped interface

Two-stringed GD system

Five multi port gas probes ready for installation in transect

Multi port gas probe units during installation in GDL

Multi port gas probe units installed in all depth

Manifold for sampling one transects 50 gas probes

The finalized biocover system

Figure 3.2. Pictures taken during construction of the biocover and establishment of the horizontal multi port gas probe units.

Figure 3.3 Pictures of the biocover at different times during the monitoring period.

4. Materials and Methods

4.1 Laboratory experiments: Methane oxidation and respiration

Two batch laboratory experiments have been carried out during the biocover study to evaluate compost respiration and methane oxidation capacity. Biocover compost was sampled at two places in November 2012 and April/May 2013 At the two occasions samples were taken in different depths and immediately sealed in tight plastic bags to avoid moisture evaporation. Sample locations are shown in Figure 4.1. First sample was taken close to T2, while the later samples were taken at 5.2 (see Figure 4.1).

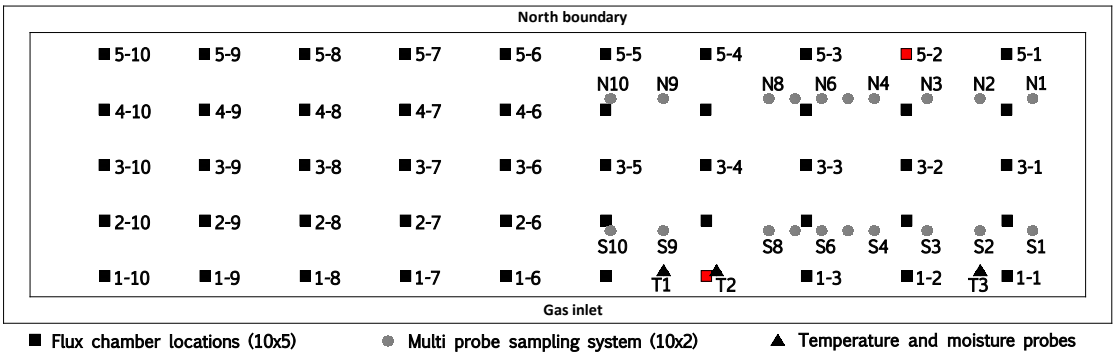


Figure 4.1 Map of the biocover with indications of locations of flux chamber measurements, the multi probe samplings system, the temperature/moisture probes and compost sampling sites.

For all batch experiments a standard experimental setup was used. Standard glass bottles (500 mL) were filled with 50g of compost, previously sieved to avoid inhomogeneities. Two types of compost were used for the study: pure compost from a certain depth and composite compost, obtained by manually mixing compost samples from different depths. The bottles were sealed with gas tight butyl rubber septa (Norsorex DC 97 rubber stoppers) and aluminium caps. Bottles intended to methane oxidation capacity evaluation were pre-treated overnight by injecting 80 mL of pure CH_4 to activate methanotrophic bacteria naturally present in the compost. Bottles were re-opened and flushed with pure air to avoid remaining CH_4. After the final sealing, gas was extracted from the bottles (120 mL for respiration bottles and 200 mL for methane oxidation bottles) and replaced with pure gases to created suitable conditions for microbial reactions. A starting O_2 concentration of about 30% volume (120 mL) has been chosen for compost respiration evaluation. Starting concentrations in bottles for methane oxidation evaluation were 12-14% volume (80 mL) for CH_4 and 32-35% volume (120 mL) for O_2. Gas samples were transferred every 12 hours from the bottles to standard vacuum 5mm 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 analysed by using a gas chromatograph (Agilent Technologies MicroGC 490). Measured concentrations were plotted with time and gas generation or consumption rates cal-

culated (O_2 and CO_2 for respiration test, CH_4 and CO_2 for methane 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 content was separately measured and generation or consumption rates normalized to dry mass (DM). For all experiment duplicates and blanks (bottle with injected gases but without compost addiction) were incubated to prove the results obtained.

The first methane oxidation batch test series started in November 2012 and had a duration of about 100 hours, while the first respiration test started in December 2012 and had a duration of 350 hours. Compost was sampled at different depths (-20cm, -40cm, -60cm, -80cm, -90cm) and incubated in three different experiments. In the first experiment, compost from each depth was incubated at the corresponding original temperatures measured during the sampling procedure (20°C, 30°C, 40°C, 55°C, 60°C). The purpose of this experiment was to measure the CH_4 oxidation and respiration potential of the compost at each depth and determine the optimal oxidation temperature. In the second experiment, all samples from all depths were incubated at the standard temperature of 20°C with the intent of identify the depth distribution of different types of bacteria. The third experiment consisted in the incubation of a mixed compost (obtained by mixing compost from all sampled depths) at eight different temperatures (4°C, 10°C, 15°C, 20°C, 30°C, 40°C, 55°C, 60°C) with the purpose of analysing the temperature dependence of CH_4 oxidation and respiration on a standard compost sample.

The second batch test series started in June 2013. The CH_4 oxidation test had a duration of about 25 hours, while the respiration test finished in 200 hours. For the methane oxidation and respiration experiments new compost samples were obtained in April 2013 at three depths in the biocover (-20 cm, -40 cm and –60 cm) in a different point from the first time (see Figure 4.1). Two different types of compost were then prepared and incubated (pure compost from -40 cm and a mixed compost from -20 cm, -40 cm and -60 cm) at two temperatures (30°C and 50°C). The main purpose of this experiment was to analyse how the methane oxidation and respiration potential of the biocover compost has changed from the freshly activated compost (first batch of November) to the consolidated bacterial community of the second batch (after 9 months of biocover operation). The same experimental procedure as for the first batch test series was used.

4.2 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 methane oxidation efficiency of the bacterial community and study the gas distribution quality in the system (confer section 4.8 for more details). In order to study the possible inhibitory effect of this tracer gas on methanotrophic bacteria pathway, two

laboratory experiments were carried out. For both experiments the same compost samples was used as for the second batch experiment series described in the previous section. Standard bottles (500 mL) were filled with 50g sieved compost and sealed with butyl rubber septa and aluminium caps as in previous batch tests. 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 pure air before the experimental procedure started. The same experimental procedure as described in the previous section was used.

The first tracer batch test started in April 2013 and had a duration of about 130 hours. The purpose of this experiment was to evaluate the eventual inhibitory influence of HFC-134a gas on bacterial activity. Compost was sampled at AV Miljø biocover in a range between -30 cm and -50 cm and incubated at 50°C to better control the methanotrophic activity. 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 methane oxidation batch tests. Five different quantities of tracer (HFC-134a) were then injected into the bottles to create five increasing mixing ratios (0 ppm, 8 ppm, 40 ppm, 90 ppm and 140 ppm).

The second tracer batch test was carried out jointly with the second methane oxidation test described in previous section. A duplicate for each bottle set was prepared and tracer was injected to obtain a standard concentration of 100 ppm. Methane oxidation rates from two compost samples (from -40 cm and a mixed compost) incubated at two temperatures (30°C and 50°C) with 12-14% volume or 18% volume of CH_4 were compared between bottles with and without tracer addiction. In this way the possibility of a differential oxidation rate in case of tracer presence was investigated. For both tests, duplicates and blanks were prepared as required from standard procedure as described previously.

4.3 Monitoring of gas supply to biocover

The gas pumping rate was continuously monitored at each of the air tight sheds confining the leachate wells by occasionally reading the accumulative gas meter (Flonidan, Denmark). The landfill gas composition in the inlet of the biocover was monitored continuously during the whole study period. An OLCT Infrared Transmitter Detector (Oldham, France) was installed in the mixing chamber and the methane percentage composition measured every 5 minutes. Data were recorded by using a GP-HR TruTrack outdoor data logger (Intech, New Zealand). Additionally, the major constituents of the gas were monitored in the mixing chamber once a week by using a Biogas 5000 portable gas analyzer (Geotech, UK). 100 mL of gas were extracted from the three inlet pipes to the mixing chamber and from the inlet pipe to the biofilter distribution system. The percentage composition of CH_4, CO_2, and O_2 was determined and manually recorded.

4.4 Moisture and temperature probes and measurements

Compost temperature and moisture content were monitored throughout the entire study period. Combined temperature and moisture probes (Five ECH2O EC-TM probes -Decagon Devices, USA) were installed in three different points and at three different depths (-20 cm, -50 cm, -75 cm or -95 cm) in the methane oxidation layer of the biocover. The location of the probes are shown in Figure 4.1.Three digital EM50 Decagon data loggers were connected to the 9 probes and programmed to measure the temperature (°C) and moisture content ($m^3 \cdot m^{-3}$VWC) every 5 minutes.

4.5 Surface screening of CH_4 and CO_2

A qualitative analysis of the spatial variability in surface emissions was carried out by observing the gas concentration of methane and carbon dioxide above ground surface. A grid was created to regularly divide the biocover area in 252 rectangular subareas of $2m^2$ each. Surface air concentrations were measured at the central spot of each subarea 10 cm above the soil surface. First, the methane surface concentration (ppm) was measured with a TVA1000B Photovac MicroFID analyzer (ThermoScientific, Waltham, USA). The surface was then screened with a Vaisala CARBOCAP® Infrared Sensor (Vaisala, USA) and CO_2 air concentrations (ppm) measured. All the screening campaigns took place during monitored weather conditions with low wind speed and stable barometric pressure to minimize spatial mixing of the sampled gas. A total of six measurement campaigns have been carried out during the whole study period. The screening was carried out on November 28th 2012, December 4th 2012, April 11th 2013, April 21st 2013, May 2nd, 2013 and May 24th 2013. Collected data were then processed by using Surfer 8 software (released by Golden Software Inc., Golden, USA) and interpolated with kriging statistical method to define iso-concentration curves.

4.6 Surface emissions of CH_4, CO_2 and tracer (HFC-134a)

The initial surface screening was followed up by a quantitative analysis of emissions from the biocover surface. With this methodology, the surface distribution of methane, carbon dioxide and tracer gas fluxes was evaluated. A series of 50 points equally distributed over the biocover surface was chosen for gas flux sampling – locations are shown in Figure 4.1. Surface emission rates were determined using static flux chambers. The flux chambers were made of stainless steel and equipped with sampling ports and a manually operated fan securing that the air inside the chamber was totally mixed during sampling. When installed, the flux chamber had a height of approximately 21 cm and an inner diameter of 30 cm (giving a total chamber volume of 15 L). The emission rates were measured by taking a time series of gas samples (1 sample per minute for minimum 5 min) from the chambers. An Innova 1312 photoacoustic multi gas monitor

(LumaSense Technologies A/S, Denmark) was used to measure concentrations of CH_4, CO_2, and tracer gas (HFC-134a). Concentrations and times of measurement were logged using a laptop pc connected to the instrument. Measurement ranges at the used configuration of the instrument were 0.4–20,000 ppmv (CH_4), 1.5–10,000 ppmv (CO_2) and 0.2-2000 ppmv (HFC-134a). In general, the concentration vs. time curves showed good linear fits ($r^2 > 0.9$) without any change in slope for the final sampling times. Fluxes were calculated from the product of the change in concentration over time (dC/dt) and the chamber volume/chamber area ratio (Scheutz et al., 2003, 2008). The detection limit of the flux chamber measurements were ±0.05 g m^{-2} d^{-1}. A spatial two-dimensional distribution of fluxes was obtained by processing calculated fluxes for each measuring day with Surfer 8 software.

4.7 Horizontal multi-port gas probes (HMPGP) – design and sampling

To evaluate the spatial vertical and horizontal distribution of loaded gas, a multi-port gas sampling system was designed. Two transects of 50 ports each were embedded in the biocover; one in the South-East part (3.5 m North of the Southern biocover boundary) of the biocover and the one in the North-East (8.5 m North of the Southern biocover boundary). Each port was designed to connect a predetermined point inside the biofilter to the outside, in order to make the sampling of the pore gas easier and avoid compaction of the compost layer by human activity. In each transect the ports were divided in five groups, each laid at a different depth in the biocover volume (-20 cm, -50 cm, -75 cm, -100 cm and -120 cm below ground surface). Each depth were labelled with different letter (A, B, C, D, E) with A being the shallowest one. In the end each port of this group of ten were placed at a different distance from the gas inlet point and labelled with a number. The ports were located at 18(#1), 16(#2), 14(#3), 12(#4), 11(#5), 10(#6), 9(#7), 8(#8), 4(#9), and 2(#10) meters from the biocover midline, as represented in Figure 3.1. As an example, the port labelled with N1A is located in the North-East part of the biofilter, 18 meters from the biocover midline and at a depth of 20 cm. The exact location (plan view) of each probe is also shown in Figure 4.1. Each tubing had an internal diameter of 2 mm giving an internal volume of 3.1 mL/m. Each bundle of 10 tubings were equipped every second meter with a gas blocking plastic plate to avoid horizontal transport of gas along the bundle.

Gas concentrations of CH_4, CO_2, O_2 (percentage) was determined using a Biogas 5000 portable gas analyzer (Geotech, Warwickshire, UK). The pump rate of the Biogas 5000 was approximately 550 mL/min, with an operating temperature range from -10°C to +50°C. The pump was set to run for 10 second, with a 30 seconds flushing prior to measurement. The operating range of the gas analyzer is between 0-100%vol for CH_4 and CO_2 and 0-25%vol for O_2. The detection accuracy is +/- 1%vol in the concentration rage of 0 to 25%vol. Samples for trace gas quantification (during the tracer release experiments – see section 4.8) were analyzed using the Innova

1312 photoacoustic multi gas monitor. Data were then processed with Surfer 8 software to obtain the vertical two-dimensional distribution of each gas for each of the transects defined by a multi-port sampling system. Data was also used for determination of vertical gas concentration profiles at selected locations.

4.8 Tracer release experiment

To evaluate the ability of the designed biocover system to evenly distribute the gas to the full footage area and to improve the determination of the biocover efficiency for methane oxidation, the injection of a tracer gas into the mixing chamber was performed. The tracer gas adopted for the field study is known scientifically as 1,1,1,2- Tetrafluoroethane or as $C_2H_2F_4$ (commercially as HFC-134a). The compound is regarded as persistent under anaerobic as well as aerobic conditions (Scheutz et al., 2004), and has a low hydrophobicity leading to a low retardation due to sorption by the compost material. A 20 L tedlar gas bag was refilled every 12h with pure HFC-134a from a pressure bottle. A constant flux of 0.2 mL/s was continuously pumped from the gas bag into the mixing chamber with the use of a high-precision peristaltic pump (Model, xx, xx). A constant flux of tracer guaranteed a stable concentration in the inlet of 80 ppm, constantly monitored during the whole experimental period. A flow-meter and flow-controller was also added between the peristaltic pump and the biocover mixing chamber to guarantee a constant influent tracer flux. The Photoacustic measuring device Innova 1412i (LumaSense Technologies, 2012) was calibrated for the analysis of CH_4, CO_2 and $C_2H_2F_4$ altogether, with a detection limit of 3.0 ppm, 5.1 ppm and 200 ppb, respectively. The inlet was tested with Innova 1412i twice a day to ensure a constant flux of tracer into the biocover system.

A selection of probes in the HMPGP was sampled and tracer concentrations determined using the Innova 1412i during the whole tracer study duration. The concentration of tracer gas as a function of time was plotted to study the tracer breakthrough curve at selected ports. A pre-test and two measuring campaigns were carried out using this methodology. A pre-test experiment took place on November 17th-18th, 2012 where gas was extracted from 18 ports (9 in the South transect and 9 in the North transect) over a period of 18 hours. The pre-test made a basis for creating a detailed testing plan for the following two studies. The first test was carried out in the period December 5th-9th, 2012 for a total duration of 67 h. During this test, 9 ports in the South transect and 9 in the North transect were repeatedly tested using Innova 1412i. The ports chosen were #1, #4, #8, respectively 18, 12 and 8 meters from the biocover gas inlet. Ports at three depths were tested for each distance (A, B, D) corresponding to -20 cm, -50 cm and -100 cm depths below ground surface. The second experiment setup was run in the period May 16th-27th, 2013 for a total duration of 262 h. As for the previous experiment, 9 ports were tested for each

transect, for a total of 18 ports. Tracer concentrations in time were measured for port #1, #7, #10, respectively 18, 9 and 2 meters from the biocover inlet. The same three depths (A, B, D) as in the first test were tested during this campaign. Tracer concentrations versus time were plotted to evaluate the tracer breakthrough in selected gas probes. At the end of the final tracer campaign, the surface flux of HFC-134a tracer was measured simultaneously with a normal CH_4/CO_2 surface flux campaigns (as described in section 4.6).

4.9 Single point CH_4 oxidation by the carbon mass balance method

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 is equal in the bottom of the compost cover. The method is based on the assumption of steady state and therefore assimilation of carbon by the methanotrophic bacteria is not accounted for (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 Equations 1 and 2 results in Equation 3, which was used to calculate the load of CH_4 entering the biofilter. Equation 4 was used to calculate the CH_4 oxidation rate.

$$J_{LFG,bottom} = J_{CO_2,surface} + J_{CH_4,surface} = J_{CO_2,bottom} + J_{CH_4,,botttom} \quad (1)$$

$$\frac{J_{CH_4,bottom}}{J_{CH_4,bottom} + J_{CO_2,bottom}} = \frac{C_{CH_4,bottom}}{C_{CH_4,bottom} + C_{CO_2,bottom}} \quad (2)$$

$$J_{CH\,4,bottom} = \frac{C_{CH\,4,bottom}}{C_{CH_4,bottom} - C_{CO_2,bottom}} \cdot \left(J_{CO_2,surface} + J_{CH_4,surface}\right) \quad (3)$$

$$R_{CH_4oxidation} = J_{CH_4,bottom} - J_{CH_4,surface} \quad (4)$$

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

4.10 Single point CH_4 oxidation by the tracer mass balance method

A new and innovative method for quantifying CH_4 oxidation was tested using injection of an inert gas inside the biocover body. The inert gas, called tracer, was mixed into the inlet flow of LFG. Tracer inlet and surface fluxes were measured (see section 4.8). The tracer 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 movement in the biocover has been reached. It is also assumed that the CH_4 and tracer gas diffuse with the same rate. The CH_4 oxidation efficiency can be determined by measuring the CH_4 to trace gas flux ratio in the inlet and at the biocover surface using the following equation:

$$MO_{Efficiency} = \frac{\left(\frac{J_{CH_4}}{J_{tracegas}}\right)_{Inlet} - \left(\frac{J_{CH_4}}{J_{tracegas}}\right)_{Surface}}{\left(\frac{J_{CH_4}}{J_{tracegas}}\right)_{Inlet}} \quad (5)$$

Tracer test using a Freon compound (HFC-134a) were conducted to quantify CH_4 oxidation. The tracer was continuously pumped at a controlled rate into a mixing chamber and distributed further to the gas distribution layer and the CH_4 oxidation layer (confer section 4.8 for more details on the performance of the tracer test). Tracer breakthrough curves in selected points of the horizontal multi-port gas probe sampling units were established by sampling and analyzing the tracer concentration over time in the duration of the tracer experiment. Also the tracer emission through the surface of the biocover at the end of the tracer experiment was measured in 50 locations by use of a mobile static flux chamber.

4.11 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 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 as described in section 4.3. Surface emissions of CH_4, CO_2 and trace gas were measured using stationary flux chambers (see section 4.6). In total, 6 campaigns were conducted in the period from January 2013 to May 2013. The tracer emissions were only measured at the end of the second tracer experiment on May 24th 2013.

5. Results and discussion

5.1 Methane oxidation and respiration of compost material

Table 5.1 shows the results regarding methane oxidation rates achieved during December 2012 (Batch 1) and May 2013 (Batch 2) laboratory experiments. Methane oxidation rates show an almost constant value over time for experiments made at 50°C. For both first and second batch, CH_4 oxidation rates range between about 57 and 79 $\mu gCH_4 \cdot gDM^{-1} \cdot h^{-1}$ oxidized. A consistent increase in oxidation rates was observed for the 30°C oxidation rates, increasing from about 24 to 123-131 $\mu gCH_4 \cdot gDM^{-1} \cdot h^{-1}$ oxidized over a period of 6 months. Methanotrophic bacteria seem to adapt faster to high temperatures, however oxidation rates are considerably lower at 50°C in comparison to 30°C in nearly all cases. At the same time the adaptation period is longer at 30°C. No considerable difference can be observed between methane oxidation capacity of a particular depth soil (-40 cm) and the mixed compost indicating that methanotrophic bacteria are equally distributed at all depths of the biocover.

Table 5.1 Methane oxidation rates for mixed (MIX) and depth specific (-40cm) compost samples determined in batch tests at two temperatures (30°C and 50°C) and with or without addition of tracer (HFC-134a).

Batch and compost	Initial CH_4 conc. (%)	CH_4 oxidation rate: No HFC-134a addition ($\mu gCH_4 \cdot gDM^{-1} \cdot h^{-1}$)		CH_4 oxidation rate: HFC-134a addition ($\mu gCH_4 \cdot gDM^{-1} \cdot h^{-1}$)	
		30°C	50°C	30°C	50°C
Batch 1 MIX (December 12)	14	24.60 ± 1.45	72.82 ± 4.66	-	-
Batch 2 MIX (May 13)	14	123.79 ± 3.85	57.53 ± 0.09	123.86	54.81
	18	131.60 ± 3.83	79.21 ± 0.61	130.71	72.49
Batch 2 -40cm (May 13)	14	126.20 ± 5.47	57.38 ± 5.91	125.91	58.08
	18	124.73 ± 6.08	71.62 ± 8.50	128.77	68.5

A wide range of CH_4 oxidation rates has been measured in a previous study (Scheutz et al., 2011c). Measured oxidation rate at 22°C ranges between 14.7 and 168.2 $\mu gCH_4 \cdot gDM^{-1} \cdot h^{-1}$ demonstrating a wide variability of the methanotrophic activity. These results are widely comparable with the one obtained in the study presented here with a methane oxidation efficiency at 30°C ranging between 24 and 131 $\mu gCH_4 \cdot gDM^{-1} \cdot h^{-1}$.

Data from compost respiration experiments showed a substantial CO_2 generation activity in the biocover compost. The CO_2 generation changes considerably with the environmental temperature. In the range of temperatures actually presents in the biocover compost (20 to 60°C) the CO_2production ranges between 8 and 50 $\mu gCO_2 \cdot gDM^{-1} \cdot h^{-1}$, with the maximum value reached for a temperature of 50°C (Table 5.2). Data from both December and June lab tests showed similar results for high temperatures (50°C) reporting a generation rate between 47 and 49 $\mu gCO_2 \cdot gDM^{-1} \cdot h^{-1}$. On the contrary at lower temperatures, CO_2 production showed an increased rate in May in comparison to December from about 5 to 15 $\mu gCO_2 \cdot gDM^{-1} \cdot h^{-1}$. This clear trend probably shows an instant adaptation of bacteria responsible of compost respiration to high temperatures and a longer lag phase for temperatures around 30°C. No relevant difference can be observed from the respiration rates obtained between compost from a specific depth (-40 cm) and the mixed soil showing an almost homogenous distribution of bacteria responsible of compost respiration.

Table 5.2 Results from compost respiration experiments carried out at different temperatures on mixed and depth-specific samples sampled both in December 2012 (Batch 1) and May 2013 (Batch 2).

Temperature	CO_2 generation rate: MIX Batch 2	CO_2 generation rate: -40cm Batch 2	CO_2 generation rate: MIX Batch 1
(°C)	($\mu gCO_2 \cdot gDM^{-1} \cdot h^{-1}$)	($\mu gCO_2 \cdot gDM^{-1} \cdot h^{-1}$)	($\mu gCO_2 \cdot gDM^{-1} \cdot h^{-1}$)
20	8.39	-	-
30	5.73	15.74	15.57
40	25.80	-	-
50	49.53	47.55	47.77
60	25.05	-	-

An increased CO_2 generation rate has been reported in other previous studies concerning methane oxidation in compost. Scheutz et al. (2011c) demonstrated how respiration rates can increase over time in case of high methanotrophic activity due to the accumulation of biomass and of easily degradable organic compounds in the compost. In this study, the average CO_2 production increased from about 4 to 8-30 $\mu gCO_2 \cdot gDM^{-1} \cdot h^{-1}$ over an experimental period of 8 months. Einola et al. (2008) found also increasing respiration activities over time in the active methane oxidation zone. This study showed an intensive CO_2 generation activity at 25°C in the first 5 cm of depths (location of the methane oxidation layer) with rates increasing from about 20 to 200-250 $\mu gCO_2 \cdot gDM^{-1} \cdot h^{-1}$ in the experimental period duration.

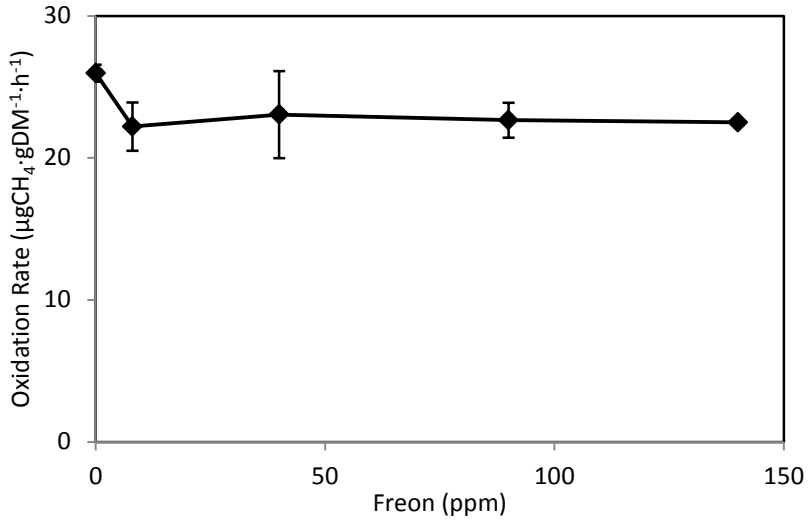


Figure 5.1 Methane oxidation rate determined in batch tests as a function of added Freon (HFC-134a) to the batches.

The result of the first experiment evaluating the potential inhibitory effect of the presence of HFC-134a to the methane oxidation activity is displayed in Figure 5.1. The figure shows an almost constant CH_4 oxidation rate for all the different HFC-134a concentrations tested. Rates were found to range between 20 and 25 $\mu gCH_4 \cdot gDM^{-1} \cdot h^{-1}$ and no noticeable influence was found in the bacteria performance due to the presence of HFC-134a. These results are also corroborated by the second tracer experiment performed in June and presented in Table5.1. Methane oxidation rates do not show any change depending on tracer presence, validating the results achieved in the previous test. This means that the performance of the tracer test in full scale in the biocover will not have any influence of the methane oxidation performance of the biocover during the tracer release.

5.2 Temperature and moisture conditions in biocover

To avoid water built-up into the boundary layer between compost and gravel material due to capillary forces a “zig-zag” shaped interface between the MOL and GDL was constructed. The efficiency of this system was tested by monitoring and comparing precipitation data from the local weather station installed at AV Miljø and the compost moisture data recorded by the probes installed at different depths and places. The comparison of these data (Figure 5.2) showed a direct correlation between relevant rain events and the local increase of the moisture content at all the depths. An increasing time delay between rain event and noticeable compost moisture increase was observed depending on depth (the higher the biocover depth, the higher the time delay). Between one relevant rain event and the following one, a constant decrease of the moisture content in the methane oxidation layer (MOL) was measured, demonstrating the efficiency

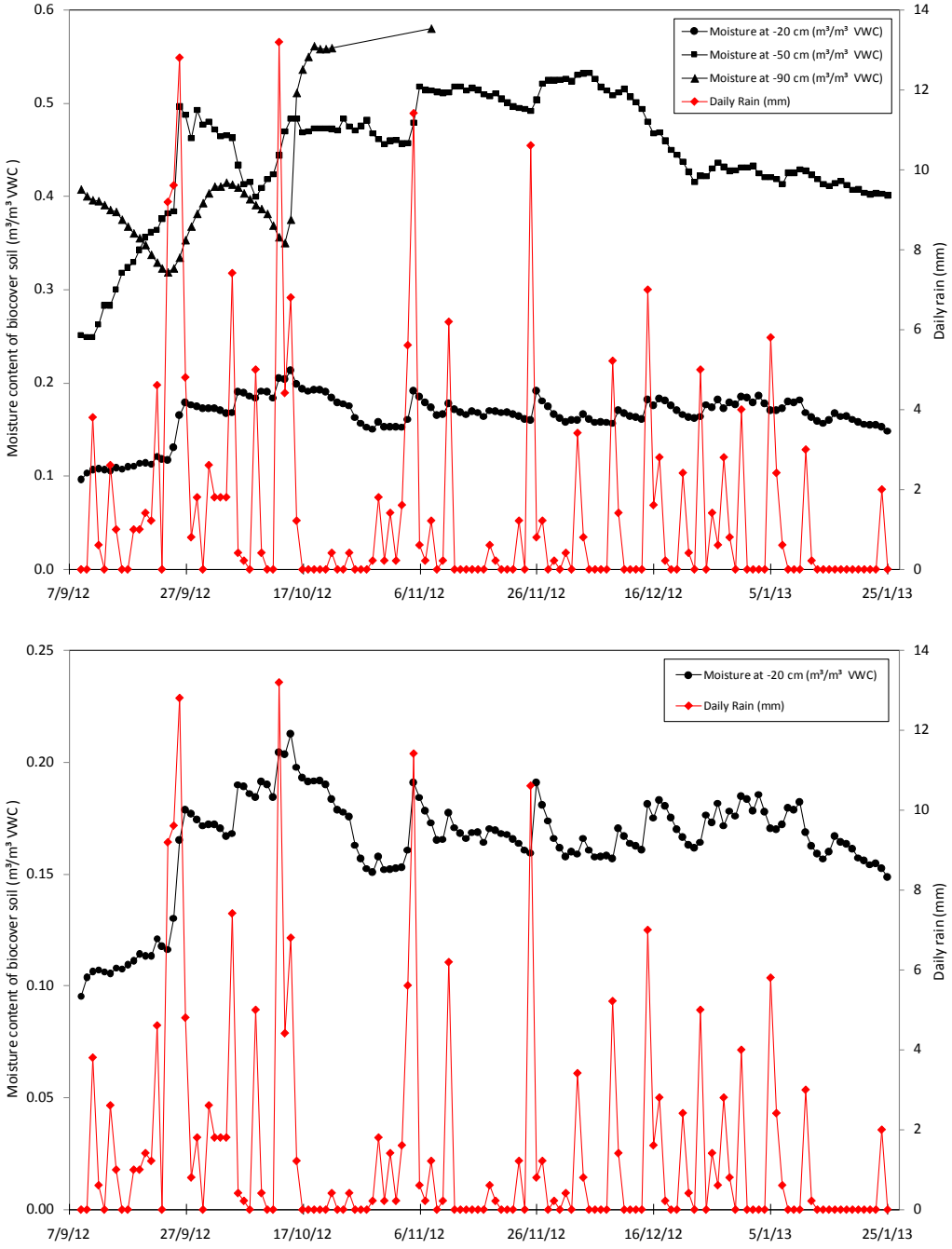


Figure 5.2 Moisture content (vol. %) in selected depths of the biocover.

of the “zig-zag shaped” system to avoid accumulation of water in the interface, which may lead to blockage of vertical gas transport. The removal of the exceeding water content from the biofilter system was further handled by the water drainage system installed, which led the excess water to the nearby sewer.

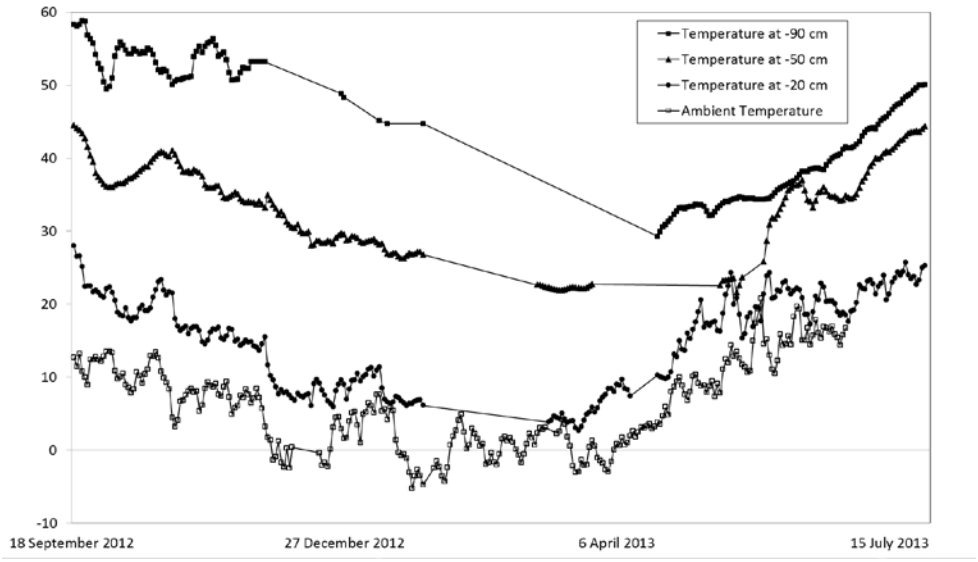


Figure 5.3 Temperatures measured in biocover at different depth in comparison to ambient temperature for the period 18 September 2012 until 15 July 2013.

Figure 5.3 shows the recorded temperatures in three different depths of the biocover over the entire monitoring period. Also the recorded ambient temperature is shown. The temperature is a very important environmental factor for microbial methane oxidation (Scheutz et al., 2009). The figure shows that significantly elevated temperatures are measured in the biocover. The highest temperatures are observed in the 90 cm depth with a typical temperature difference to ambient temperature of Δ50°C in the beginning of the period decreasing to about Δ25°C in the last period (April – July 2013). The temperatures are generally lower at more shallow depths, but the same trend of decreasing temperature differences over the experimental period (Δ30°C decreasing to Δ15°C for the 50cm depth, and Δ10°C decreasing to Δ5°C for the 20cm depth). The reason to the elevated temperatures must be the oxidation of methane and compost respiration (which both are exothermic reactions) in combination with heat transport from below (temperatures in the interior of the landfill have not been measured but is expected to be much higher than normal soil temperatures (Coccia et al., 2013)). The observed gradually decreasing temperature difference in all three depths is probably due to a lower heat generation from the continuously maturing compost during the ten month period. Similar observations were done at the Klintholm biocover system (Kjeldsen et al., 2014).

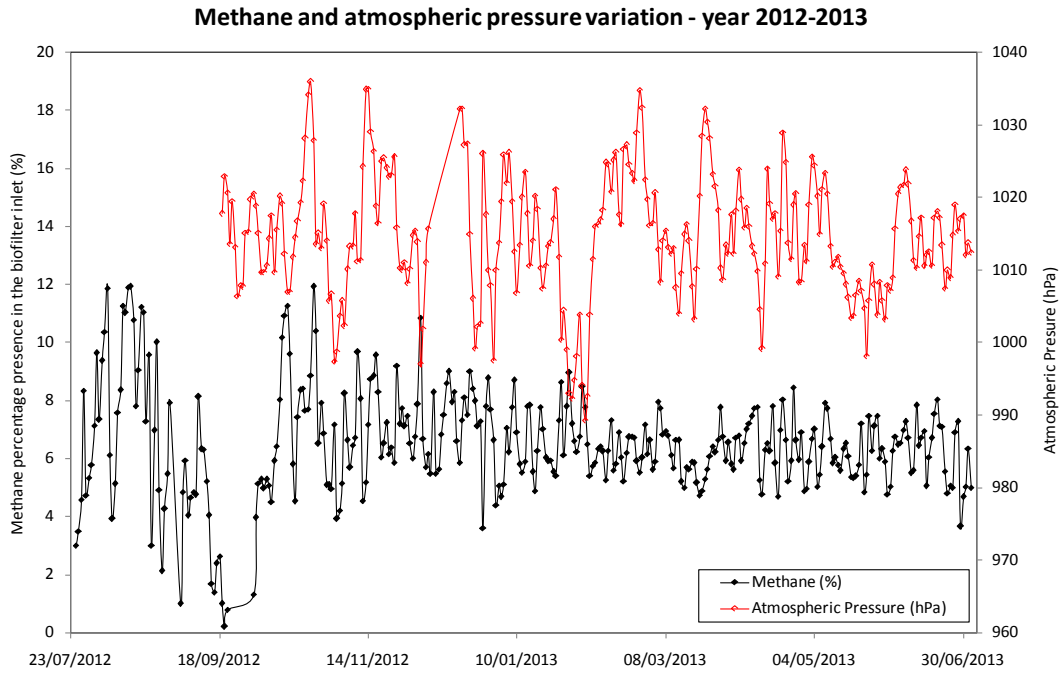


Figure 5.4. Concentration of CH_4 (in %) in the mixing chamber during the experimental period. Also the recorded barometric pressure (in hPa) is shown.

5.3 Gas load to biocover

The three pumps controlling the gas flow to the mixing chamber gave very constant gas loading of totally 182 m^3/day (data not shown). Figure 5.4 shows the CH_4 concentration in the mixing chamber for both 2012 and 2013. On the figure the recorded barometric pressure is also shown. The figure shows that the CH_4 concentration is fluctuating over time with typical concentration in the range of 3-12 %(vol.). The fluctuations seemed to some extent to be correlated with barometric pressure changes, however there must also be other factors controlling the CH_4 content for instance the pressure history some period back in time. . As previously demonstrated in other studies (Christophersen and Kjeldsen, 2001; Czepiel et al., 2003; Fredenslund et al., 2010; Kjeldsen and Scheutz, 2011) an inverse correlation can be observed between the two factors which identify in the atmospheric pressure the most significant influence factor on methane emissions from landfill bodies.

As a result of the presented observation it is clear that the methane load was fluctuating over time. Table 5.3 shows the manually measured gas composition in the mixing chamber. It is obvious that the leachate collection system had some openings to the atmosphere where atmospheric air was intruding, leading to oxygen content in the gas in the range of 5-13%(vol.). The implication of this was, however, that the MOL was loaded with oxygen not only by diffusion from the top but also due to the content in the loaded gas mixture. From the total pump rate and the methane content the typical CH_4 load can be deducted. Assuming a CH_4 content of 10

%(vol.), the load is 13 kgCH_4/day which is about 50% of the estimated load from the short time test of the three participating leachate wells (see section 2.1). This may be due to a larger dilution in the leachate collection system over the longer time period.

Table 5.3. Gas composition in mixing chamber measured manually by IR device.

Date	IR Device -Biogas 5000				Sensor
	CH_4	CO_2	O_2	N_2	CH_4
30/07 2012	4.8	5.1	12.9	77.2	4.2
02/08 2012	10.6	7.8	8.1	73.5	9.6
03/08 2012	6.9	6.2	10.5	76.4	5.7
07/08 2012	6.3	5.4	12.8	75.5	5.7
07/09 2012	6.3	7.0	11.5	75.2	4.0
19/09 2012	2.0	3.3	17.2	77.5	1.3
23/09 2012	1.2	2.2	17.9	78.7	1.3
26/09 2012	2.8	4.3	13.7	79.2	1.3
28/09 2012	0.7	2.7	16.9	79.7	1.3
03/10 2012	6.6	6.9	12.2	74.3	5.2
10/10 2012	8.2	6.5	11.5	73.8	7.1
16/10 2012	6.5	5.7	13.4	74.4	10.2
24/10 2012	12.8	10.7	5.4	71.1	12.3
31/10 2012	5.3	5.4	14.0	75.3	4.4
07/11 2012	6.0	5.5	13.4	75.1	5.7
14/11 2012	7.9	7.3	10.0	74.8	7.2
20/11 2012	7.7	6.7	10.7	74.9	6.5
28/11 2012	8.8	7.6	11.0	72.6	7.1
08/12 2012	6.8	5.9	12.4	74.9	5.5
11/03 2013	10.7	8.0	8.9	72.4	6.1
09/04 2013	8.6	6.5	12.7	72.2	7.2
11/04 2013	9.6	7.0	10.2	73.2	7.8
18/04 2013	5.8	5.0	16.2	75.3	6.9
02/05 2013	6.8	6.7	11.0	75.5	5.9
24/05 2013	6.8	7.5	10.5	75.2	5.4
Average	6.7	6.1	12.2	76.1	5.8

5.4 Surface screening of CH_4 and CO_2

Screening campaigns performed on the biocover surface for both CO_2 and CH_4 showed an even distribution of the surface gas emissions. Figure 5.5 reports the iso-concentration plots obtained with Surfer 8 software for CH_4 during four screenings performed in different seasons and periods of the study. Very low methane concentrations are present all over the biocover surface in an average range of 2-3 ppm, close to the background atmospheric concentration (1.7 ppm). A modest emission hot spot has been identified in the South-East corner during the

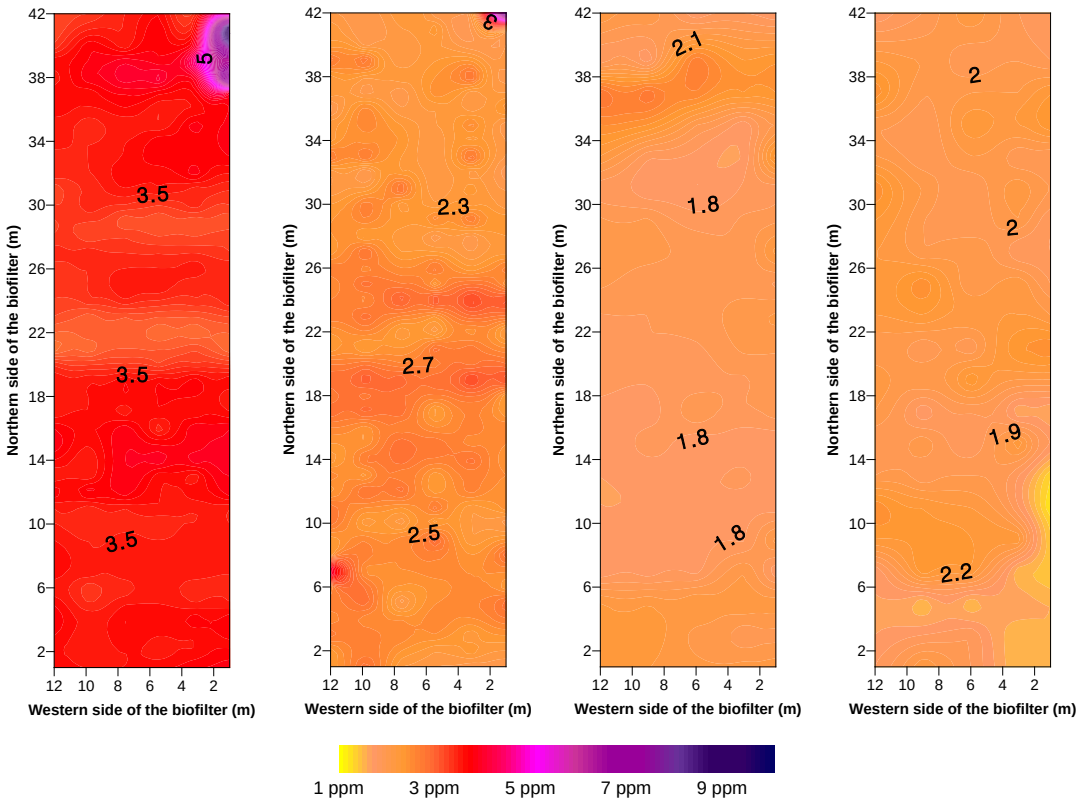


Figure 5.5: Iso-concentration plots for CH_4 on the biocover surface from 4 different screening campaigns, carried out on November 28th , 2012; December 4th , 2012; April 11th , 2013; May 5th 2013. Concentrations are expressed in (ppm).

first campaign (9 ppm) and also in the following ones (5 ppm). The hot spot has disappeared in following screenings and has not been measures again from January 2013 on. The surface CO_2 screenings presented in Figure 5.6 show an almost homogeneous distribution, close to the atmospheric concentration (400 ppm). The average measured values ranged between 400 and 420, with a general difference between South and North surface of 10 ppm. Several CO_2 hot spots were found on the biocover surface in different places from time to time, but no stable and substantial hot spots have been identified during the whole study. In general the two screenings together showed that we had succeeded in avoiding formation of significant hot spot areas probably as a result of a good distribution of the gas from the mixing chamber into the GDL and further up in the MOL.

5.5 Surface fluxes of CH_4, CO_2 and tracer (HFC-134a)

Both CH_4 and CO_2 iso-flux surface plots support the previous observations. For all flux chamber measurement campaigns, CH_4 surface emissions show a homogeneous spatial distribution. Examples of the iso-flux distribution for CH_4 are represented in Figure 5.7 - graph 1a-1b. Graph 1a refers to the January 25th campaign and shows an average surface flux of 0.05 $gC \cdot m^{-2} \cdot d^{-1}$, while

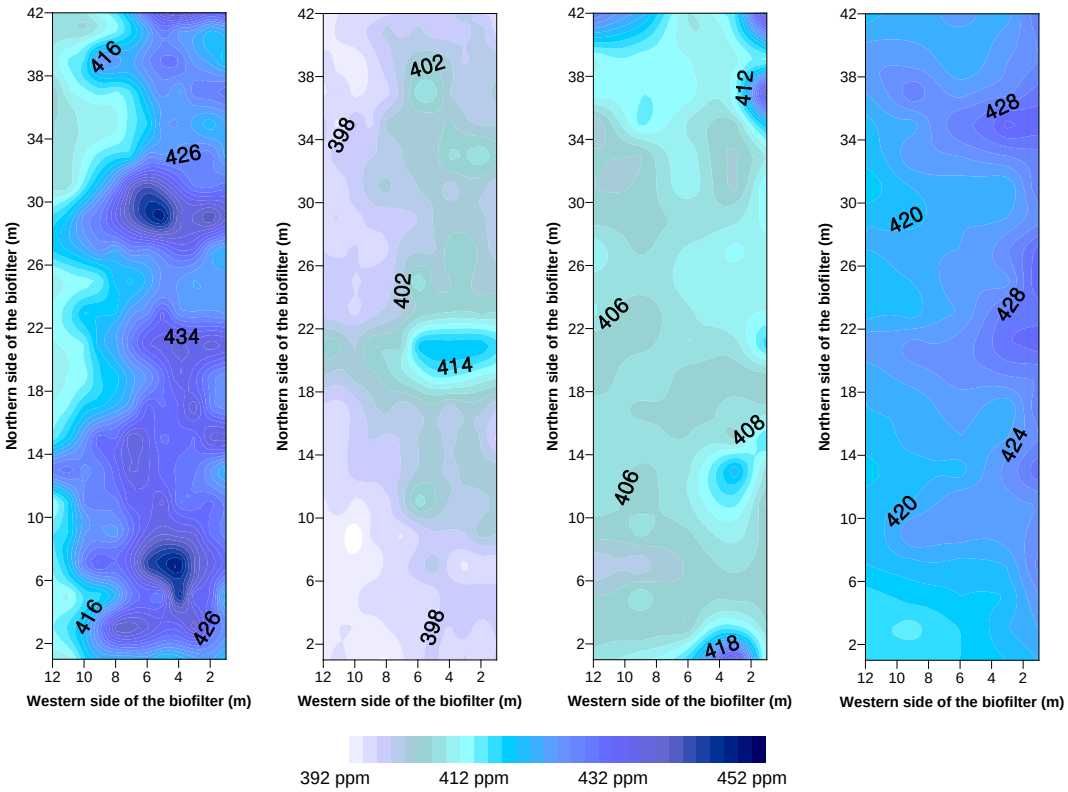


Figure 5.6: Iso-concentration plots for CO_2 on the biocover surface from 4 different screening campaigns, carried out on (a) November 28th , 2012, (b) December 4th , 2012, (c) April 11th , 2013 and (d) May 5th , 2013. Concentrations are expressed in (ppm).

the second one refers to the May 24th flux chamber campaign and shows an average flux of 0.6 $gC \cdot m^{-2} \cdot d^{-1}$. Also CO_2 surface fluxes indicate a modest spatial variation, with an average of 50 $gC \cdot m^{-2} \cdot d^{-1}$ for both January 25th and May 24th field measurement (Graphs 2a and 2b). Hot spots have been found on the biocover surface in different points from time to time. The presence of these preferential emission points is probably due to changing gas concentrations inside the biocover and to other environmental factors. On the other hand, the absence of constant hot spots through all the flux chamber campaigns demonstrates the homogeneity of emissions and the efficient design of the biocover.

The surface plot of tracer gas fluxes could be considered a precise picture of the distribution quality of the biocover system, and particularly of the GDL efficiency. Graph 3b in Figure 5.7 shows the HFC-134a distribution (May 24th campaign), indicating a modest spatial variation of tracer gas. The average range of tracer emission fluxes was found to be between 0.2 and 0.5 $gC_2H_2F_4 \cdot m^{-2} \cdot d^{-1}$, showing good distribution and homogeneous surface emission of gas. By using kriging interpolation of the 50 tracer emission measurements over the entire biocover area using the Surfer 8 software, the whole emission of tracer was found to be 70.8 $gC_2H_2F_4 \cdot d^{-1}$, which is

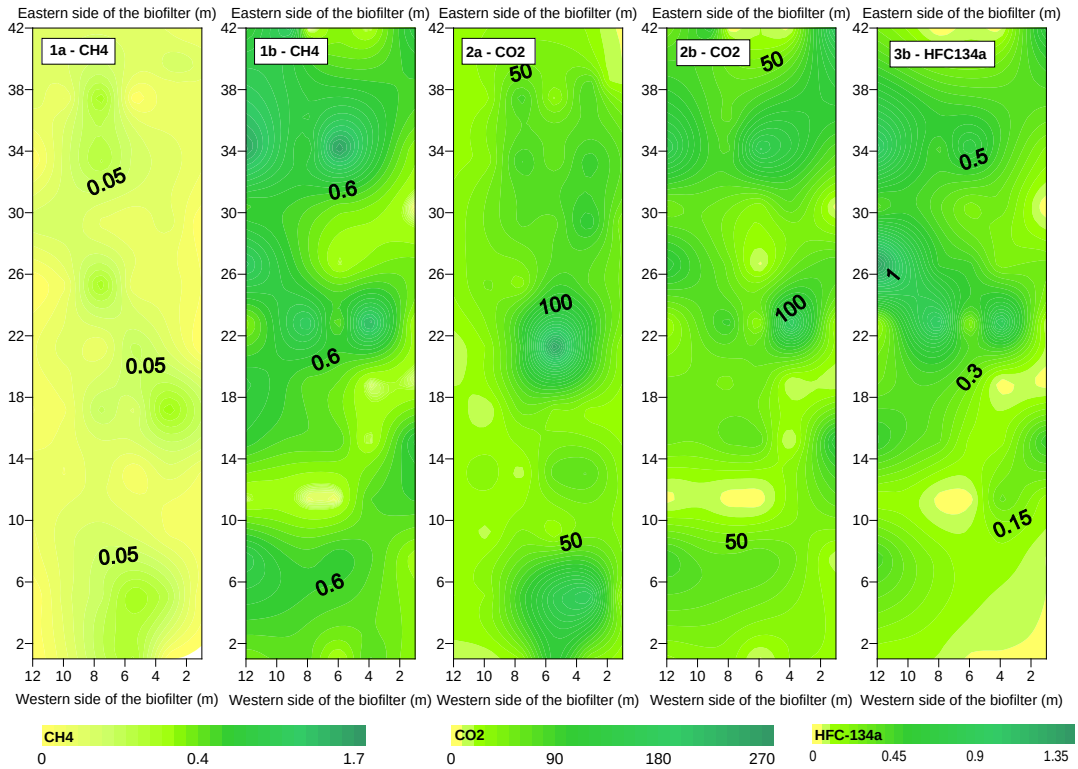


Figure 5.7: Iso-flux plots for CH_4 (1), CO_2 (2) and HFC-134a (3) on the biocover surface from 2 different flux chamber campaigns, carried out on (a) January 25th and (b) May 24th. Surface fluxes are expressed in $gC \cdot m^{-2} \cdot d^{-1}$ for CH_4 and CO_2 and in $gC_2H_2F_4 \cdot m^{-2} \cdot d^{-1}$ for the tracer (HFC-134a).

about 105% of the tracer loading rate (67.5 $gC_2H_2F_4 \cdot d^{-1}$). This shows that the tracer experiment had reached stationarity at the 12 day duration of the test, and that all loaded gas was finally leaving the biocover system as surface emissions.

5.6 Spatial evaluation of pore gas composition in biocover

The differential composition of the inlet gas could theoretically effect the biocover system stability and efficiency. Vertical 2D gas profiles of the biocover East sector showed an almost constant distribution and concentration of the analysed gases (CH_4, CO_2 and O_2) even with different inlet gas and atmospheric pressure conditions. The average percentage concentration of studied gases (CH_4, O_2 and CO_2) into the compost pore volume is represented in Figure 5.8. Methane and oxygen vertical profiles show the same trend with higher concentrations close to the GDL and lower close to the biocover surface. The bottom concentration of CH_4 ranges between 3 and 5%, with a final surface concentration after methanotrophic bacteria activity of about 0%. Oxygen is also consumed both during the compost respiration process and the methane oxidation with a decrease from 6% at 120 cm depth to 1% in 20 cm depth. The oxygen presence and homogeneous distribution in the biocover is of the most important factors since

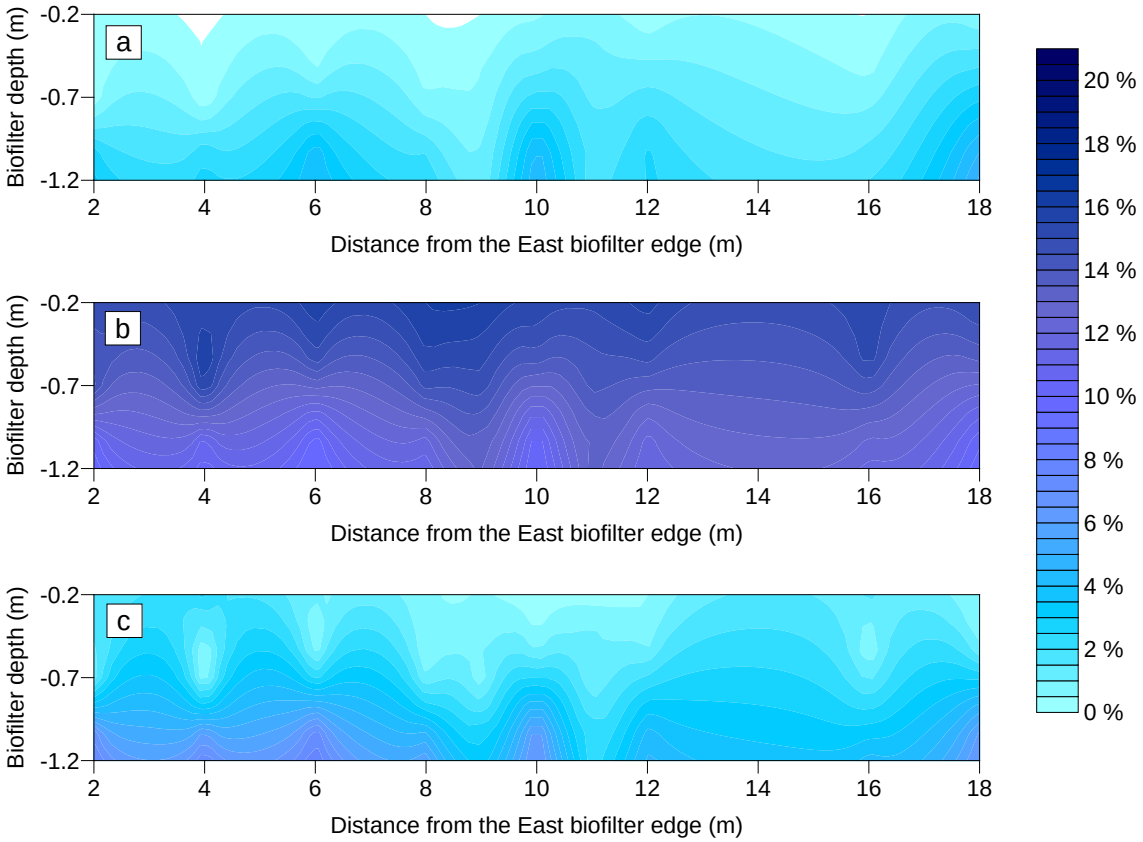


Figure 5.8 Gas composition (a: CH_4, b: CO_2, c: O_2) in the pore gas of the biocover as a function of biocover depth and location from east biofilter edge as measured in the southern horizontal multi gas probe system.

oxygen is normally the limiting factor for methanotrophic processes. Since the inlet gas contains significant quantities of oxygen the biocover is not only loaded by oxygen diffusion from the top but also by the inlet gas itself. This is different from traditional biocover systems which are loaded by a more pure LFG not containing O_2. In the end CO_2 percentage distribution increase from the bottom to the top of the biocover MOL with a starting 12% concentration which reaches 18-20% close to the surface. This indicate that methane oxidation is taking place at the bottom of the MOL and probably already in the GDL. The general trend of the three gases clearly shows that methanotrophic bacteria are active in the system and that CH_4 and O_2 are consumed and CO_2 is generated.

5.7 Evaluation of gas distribution by the tracer experiment

The HFC-134a concentration in the mixing chamber was constantly monitored during the whole measurement campaign duration to guarantee a constant flux of the tracer in the biocover distribution system and a constant tracer concentration (about 82 ppm). The concentration of tracer (HFC-134a) in the mixing chamber is represented by the red line in both Figure 5.9 and

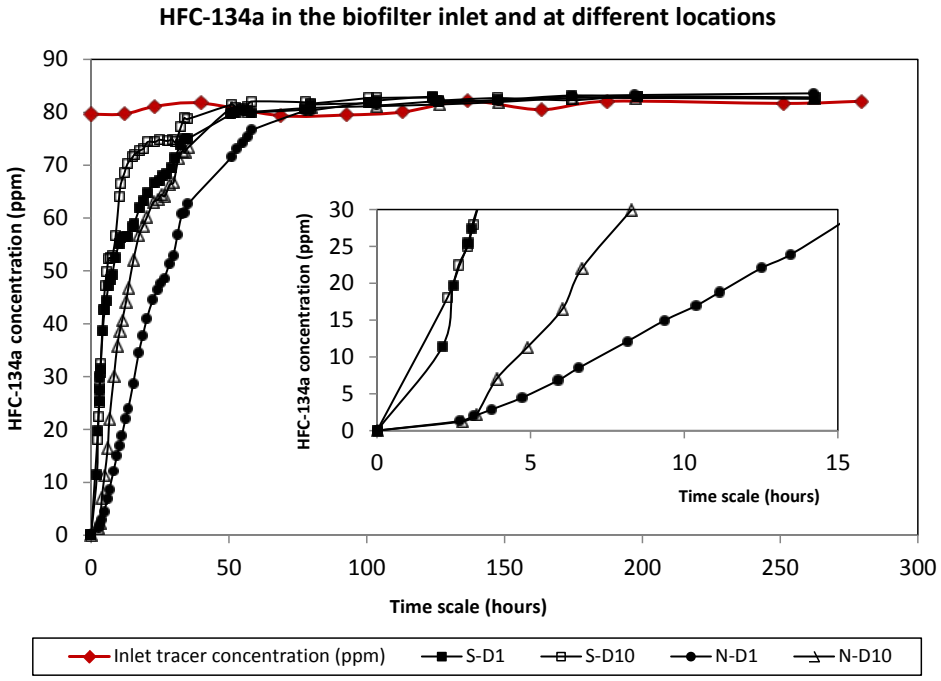


Figure 5.9. Concentrations of tracer (HFC-134a) in the mixing chamber representing influent and in gas probes placed in the gas distribution layer at different locations in the south and north transects.

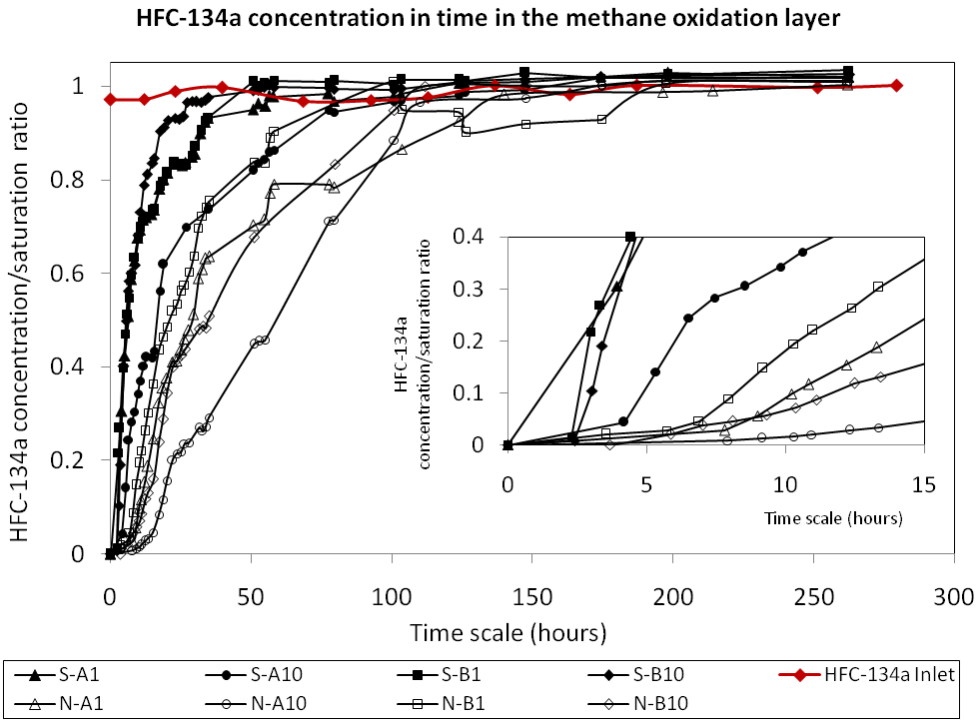


Figure 5.10. Concentrations of tracer (HFC-134a) in the mixing chamber representing influent and in gas probes placed in the methane oxidation layer at different locations in the south and north transects.

5.10 and shows a stable and constant trend of the gas, with natural fluctuations between 80 and 84 ppm (0.97-1.03 standardized).

The breakthrough curve of the tracer gas for each probe has been drawn by plotting HFC-134a concentrations (ppm) and time (hours). The standardized concentration (from 0 to 1) has then been calculated by dividing the point concentrations for the maximum concentration at saturation (82 ppm). Data collected from the two tracer injection campaigns and plotted as breakthrough curve gave interesting results regarding the gas distribution in the biocover.

During the first measurement campaign (December 5^{th}-9^{th} , 2012) the stationary concentration of 82 ppm were reached for all the probes installed in the South part of the biocover. The stationary concentration level was achieved for all South probes between 50 and 100 hours from the initiation of HCF-134a injection. The stationary concentration level was also reached in the North part of the biocover for D (-100 cm deep) and B (-50 cm deep) probes but never for probes A (-20 cm) due to the premature termination of the campaign. During the second measurement campaign (May 16^{th}-27^{th}, 2013) tracer stationarity was reached in both South and North parts of the biocover at all depths. Probes placed in South at all depths (GDL and MOL) and probes placed in the North GDL reached a stable concentration level between 50 and 100 hours from the injection start (Figure 5.9 and 5.10). Probes placed in the North biocover MOL reached a stationary conformation between 100 and 200 hours after tracer injection start (Figure 5.10).

In general, the tracer experiments gave steep breakthrough curves with similar breakthrough times of probes from the south transect independent of the distance from the inlet point. This shows an efficient and even delivery of gas in the un-slotted gas delivery pipe and the first part of the slotted gas distribution pipes. Breakthrough curves in probes from the north probe transect showed, however, longer breakthrough times indicating a slower distribution of gas to the northern part of the biocover.

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

Figure 5.11 shows representative gas composition profiles measured at locations from both the Southern and Northern transects. 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 contains significant concentrations of oxygen.

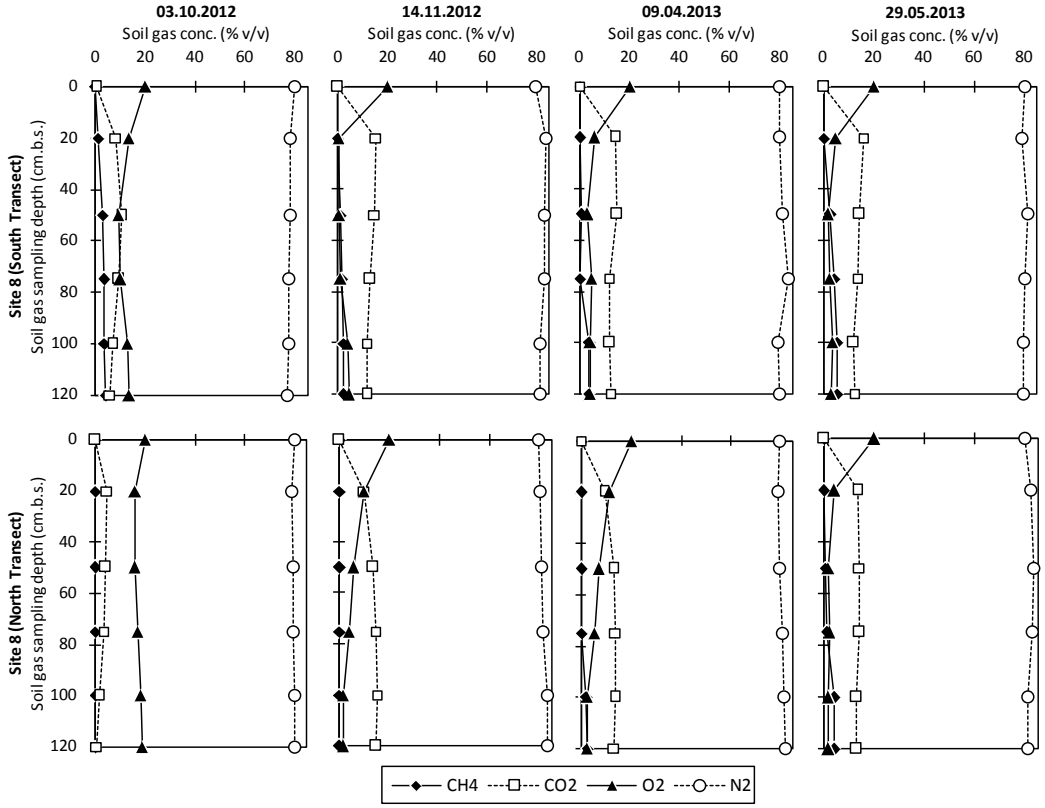


Figure 5.11 Gas profiles for CH4, CO2, O2 and N2 in selected locations and times.

Methane oxidation in the MOL can also be demonstrated by the vertical distribution of the ratio between the concentration of CH_4 and CO_2 with the concentration of HFC-134a showed in Figure 5.12. The profiles are measured after stationarity has been reached. Profiles represented in Figure 5.12 shows clearly how CH_4 and CO_2 change in the biocover volume due to methanotrophic activity. The ratio $CH_4/C_2H_2F_4$ shows clearly a decreasing trend from the bottom to the upper surface of the biocover demonstrating that CH_4 has been oxidized during its flow through the biocover pores. At the same time the ratio $CO_2/C_2H_2F_4$ describe an increasing trend during its flow through the biocover. In this case the CO_2 production from methane oxidation phenomenon is not the only reason of the ratio variation because compost respiration is also active in the biocover.

Table 5.4 compares CH_4 oxidation efficiencies and rates obtained by using the carbon mass balance method (see section 4.9) and the trace gas method (see section 4.10) performed in 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%. 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

Table 5.4. Comparison of CH_4 oxidation efficiencies and CH_4 oxidation rates obtained using two different methods; the carbon mass balance method and the trace gas 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

CMB: carbon mass balance; TMB: tracer mass balance; - 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%.

emitted CO_2 is be due to compost respiration. Considering CO_2 from compost respiration in the carbon mass balance will reduce the CH_4 oxidation efficiency further. 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}. 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 14 g m^2 d^{-1} (see next section).

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

As shown in section 5.5 very low CH_4 fluxes were recorded in general varying between negative emissions (-0.006 mol m^{-2} d^{-1}) indicating atmospheric uptake to maximum CH_4 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. Table 5.5 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. Overall, the CH_4 oxidation efficiency of the biocover varied between 81 and 100% - in most cases over 94% - and showed that the pilot plant biocover 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}. The average biocover performance was estimated to be around 14 g m^2 d^{-1}, which is relatively low in comparison to previous biocover studies. Considering the high CH_4 oxidation efficiency it is likely the biocover is over dimensioned and could actually oxidize a higher CH_4 load if needed.

The laboratory-measured methane oxidation potential as function of temperatures can be used to estimate the whole biocovers methane oxidation potential (assuming that no oxygen limitation of the methane oxidation is occurring in the field). The biocover MOL was divided in five horizontal layers (20cm each) and temperature homogeneity assumed for each stratum. Reference temperatures have been chosen based on data from probes installed in the biocover soil (as described in section 5.2). Then, the maximal CH_4 oxidation potential of the biocover was calculated by considering the methanotrophic oxidation rate measured during the three laboratory studies for each reference temperature (results reported in section 5.1). The specific CH_4 oxidation potential was calculated in this way to be around 300 $gCH_4 \cdot m^{-2} \cdot d^{-1}$. The results of the field measurement campaigns showed significantly different results. The average specific biocover performance was as previous mentioned calculated to be between 18 and 23 $gCH_4 \cdot m^{-2} \cdot d^{-1}$, but peaks of noticeably higher value where measured in different positions from time to time (as reported in the previous section). It is reasonable to conclude that the maximal potential of the biocover is several times higher than the average performance calculated with the whole-biocover campaigns. This is both shown from theoretical (lab experiments) and field (point flux chambers). The reason for the specific biocover performance of 18-23 $gCH_4 \cdot m^{-2} \cdot d^{-1}$ is probably that the load of CH_4 injected in the biocover is too low compared to the biocover oxidation potential.

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. High efficiencies have been found also during low ambient temperatures, which in previous studies has brought the methanotrophic activity to a stagnation (Stern at al., 2007). Reasons for the substantial efficiency of the biocover are the optimal distribution of O_2 at every depth, and the homogeneous gas distribution in the system due to the good design of the GDL. Moreover, bacterial growth was constantly guaranteed by the presence of the optimal temperature for methanotrophs. Previous studies reported optimal temperature for methanotrophic bacteria the range between 25°-30°C (Scheutz et al., 2011) and between 30°-40° (Streese and Stegmann, 2003). Gas profiles (Figure 5.11) showed that the maximal CH_4 oxidation activity was measured between -40cm and -90cm, where the optimal temperature is located and where also O_2 was present.

Table 5.5. Overview of CH_4 oxidation efficiencies and CH_4 oxidation rates obtained during six measuring campaigns performed from January to May 2013.

Date	Inlet load		Surface emission		CH_4 oxidation rate		CH_4 oxidation efficiency
	$gC\text{-}CH_4d^{-1}$	$gC\text{-}CO_2d^{-1}$	$gC\text{-}CH_4d^{-1}$	$gC\text{-}CO_2d^{-1}$	$gC\text{-}CH_4\ d^{-1}$	$gCH_4\ m^2d^{-1}$	%
25.01.13	6.930	6.121	19	25.007	6.911	18.3	100
08.03.13	7.732	5.370	332	88.386	7.516	15.0	96
11.04.13	8.066	4.537	1.541	77.623	6.526	13.0	81
21.04.13	7.020	6.265	35	36.277	6.985	14.0	100
02.05.13	6.396	7.242	30	39.489	6.366	12.7	100
24.05.13	4.046	3.760	246	28.219	3.800	7.6	94

5.10 Evaluation of compost respiration by biocover carbon balance

The Mass Balance application as shown in Table 5.5 proved that the total emitted Carbon ($C\text{-}CH_4$ and $C\text{-}CO_2$) from the biocover surface was considerably higher than the Carbon load into the system. Considering the CO_2 load in the inlet (about 6-7 $kgC\cdot d^{-1}$) and the CO_2 generated from methanotrophic activity (about 6-7 $kgC\cdot d^{-1}$), the additional CO_2 is probably produced from respiration of the compost. The average estimated carbon flux contributed by compost respiration activity based on field measurements was between 11 and 20 $kgC\cdot d^{-1}$. However, during the 2nd and the 3rd campaign a considerably higher $C\text{-}CO_2$ flux was estimated (about 40 $kgC\cdot d^{-1}$) due to the presence of exceptionally high emission hot spots.

The respiration potential of the biocover compost was estimated using the laboratory data previously presented. The MOL was divided in 5 horizontal parallel layers and for each layer a representative temperature was assumed. Data collected from Temperature-Moisture content probes (confer section 5.2) installed at different depths in the biocover MOL was analysed. A flexible temperature range was adopted based on the corresponding probes data to simulate the changing seasonal temperature trend. Respiration rates calculated with the two laboratory experiments have been used (as presented in section 5.1). 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 increased from 10-18 $kgC\cdot d^{-1}$ in January to 17-33 $kgC\cdot d^{-1}$ in the end of May, which is in the same range as the previously reported average estimated carbon flux contributed by compost respiration activity based on field measurements (11-20 $kgC\cdot d^{-1}$). Only the 2nd and the 3rd measuring campaign showed field results 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.

6. Conclusion and perspectives

Newer landfills are often receiving waste with lower organic content leading to a lower, but still significant gas generation. The landfills are often lined and not equipped with a gas extraction system. In such cases the generated gas is often emitted to a large extent through the leachate wells directly into the atmosphere. In order to evaluate the possibility of mitigating the gas emission by a semi-passive system where gas is collected from leachate wells and fed into an established biocover system where the methane contained in the collected gas is microbially oxidized, an innovative pilot-scale biocover system was established at the AV Miljø landfill.

Very even gas distribution was achieved in a 500 m^2 biocover system by use of a new-developed gas distribution system. Even gas distribution is a prerequisite for obtaining an efficient biocover without the formation of emission hot spots. The even gas distribution was documented by methane and carbon dioxide concentration screenings at the biocover surface and by the performance of a tracer experiment showing equal travel times in different parts of the gas distribution system and even distributed emissions of tracer over the biocover area.

By installation of moisture and temperature probes it was documented that significantly elevated temperatures were achieved in the biocover due to self-heating methane oxidation and compost respiration processes, which were able to maintain a high efficiency of the biocover system (higher than 95%) in the cold season. Moisture content logging in several depth of the biocover system documented that the system was efficient in discharging excess precipitation. I all depth a sudden increase in moisture content followed larger rain events, but decreased gradually after the rain had stopped.

Methane balances of the biocover by measuring the methane load to the biocover system and the emitted methane though the biocover surface (through integration of flux measurements performed in over fifty point on the biocover surface) showed that the system had a very high methane oxidation efficiency independent of the time of the year. – in most cases close to 100% efficiency. The active methane oxidation process was further documented by point measurement of the methane oxidation capacity using a carbon balance approach and a new-developed method using information about methane and tracer fluxes. The methane load to the biocover system was in generally lower than anticipated and resulted in relative low methane oxidation rates – on an absolute scale (from between 18 and 23 $gCH_4 \cdot m^{-2} \cdot d^{-1}$)

The active methane oxidation process was also documented through the performance of laboratory batch tests which also showed high methane oxidation at temperatures up to 60°C,

which was the maximal observed temperature in the methane oxidation layer of the biocover system. The performed laboratory experiments also indicated that the biocover system would have been able to oxidize a significantly higher methane load than the load obtained by gas extraction from the three leachate wells.

At many landfills a significant part of the generated landfill gas may emit through leachate wells. At such landfills the methane emissions 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 methane load to the biocover system could be significantly increased to evaluate the in-field capacity of the biocover system.

With an additional performance evaluation of the biocover under higher methane loads a more realistic methane oxidation capacity could be determined and used for dimensioning a full-scale biocover system for the AV Miljø Landfill. At a time where the landfill is finally capped the leachate and inspection well heads could be changed to gas-tight versions. In this way a better control of the gas routed in the leachate collection system could be obtained, and new permanent biofilter systems with the right dimensions based on the final evaluation of the methane generation at the landfill could be constructed as the landfill gas after-care installation leading to a low methane emission in the after-care period. Obtaining a realistic methane oxidation capacity of biofilters similar to the constructed pilot-scale biofilter a mitigation cost (in DKK/tons CO_2-equivalences) can be calculated.

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