Evaluation of methane oxidation efficiency of two biocovers: field and laboratory results

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Roncato, C.D.L. and Cabral, A.R. (2012). Evaluation of methane oxidation efficiency of two biocovers: field and laboratory results. J. Env. Eng. (ASCE), 138(2): 164-173

Abstract

Biocovers constitute a promising technology to reduce fugitive and residual emissions from landfills throughout their operational life and after gas collection systems are turned off. The aim of this study was to assess the efficiency of two substrate materials to oxidize CH_4 into CO_2 under field conditions and in the laboratory (column tests). The two substrates evaluated were: 1) a mixture of sand and compost, and 2) a mixture of the sand–compost with gravel. The oxidation rates obtained in the field attained a maximum of 576 g CH_4 m^{-2} d^{-1} for one of the substrates and 352 g CH_4 m^{-2} d^{-1} for the other. These maximum values were much higher than those obtained in the laboratory: 115 and 118 g CH_4 m^{-2} d^{-1}, respectively, for an oxidation efficiency of 96% in both cases. The exact causes for the discrepancy between field and laboratory results were not identified, but it was hypothesized that vegetation on the surface of the passive methane oxidation biocovers (PMOBs) greatly improved methane oxidation efficiencies in the field. The results obtained in this study show that laboratory column tests constitute a reliable means to evaluate potential candidate materials for biocovers. However, the maximum oxidation efficiencies may not be the same as those obtained in the laboratory.

Keywords: landfills emissions, column tests, field tests, greenhouse gases

1. Introduction

In small and old landfills, gas collection is not cost effective, and as a result all the biogas produced is allowed to escape into the atmosphere, constituting what is called fugitive emissions. Residual emissions are expected to be released from landfills after gas collection systems are turned off. Methane emissions from landfills, particularly fugitive and residual emissions, can be oxidized to CO_2 in the presence of oxygen through microbial methane oxidation in landfill cover soils or biocovers (Humer and Lechner 1999; Hilger et al. 2000; Huber-Humer et al. 2008; Ait-Benichou et al. 2009; Scheutz et al. 2009).

The most recent IPCC Working Group III assessment report lists biocovers and biofilters as upcoming technologies and practices to mitigate methane emissions projected to be commercialized before 2030 (Bogner et al. 2008). The capacity of this top layer to oxidize CH_4 depends on both the physical and chemical properties of the cover material, on the biogas flow rate and its quality, and on climatic parameters such as atmospheric pressure and temperature (Einola et al. 2007; Scheutz et al. 2009, etc.).

The prospect of mitigating CH_4 emissions passively and for a long period, prompted an ongoing multidisciplinary project (started in November 2005) that jointly considers geotechnical, hydraulic, physico-chemical, environmental, climatic and microbiological aspects in assessing the oxidation efficiency of landfill cover materials that would act as passive methane oxidation biocover (PMOB). The study presented herein is part of this project. It presents and analyzes the results obtained during the 2009 monitoring campaign for two of the experimental plots, namely, PMOB 2 and PMOB 3B. Moreover,

this article presents the results of two oxidation tests that were performed in the laboratory with substrate sampled from the same two experimental PMOBs. Comparisons between CH_4 oxidation rates and efficiencies obtained in the field and in the laboratory are limited because of important differences in boundary conditions, such as substrate thickness and presence of vegetation (which allow for more efficient moisture exchange and development of methanotrophic colonies). However, this type of comparison is fundamental if column tests are to earn credibility and become a reliable tool for evaluating candidate substrate materials for future full-scale projects.

2. Materials and methods

2.1 Field experiments

Three experimental plots measuring 2.75-m (W) x 9.75-m (L) were constructed during the summer of 2006, within the existing final cover of the St-Nicéphore landfill, located in Quebec, Canada, in an area where the waste mass is approximately 5-years old. PMOB 2 (Figure 1a) included a 0.80 m thick layer of substrate underlain by a 0.10 m thick gas distribution layer (GDL) consisting of 6.4-mm net gravel and 0.3 m of 12.7-mm net gravel. The substrate layer consists of a mixture of 5 volumes of mature compost sieved through a 12 mm industrial sieve and 1 volume of coarse sand (D_{10} = 0.07 mm; D_{85} = 0.8 mm; Cu= 4.3). The compost was sieved prior to mixing with the sand. More details on the compost and the mixture can be found in Jugnia et al. (2008). The substrate layer was placed in four 0.2 m layers and compacted with a vibrating plate to obtain layers with an average dry unit weight of 840 kg m^{-3} and total porosity (n) equal

to 63%. The specific gravity (Gs) of the sand-compost mixture is equal to 2.24 and the organic matter of the substrate was 18% (Cabral et al. 2010a).

In 2007, in order to increase the air filled porosity (AFP) of PMOB 3, thereby improving air penetration, this plot was re-excavated and PMOB 3B was constructed in its place with the configuration shown in Figure 1b (Cabral et al. 2008). The substrate of the new plot is composed of one volume of the same mixture of sand-compost of PMOB 2 and one volume of 6.4-mm gravel. The substrate was compacted to a unit weight of 1428 kg m^{-3}, with n equal to 48%. The Gs was 2.74 and the organic matter of the substrate was 6%. The GDL is composed of 0.10 m of 6.4-mm gravel and 0.8 m of coarser gravel (12.7-mm). A biogas distribution system was installed within the GDL to feed the two PMOBs with biogas from a well, constructed for the purposes of this study. The PMOB was lined using a 1.5-mm thick HDPE geomembrane (GM) in order to isolate them from the existing 2.7-m thick silty cover.

Part of the instrumentation installed within the PMOBs consisted of the following probes: water content, suction, temperature and gas (Figure 1c). Gas profiles of O_2, CO_2 and CH_4 were obtained from weekly samplings collected from the aluminum gas probes and analyzed *in situ* using a portable gas meter (Columbus Instruments Inc.) equipped with infrared sensors able to detect CO_2 and CH_4 on a scale from 0 – 100 vol% and an electrochemical sensor calibrated to detect O_2 from 0 – 21 vol%. The detection limit for CH_4 and CO_2 was 2 vol% and 1 vol% for O_2. For every visit to the site, only one sample for each depth of each profile was taken.

Figure 1 – Configuration of PMOB 2 (a) and PMOB 3B (b) and a representative plan view of both (c)

In order to determine CH_4 surface emissions (J_{out}), the static chamber technique was performed at two different points on each experimental plot (Figure 1c). The chamber used was manufactured by Odotech Inc. (following an EPA design) and a diameter of 0.49 m and a height of 0.4 m. The perimeter of the Plexiglas® chamber was sealed with a bentonite paste to prevent intrusion of atmospheric air into the chamber. The evolution of CH_4 concentrations within the chamber were monitored every 5 seconds over a 6 minute interval using a portable FID (TVA 1000B, Thermo Scientific) equipped with a data acquisition system. The detection limit of FID was 0.1 ppm of CH_4 within the 0 – 5% range (beyond this value the equipment shuts down). In order to reduce induced suction due to pumping by the FID equipment, a three-way valve was installed, which permitted the operator to shift from pumping inside the chamber (for 15 s) to pumping atmospheric air (30 s). This procedure was retained after testing other combinations. Static chamber measurements were performed weekly. Surface emissions of CH_4 (J_{out}) were calculated according to the equation:

$$J_{out} = \frac{V}{A}\frac{\Delta C}{\Delta t} \quad (1)$$

where J_{out} is in mg/m²/s, V is the chamber volume (m³), A is the internal surface area (m²), and $\Delta C/\Delta t$ (mg/m³/s) represents the slope of the plot relating change in gas concentration to time.

A Sage Integral Prime (SIP) mass flow meter connected to a data logger permitted continuous monitoring of the CH_4 loading (J_{in}) into the PMOBs. The following equations were used to calculate the methane oxidation efficiency (f_{ox}) and the oxidation rate (J_{ox}) of the PMOBs on the basis of methane mass balance:

$$f_{ox} = \frac{J_{in} - J_{out}}{J_{in}} \times 100 \quad (2)$$

$$J_{ox} = \frac{J_{out}}{(f_{ox}^{-1}) - 1} \quad (3)$$

where f_{ox} is the CH_4 oxidation efficiency (%), J_{in} is the CH_4 loading (g CH_4 m^{-2} d^{-1}) and J_{ox} the CH_4 oxidation rate (g CH_4 m^{-2} d^{-1}). In this equation J_{out} is in g CH_4 m^{-2} d^{-1}.

CH_4 surface concentration scans were performed using the FID equipment, following a pre-defined path inside the PMOBs. Gas samples were obtained continuously every 5 seconds, with a probe maintained at a distance of approximately 0.1 m above the surface. Sampling was made at a total of 60 points on the surface of each PMOB.

Isotope analyses were performed at the G.G.Hatch Lab (Department of Earth Sciences, University of Ottawa). The GC-C-IRMS system consists of a HP 5890 Series II gas chromatograph (GC) coupled with a Delta Plus (Thermo-Finnigan) isotopic ratio mass spectrometer via a combustion interface GC combustion III. The GC column was a PoraPlot Q (Varian, CP-7551) plot-fused silica column (25 m, 0.32 mm). The results obtained were normalized (re-calculated versus VPDB) using two international gas standards: NSG1 (RM8559) with methane ^{13}C value of -29.11 and NSG2 (RM8560) with

a methane ^{13}C value of -44.84. Precision and accuracy on 3rd blind standard (internal) was better than 0.2permil.

2.2 Column tests

For the oxidation column tests performed in the laboratory, an artificial biogas was used. The CH_4 and CO_2 concentrations were equal to 50% (vol%). Plexiglas® columns (Ø = 0.15 m) were built, with a perforated plate at the bottom and lateral sampling ports were inserted at every 0.10 m (Figure 2). Sampling ports along the columns were equipped with silicone septa to enable collection of gas samples via a syringe needle. Several column tests were performed with the substrate taken of PMOB 2 and 3B. The substrate of PMOB 2 had been exposed to methane for three years, whereas the substrate of PMOB 3B had been exposed for 2 years. Only two representative tests are discussed herein. For the first, the 0.30-m-high substrate sample of PMOB 2 was compacted with an initial degree of (water) saturation, S_r, equal to 69% (AFP = 20%). For the 2nd column test, the PMOB 3B substrate was tested with a substrate height of 0.30 m and S_r equal to 41% (AFP = 28%). During the tests, the CH_4 loading was measured using micro flow meters (Gilmont, GF-3060). Loadings varied between 0.15 and 2.30 ml min^{-1} (8 and 127 g CH_4 m^{-2} d^{-1}). Atmospheric air was introduced at the top of the columns by a peristaltic pump. Gas samples were taken from the head-space and the sampling ports. Three to four times a week 1-ml gas samples were collected from the columns and analyzed using a Micro GC 3000A gas chromatograph (Agilent), equipped with two columns: a Molsieve 5A to quantify the O_2, N_2 and CH_4 and a

PoraPlot Q to quantify the CO_2. The entire methodology for column tests is described by Roncato (2009).

Figure 2 – Schematic of the column tests

3. Results and discussion

3.1 Methane removal efficiencies in the field

PMOB 2

Figure 3 shows the evolution with time of the CH_4 loading ("CH_4 in") and outflux ("CH_4 out"), oxidation rate and oxidation efficiency for PMOB 2. During the first month of the 2009 monitoring campaign the CH_4 loading was kept low to allow full development of vegetation and re-establishment of the methanotrophic colony. The initial loading of 8 g CH_4 m^{-2} d^{-1} corresponds to a residual loading that may be found at the base of a landfill cover decades after the final cover installation (Stegmann et al. 2007). Once the vegetation was well established, the CH_4 influx was gradually increased from 20 g CH_4 m^{-2} d^{-1} to 580 g CH_4 m^{-2} d^{-1}. Previous experience both in the field and the laboratory (Roncato et al. 2010; also see the results presented in this paper; also some yet unpublished results) show that a gradual increase in loading allows to attain higher oxidation efficiencies.

Figure 3 – Evolution of CH_4 loading, emissions and oxidation efficiencies for PMOB 2

Fluctuations in oxidation efficiencies were observed until the end of July. This fluctuation may have been partly caused by adaptation of the system to the increase in loading, partly by fluctuations in the degree of saturation (S_r) within the profile of this PMOB; the latter being caused by intense rainfall in June and July 2009, during which several precipitation events of more than 15 to 20 mm were recorded.

The maximum oxidation rate for PMOB 2 in 2009 was 576 g CH_4 m^{-2} d^{-1} with an oxidation efficiency of 99%. This value corresponds to the maximum CH_4 influx that could be supplied by the biogas well because the controlling valve was left completely open from September 1. After this date, it can be observed that CH_4 influx naturally decreased. According to information provided by the landfill manager, biogas pumping around the sector where the PMOB is installed increased in September (personal communication). Possible spatial variation in gas production or unaccounted for preferential landfill gas flow paths (Einola et al. 2009) cannot be ruled out. The CH_4 surface outflux measured with the chambers from June to September varied from 0 (below detectable limits) to 21.9 g CH_4 m^{-2} d^{-1} (maximum value), with an average of 4.2 g CH_4 m^{-2} d^{-1} and a standard deviation of 5.8 g CH_4 m^{-2} d^{-1}.

The air temperature between June 15 and September 30, 2009 varied between 3 – 30°C, with an average of 18°C. During the same period, the temperature within the first 0.25 m of the PMOB averaged 28°C. During the month of September, when oxidation rates were peaking, the air temperature averaged 14°C, while the average temperature near the surface (0.0 - 0.25 m) topped 33°C, which is in the range of optima temperature for CH_4 oxidation (e.g. Humer and Lechner 1999; Scheutz et al. 2009). The fact that soil temperatures were much higher than average air temperatures can be

attributed to exothermic reactions, such as methane oxidation. One must acknowledge that temperature increase is also influenced by soil respiration. Unfortunately, the individual influence of each phenomenon could not be estimated.

During the last 3 weeks of the monitoring campaign, the average air temperature dropped abruptly to 6°C, with a minimum of – 5°C. Concomitantly, the average substrate temperature down to 0.25 m dropped to 14°C. As expected, this unusually cold weather in 2009 caused a noticeable decrease in oxidation rates, thus in the CH_4 oxidation efficiencies (Figure 3). The same kind of observation has been made in other studies (e.g. Czepiel et al. 1996; Boeckx et al. 1999; Gebert et al. 2003). Comparatively, towards the end of the 2008 field campaign (which followed the same sampling and testing methodologies described above), temperatures were milder than in 2009 (Cabral et al. 2010a), which explains why the oxidation efficiencies in October 2008 were still high (oxidation rates in early October peaked at 804 g CH_4 m^{-2} d^{-1}), whereas in 2009 they dropped suddenly. In cold areas, or during the winter, CH_4 oxidation may be significantly reduced or even come to a standstill (Scheutz et al. 2009). Einola et al. (2007) found that CH_4-oxidizing microorganisms were still able to oxidize CH_4 at temperatures varying between 1 – 19°C.

Figure 4 presents the correlation between CH_4 oxidation rates and CH_4 influx. Oxidation rates increased with the increasing CH_4 influx, following a slope almost equal to unity (R^2 = 0.999), meaning that oxidation efficiencies were very close to 100% during the 2009 campaign. The last three oxidation rate measurements (taken in October) were not considered in the regression analysis, because, the surface of the PMOB was frozen, making it difficult to properly perform the adopted flux chamber protocol. As a

consequence, there is reasonable doubt about the validity of the values obtained in October (including the oxidation efficiency values shown in Figure 3).

Figure 4 – Methane oxidation rates as a function of CH_4 loading

Figure 5 – Representative gas profiles for different CH_4 loadings of PMOB 2: (a) low; (b) mid-range; and (c) high CH_4 loading

Figure 5 presents gas profiles representative of three different CH_4 loading steps, with each gas profile representing an average of four profiles within PMOB 2 (Figure 1c). Figure 5a represents the case of a relatively low CH_4 loading. In this case, the CH_4 concentration started to decrease at the interface between the substrate and the gas distribution layer, i.e. at a depth of 0.82 m. This decrease is partly caused by dilution of atmospheric air (the high N_2 concentrations at depth indicate that atmospheric air penetrates until the bottom of the substrate layer). However methane oxidation is definitely taking place, as confirmed by stable isotope analyses (see representative set of results in Table 1) and mass balance calculations performed to obtain the results in Figure 3.

It can be observed in Figure 5a that near the surface (from 0.40 to 0.10 m) the slopes of the CO_2 and CH_4 profiles are nearly parallel; in fact the CO_2 concentration profile is slightly steeper. Assuming that CO_2 and CH_4 are diluted equally by atmospheric air and that oxidation was not taking place (i.e. there was only dilution), the ratio between the concentrations of CO_2 and CH_4 in the soil pores would be maintained and the two

profiles should converge when approaching the surface. As a consequence, the near parallelism observed indicates that CO_2 is being generated as a result of oxidation and soil respiration. Enrichment in ^{13}C was observed in the upper part of the profile, confirming that CH_4 oxidation was taking place (Table 1 and Cabral et al. 2010b); most of it near the surface. Soil respiration was not measured directly; it is thus impossible to assess its relative importance to oxidation. The much steeper CO_2 profile at depth, which crossed the CH_4 profile at 0.70 m, is a clear indicator that CH_4 oxidation is occurring deep in the substrate layer. Again, this is backed by stable isotope data (e.g. Aug. 24, P2P4, in Table 1). The results of stable isotopes will be discussed in another article.

Table 1 – Oxidation efficiencies determined by stable isotopes for PMOB 2 and 3B

Figure 5b presents the gas profiles for a mid-range CH_4 loading. The concentration of CH_4 at the bottom of the substrate increased with the increase in CH_4 loading and, as expected, the CH_4 and CO_2 profiles crossed at a shallower depth (0.60 m). Again, dilution by atmospheric air (N_2 penetrates deep into the substrate layer) may partly explain the drop in CH_4 concentrations. In view of repeated evidence of CH_4 oxidation (Figure 3; Table 1), it is possible to attribute the sharp drop in CH_4 at depth - at least in part - to biotic oxidation.

The gas profiles relative to the maximum biogas loading are shown in Figure 5c. The high influx results in more difficult atmospheric air penetration, and oxidation was taking place mostly within the first 0.40 m, where air was able to penetrate. The sharp decrease in O_2 concentrations within the first 0.10 m of the biocover shows that most of

the biological activity was happening very near the surface. The CO_2 concentration profile crossed the CH_4 profile at approximately 0.15 m, which in absence of more precise indicators (such as biotic activity), approximately identifies the oxidation front. The latter does not diminish the relative importance of soil respiration.

Several scans of methane concentration at the surface of PMOB 2 were performed. For low CH_4 loadings, the maximum recorded CH_4 surface concentration was 76 ppm. For mid-range loadings, the maximum CH_4 concentration was 103 ppm. Finally, the maximum CH_4 concentration observed when the PMOB was submitted to high loadings was 344 ppm, which is still below the maximum surface concentration allowed by the Quebec landfill regulation, i.e. 500 ppm.

In absolute terms, the above-mentioned values of surface concentrations may be considered high. However, it must be acknowledged that the loadings applied were quite high. Indeed, low loadings herein are in the range of 50 g CH_4 m^{-2} d^{-1}. When the maximum surface concentration was 344 ppm, the flux measured with the chamber was ~ 20 g CH_4 m^{-2} d^{-1}, while the CH_4 loading was 580 g CH_4 m^{-2} d^{-1} (thus an efficiency of 97%, despite the relatively high outflux value in absolute terms).

Observed low concentrations of CH_4 along the PMOB 2's perimeter, as well as variability in the locations of registered peaks were interpreted as an indication that the seal along the interface between the substrate and the geomembrane was good enough to prevent gas leaks. As a consequence, the surface point measurements were considered representative of the entire surface of the PMOB.

PMOB 3B

Figure 6 shows the evolution with time of the CH_4 loading ("CH_4 in") and outflux ("CH_4 out"), oxidation rate and oxidation efficiency for PMOB 3B. As a result of some technical problems with monitoring equipment, monitoring of this PMOB started one month after PMOB 2. By the time biogas was introduced, the vegetation was fully developed. Due to the delay, it was decided to increase the CH_4 loading at a faster pace than for PMOB 2 (Figure 3). The initial loading of 20 g CH_4 m^{-2} d^{-1} was maintained for only one week, after which it was brought to 50 g CH_4 m^{-2} d^{-1} for two weeks and then quickly increased to 352 g CH_4 m^{-2} d^{-1}.

Figure 6 – Evolution of CH_4 loading, emissions and oxidation efficiencies for PMOB 3B

Contrary to what was observed in PMOB 2, fluctuations in CH_4 oxidation efficiencies were not observed during the month of July. The greater air-filled porosity of the substrate material of PMOB 3B, as compared with that of PMOB 2, was likely responsible for this situation. Indeed, it was observed that the S_r of PMOB 3B did not fluctuate as much, i.e. the PMOB was not as affected by intense rainfall during July.

The maximum oxidation rate for PMOB 3B in 2009 was 352 g CH_4 m^{-2} d^{-1} with an oxidation efficiency of 100%. Following this maximum CH_4 influx, technical problems led to a decrease in loading, but oxidation efficiencies remained close to 100%. On Sept. 1, the loading was increased to almost 300 g CH_4 m^{-2} d^{-1} and a similar oxidation rate was

obtained on Sept. 4, 2009, for an efficiency of nearly 100%. As in the case of PMOB 2, the inlet valves were left fully open after Sept. 1. Any subsequent fluctuation in loading was natural, as explained earlier. The CH_4 surface outflux measured with the chambers from June to September varied from 0 (below detectable limits) to 22.2 g CH_4 m^{-2} d^{-1} (maximum value), with an average of 4.2 g CH_4 m^{-2} d^{-1} and a standard deviation of 6.1 g CH_4 m^{-2} d^{-1}.

Between June 15 and Sept. 30, the temperature within the first 0.25 m of PMOB 3B averaged 30°C, i.e. slightly higher than the average obtained for PMOB 2 during the same period; partly due to the coarser nature of the material. During the month of September, when oxidation rates were high, the average temperature near the surface topped 37°C; again higher than the average temperature of PMOB 2. During the last 3 weeks of the monitoring campaign, penetration of cold air brought the average substrate temperature near the surface (0 - 0.25 m) down to 11°C, whereas the average substrate temperature for PMOB 2 for the same period and layer was 14°C. Likewise in PMOB 2, the cold temperatures in October 2009 caused a significant decrease in oxidation rates (Figure 6), which dropped from nearly 100% to approximately 50%.

The maximum oxidation rate for PMOB 3B was 352 g CH_4 m^{-2} d^{-1}, while for PMOB 2 it was 576 g CH_4 m^{-2} d^{-1}. In fact, in both cases, the maximum oxidation capacity may never have been attained. On one hand, the valves were fully open right from the beginning of Sept. 2009, which caused the oxidation rates to top at a value equal to the loading rate (oxidation efficiencies = 100%). Were it not for the cold weather and the impossibility of feeding further biogas into the two PMOBs, it is expected that oxidation rates might have increased further. Indeed, the maximum rate attained at PMOB 2 in

2008 was 804 g CH_4 m^{-2} d^{-1} (Cabral et al. 2010a, who suggested that the latter value might not be maximum capacity). No comparison can be made with earlier performances of PMOB 3B, because a leak in the geomembrane liner occurred in 2008 (it was repaired in the Spring of 2009; Roncato et al. 2010). Before 2008, PMOB 3B was constituted of other materials (and was referred to as PMOB 3).

The same type of correlation observed in Figure 4 (PMOB 2) was obtained for PMOB 3B. The coefficient of determination, R^2 was equal to 0.999, if the last three oxidation rate measurements (taken in October) are discarded, for the same reason evoked when discussing the results of Figure 4.

Figure 7 – Representative gas profiles for different CH_4 loadings of PMOB 3B: (a) low; (b) mid-range; and (c) high CH_4 loading

Figure 7 presents gas profiles representative of three different CH_4 loading steps, with each gas profile representing an average of four profiles within PMOB 3B. Figure 7a represents the case of a relatively low CH_4 loading, where it can be observed that the CH_4 concentration started to decrease within the gas distribution layer (identified as gravel layer in Figure 7). This decrease was partly caused by dilution of atmospheric air, given the fact that atmospheric N_2 concentrations at depth were quite high, but also in part to methane oxidation. Indeed, the mass balance calculations performed to obtain the results in Figure 6 and the enrichment in ^{13}C observed higher up in the profile (Table 1) confirm the latter assertion.

The fact that CO_2 concentrations were higher than the CH_4 concentrations well below the interface between the GDL and the substrate indicates that methane oxidation was occurring deep inside the GDL. Again, this is backed by stable isotope data (e.g. for the Aug. 20 data in Table 1). It can be observed in Figure 7a that the slopes of the CO_2 and CH_4 profiles were nearly parallel from 0.82 to 0.20 m. Likewise in PMOB 2, the near parallelism observed indicates that CO_2 was being generated as a result of oxidation and soil respiration.

Figure 7b presents the gas profiles for a mid-range CH_4 loading. The concentration of CH_4 at the gas distribution layer increased with the increase in CH_4 loading. The CH_4 and CO_2 profiles crossed at a shallower depth (~ 0.30 m), i.e. at the interface between the substrate and the GDL. Compared with the profiles for a mid-range CH_4 loading for PMOB 2, the crossover between the CH_4 and CO_2 profiles moved up further in the case of PMOB 3B than in PMOB 2. It can be hypothesized that the very coarse structure of the GDL in PMOB 3B did not allow sufficient retention time for oxidation to occur. However, when CH_4 reached the interface, there was sufficient microbial activity to drastically reduce the CH_4 concentration, despite the higher loading. Again, dilution by atmospheric air may partly explain the drop in CH_4 concentrations within the substrate.

The gas profiles relative to a high biogas loading are shown in Figure 7c. The high biogas influx prevented atmospheric air from reaching the base of the GDL, where N_2 concentration values were almost equal to zero. Oxidation was taking place mostly within the substrate, i.e. in the region where air was able to penetrate and where the CO_2 and CH_4 profiles crossed (at a depth of 0.15 m), which approximately defined the oxidation front for this particular case.

As with PMOB 2, several scans of methane concentration at the surface of PMOB 3B were performed. For low CH_4 loadings, the maximum recorded CH_4 surface concentration was 12 ppm. For mid-range loadings, the maximum CH_4 concentration was 248 ppm. Finally, the maximum CH_4 concentration observed when the PMOB was submitted to high loadings was 500 ppm, or the maximum surface concentration allowed by Quebec landfill regulations. As with PMOB 2, despite the fact that the surface concentrations may be considered high in absolute terms, they represent a small fraction of the very high loadings, i.e. the overall efficiencies remained quite high.

3.2 Methane removal efficiencies in column tests

Figure 8 shows the evolution with time of the CH_4 loading ("CH_4 in") and outflux ("CH_4 out"), oxidation rate and oxidation efficiency for column PMOB 2 (Figure 8a) and column PMOB 3B (Figure 8b). The values of methane loading and influx of atmospheric air into the headspace are given in Table 2 for selected moments that are commented below.

Figure 8 – Evolution of CH_4 loading, emissions and oxidation efficiencies for substrate of (a) PMOB 2 ("column PMOB 2"); and (b) PMOB 3B ("column PMOB 3B")

Table 2 – Methane and air loadings during column tests

The CH_4 influx was gradually increased and the oxidation efficiency remained at 100% until the CH_4 influx attained 70 g CH_4 m^{-2} d^{-1}. Increasing the loading further to 100 g CH_4 m^{-2} d^{-1} led to a drop in oxidation efficiency: 83% for column PMOB 2 and 95% for column PMOB 3B. It was hypothesized that if the CH_4 loading was maintained at 100 g CH_4 m^{-2} d^{-1} for a few days, the microbial population would adapt to the higher loading and the efficiency would return to 100%. In fact the return to ~100% CH_4 oxidation efficiency did happen, but it was not possible to confirm with certainty that this positive outcome was indeed associated with adaptation of the methanotrophic population to the higher loading. This could be the object of further studies.

Another and more abrupt drop in oxidation efficiency was observed when the CH_4 loading attained approximately 125 g CH_4 m^{-2} d^{-1}. This drop was more noticeable in column PMOB 2 than in column PMOB 3B, because of the better aeration observed within the coarser substrate of PMOB 3B. This is clearly observed in the N_2 profiles for day 28 in Figure 9 (also see the profiles in Figure 10). This time, however, keeping the CH_4 loading approximately constant for a few days did not result in complete recovery of the oxidation efficiency. Instead, the oxidation efficiency gradually decreased. This seems to indicate that a threshold loading might have been exceeded and that the threshold value would be in the vicinity of 100 g CH_4 m^{-2} d^{-1}. The system does not seem to be able to take in greater CH_4 loadings, at least under the laboratory conditions imposed.

The potential causes of the drop in the oxidation efficiency are associated with less efficient oxygenation of the substrate and include: (a) high upward fluxes; and (b) an increase in the degree of saturation (S_r) of the substrates during the test. The latter fact

was also observed by others, including Hilger et al. (2000) and Albanna and Fernandes (2009). Indeed, measurements made in the beginning and in the end of the tests showed that the degree of saturation increased, particularly near the surface of the samples (Table 3). This increase in S_r was caused by the combined effects of oxidation, which also produces water, and of condensation in the headspace (due to humidification of the inlet air and synthetic gas), which eventually moistens the surface of the sample.

Table 3 – Initial and final degrees of saturation of column tests

The influence of the increase in S_r in reducing oxidation efficiencies is clearly illustrated in the results presented in Figure 9, where N_2 profiles for 3 different stages of the column tests are shown. For both column tests, it was decided to compare the profiles for similar loadings (~ 70 g CH_4 m^{-2} d^{-1}), lower than the threshold loading (~ 100 g CH_4 m^{-2} d^{-1}), before and after stressing the system at maximum loading (day 28 for both tests; loading ~ 125 g CH_4 m^{-2} d^{-1}). Despite the fact that the same loading was being applied on days 15 and 36 on column PMOB 2 (Figure 9a) and days 15 and 43, on column PMOB 3B (Figure 9b), it can be observed that N_2 did not penetrate as deeply at the end of the test. It can be hypothesized that this is the result of an increase in S_r. In addition, it must be noted that the atmospheric air influx was higher after stressing the system with high loadings than before (Table 2). As expected, the variations in N_2 penetration were more noticeable within the finer substrate (PMOB 2).

Figure 9 – Nitrogen profiles for column PMOB 2 (a) and column PMOB 3B (b)

Given the limited O_2 penetration, the loading was lowered to ~ 70 g CH_4 m^{-2} d^{-1} (day 35; Figure 8), as a first step in trying to boost oxidation efficiencies. Although the procedure proved successful, efficiencies never rebounded back to 100% (Figure 8).

The maximum oxidation rates for columns PMOB 2 and PMOB 3B were, respectively, 115 and 118 g CH_4 m^{-2} d^{-1}, for an oxidation efficiency of 96% in both cases. These oxidation rates are comparable to rates found in the literature for similar substrates (Kightley et al. 1995; De Visscher et al. 1999; Humer and Lechner 2001; Stein and Hettiaratchi 2001; Scheutz et al. 2003).

In another study (Roncato et al. 2010), a series of column tests were performed to assess the influence of a thicker substrate (0.45 m) and a greater initial S_r (63%). Only the substrate of PMOB 3B was tested. For the test with a thicker substrate, the maximum oxidation rate reached 145 g CH_4 m^{-2} d^{-1}. A thicker substrate results in increased retention times for transported CH_4, leading to improved oxidation rates (Stern et al. 2007). For the test with a greater initial S_r, the maximum oxidation rate was only 50 g CH_4 m^{-2} d^{-1}, i.e. nearly half the oxidation rate obtained in the present study. As discussed previously, greater values of degrees of saturation result in shallower penetration of O_2.

The laboratory oxidation rates are significantly lower than the maximum oxidation rates obtained in the field for the same substrates, i.e. 576 g CH_4 m^{-2} d^{-1} for PMOB 2 (Figure 3) and 352 g CH_4 m^{-2} d^{-1} for PMOB 3B (Figure 6). It is hypothesized that in field conditions, the presence of vegetation leads to a more efficient exchange of moisture

with the atmosphere through evapotranspiration, whereas in the laboratory, moisture keeps accumulating within the sample. Most importantly, vegetation enhances oxygenation (aeration) near the surface, boosting oxidation rates (Nagendran et al. 2006; Wang et al. 2008; Xiaoli et al. 2009).

One cannot deny that vegetation root respiration competes for the O_2 required for CH_4 oxidation. However, the positive effects the root zone has on the structure of the cover material cannot be neglected. Indeed, O_2 transport is improved substantially by the new structure created by the development of roots. In addition, roots offer support to methanotrophic colonies, and it is within the root zone where the greatest colonies are commonly found (e.g. Watanabe et al. 1997; Stralis-Pavese et al. 2004; Wang et al. 2008).

Figure 10 – Representative gas profiles for different CH_4 loadings of column PMOB 2 (a,b,c) and column PMOB 3B (d,e,f)

Figure 10 presents gas profiles representative of three different CH_4 loading steps during the test with columns PMOB 2 (Figure 10a,b,c) and PMOB 3B (Figure 10d,e,f). Figure 10a and Figure 10d represent the case of a relatively low CH_4 loading, where it can be observed that the CH_4 concentration already started to decrease at the bottom of the substrate. This decrease was caused in part by dilution of atmospheric air introduced on the top of the columns, in part by methane oxidation. For low CH_4 loadings, the profiles obtained in the laboratory are very similar to those in the field (Figure 5a; Figure 7a).

Figure 10b and Figure 10e present gas profiles when the threshold CH_4 loading was being applied to columns PMOB 2 and PMOB 3B, respectively. The substrate of PMOB 3B allowed deeper penetration of atmospheric air for the same CH_4 loading. Under field conditions, despite a higher mid-range methane loading, O_2 and N_2 penetrated deeper (Figure 5b; Figure 7b) than in the lab columns.

Figure 10c and Figure 10f show gas profiles for the maximum CH_4 loading for columns PMOB 2 and PMOB 3B. Again, the substrate of PMOB 3B allowed a better penetration of atmospheric air than the substrate of PMOB 2, and oxidation seems to have occurred at greater depths. According to the profiles obtained for this loading level in column PMOB 2 (Figure 10c), oxidation seems to occur only in the upper crust of the sample.

4. Conclusions

The aim of this study was to assess the efficiency of two experimental passive methane oxidation biocovers (PMOB) and the associated efficiency of columns constructed in the laboratory using the same materials, i.e. a mixture of sand and compost and a mixture of the latter with gravel.

The oxidation rates obtained in the field attained a maximum equal to 576 g CH_4 m^{-2} d^{-1} for PMOB 2 and 352 g CH_4 m^{-2} d^{-1} for PMOB 3B. Higher loadings might have been reached in the field (as was the case in 2008, when it reached 804 g CH_4 m^{-2} d^{-1}), were it not for insufficient gas pressure in the feeding well, followed by the early arrival of a cold front, which caused a drastic drop in methane oxidation rates and efficiencies. These maximum values were nonetheless much higher than those obtained in the

laboratory (115 and 118 g CH_4 m^{-2} d^{-1}, respectively, for an oxidation efficiency of 96% in both cases).

The exact causes for the discrepancy between field and laboratory were not identified, but it was hypothesized that vegetation on the surface of the PMOBs greatly improved methane oxidation efficiencies in the field. Without the presence of vegetation, the degree of saturation within laboratory samples increased, reducing air penetration, hence methane oxidation efficiencies. However, despite the discrepancies, the results show that laboratory column tests constitute a reliable means to evaluate potential candidate materials for biocovers. Laboratory oxidation rates would tend to underestimate the maximum CH_4 oxidation rates and efficiencies that might be attained in the field.

An attempt was made to evaluate the resiliency of the systems tested in the laboratory to continuous increases in methane loadings. It was observed that when oxidation efficiencies dropped after continuous increases, it was possible to re-establish the previous oxidation efficiencies by maintaining the same loading for a few days (rather than continuing to increase it). By the end of the tests, in order to maintain efficiencies at the same high levels, the loadings had to be decreased. This was caused by the observed increases in degree of saturation of the samples.

Acknowledgements

This work was supported in part by a grant from the Natural Science and Engineering Research Council of Canada (NSERC), BIOCAP Foundation Canada, the

Biotechnology Research Institute (NRC) and Waste Management Canada under strategic grant # GHG 322418-0; and in part by a Cooperative Research and Development grant from NSERC and Waste Management (grant # CRD 379885-08). The invaluable help of Jean-Guy Lemelin, technician, as well as of the numerous students involved in the project, namely M. Capanema, M. Létourneau, C. Chardin, E. Martel-Poliquin and I. Hernandes, must also be acknowledged.

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Figure 1 – Configuration of PMOB 2 (a) and PMOB 3B (b) and a representative plan view of both (c)

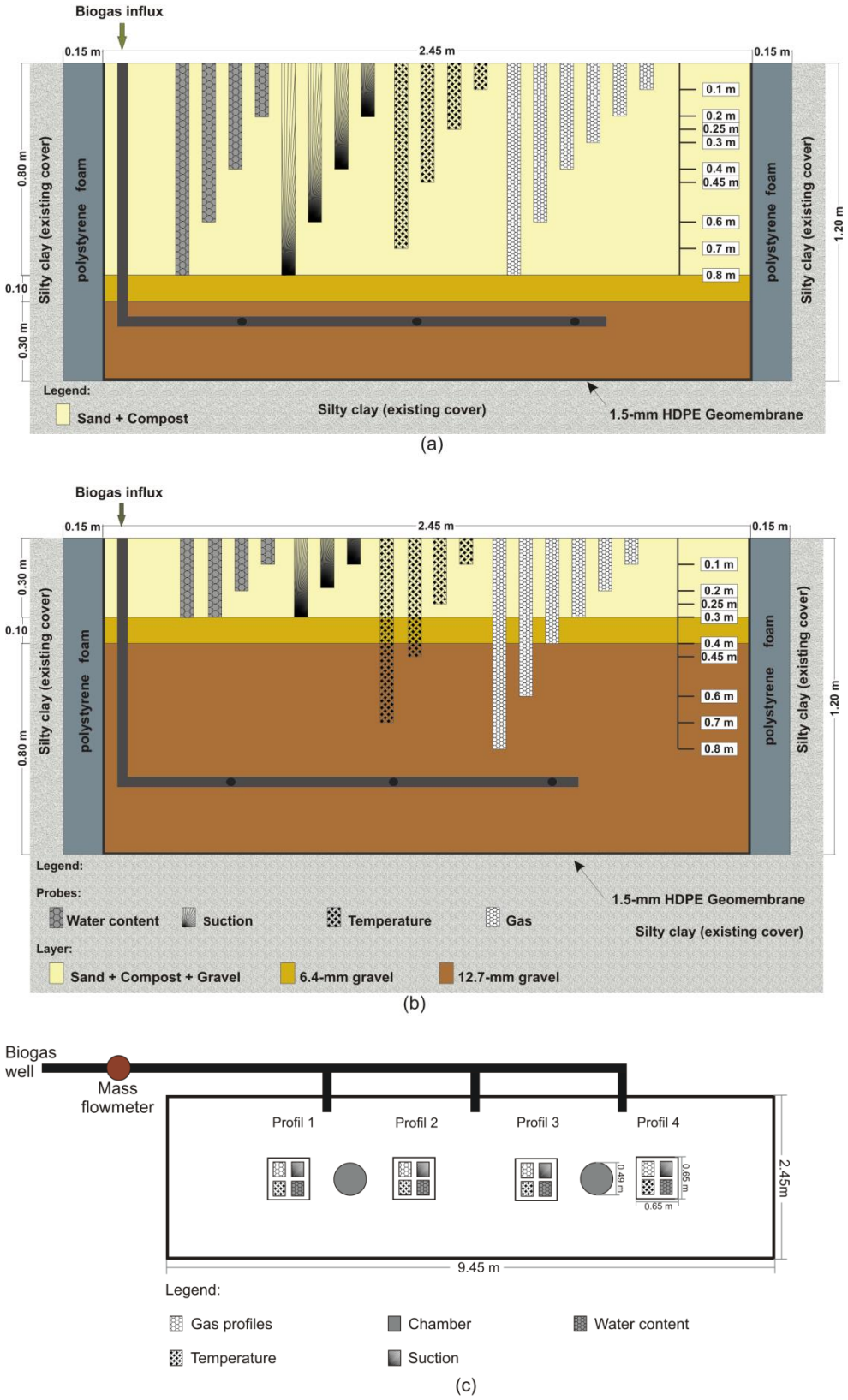

Figure 2 - Schematic of the column tests

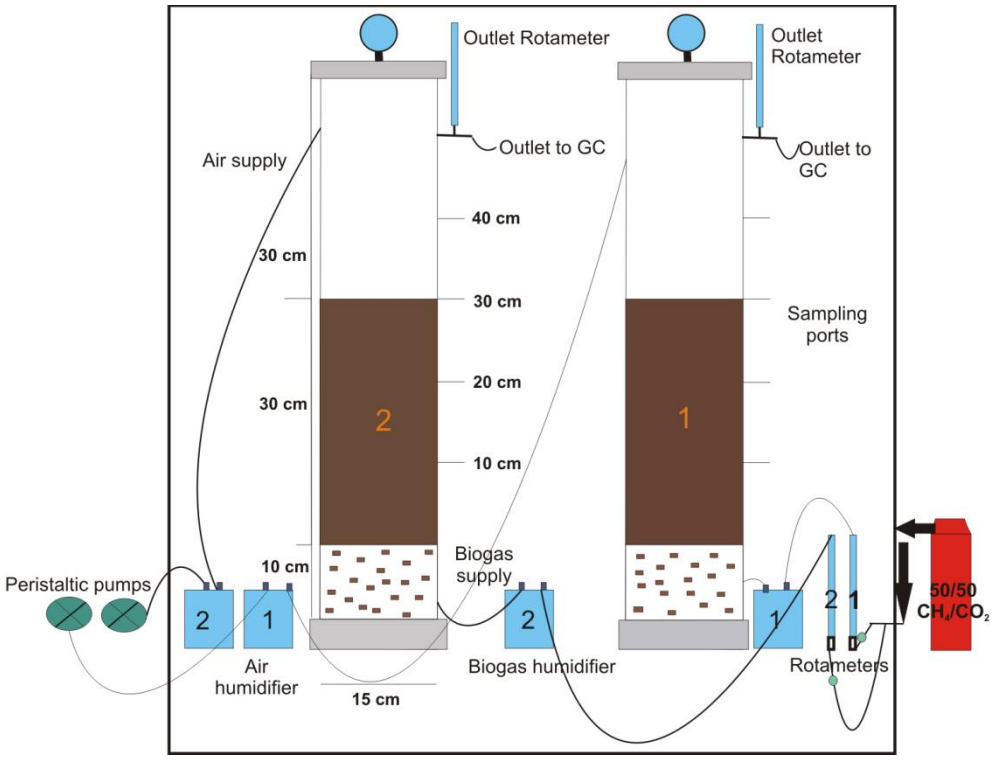

Figure 3 - Evolution of CH_4 loading, emissions and oxidation efficiencies for PMOB 2

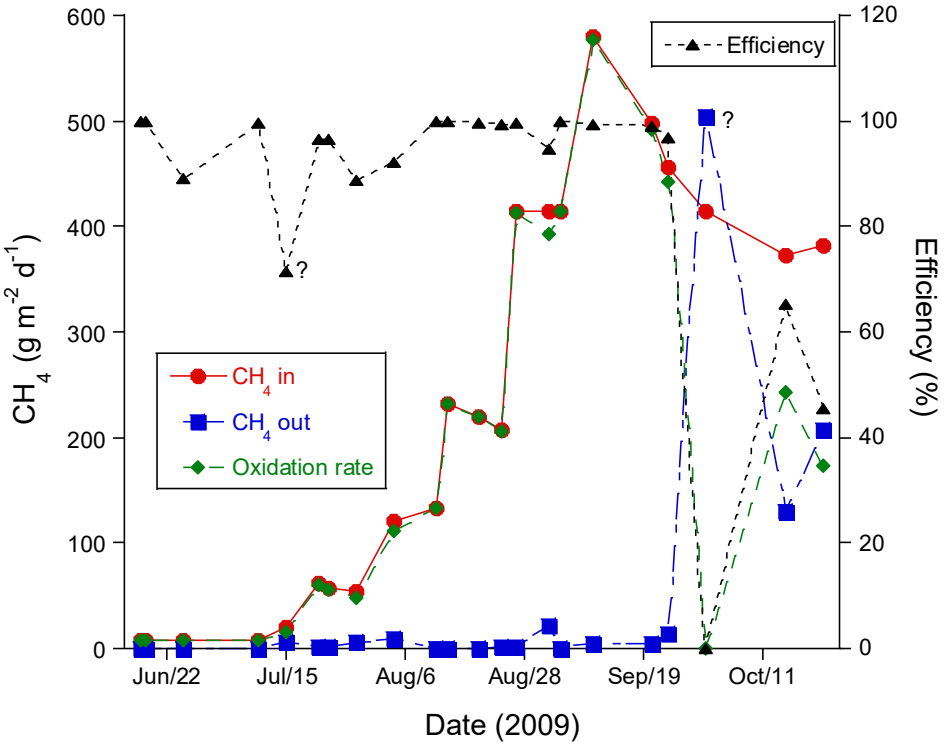

Figure 4 - Methane oxidation rates as a function of CH_4 loading

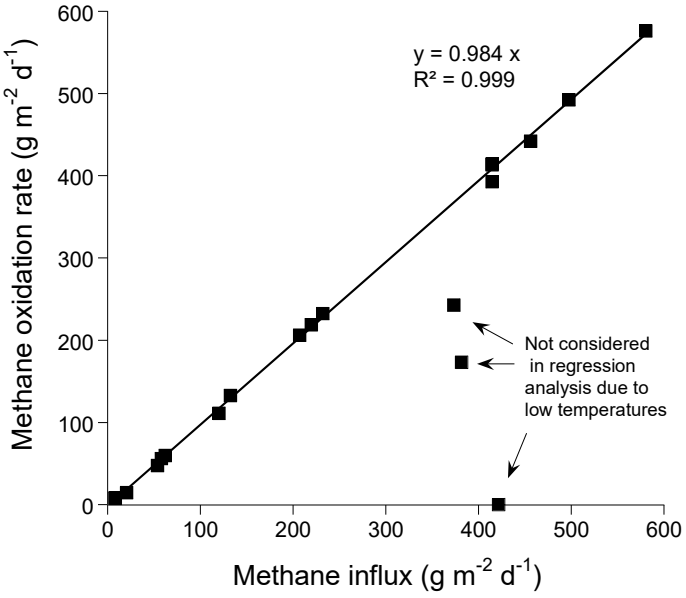

Figure 5 - Representative gas profiles for different CH_4 loadings of PMOB 2: (a) low; (b) mid-range; and (c) high CH_4 loading

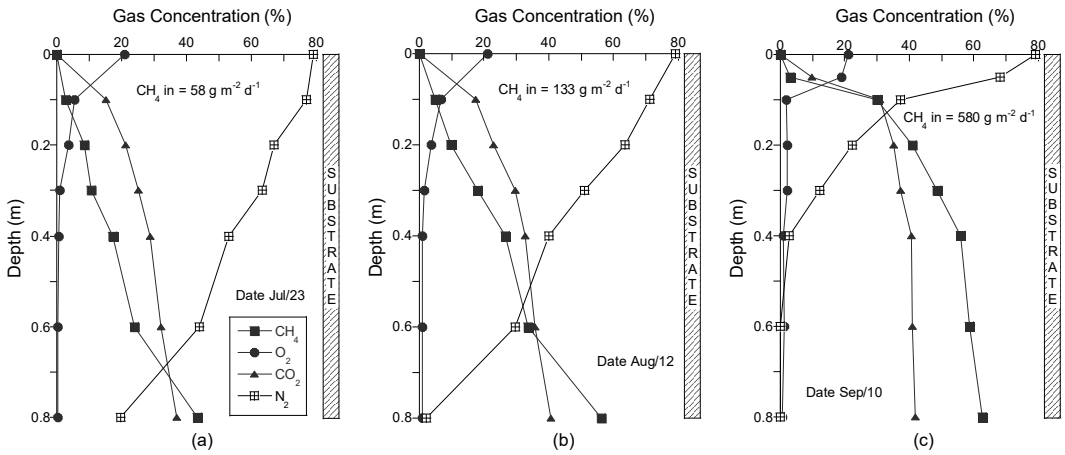

Figure 6 – Evolution of CH_4 loading, emissions and oxidation efficiencies for PMOB 3B

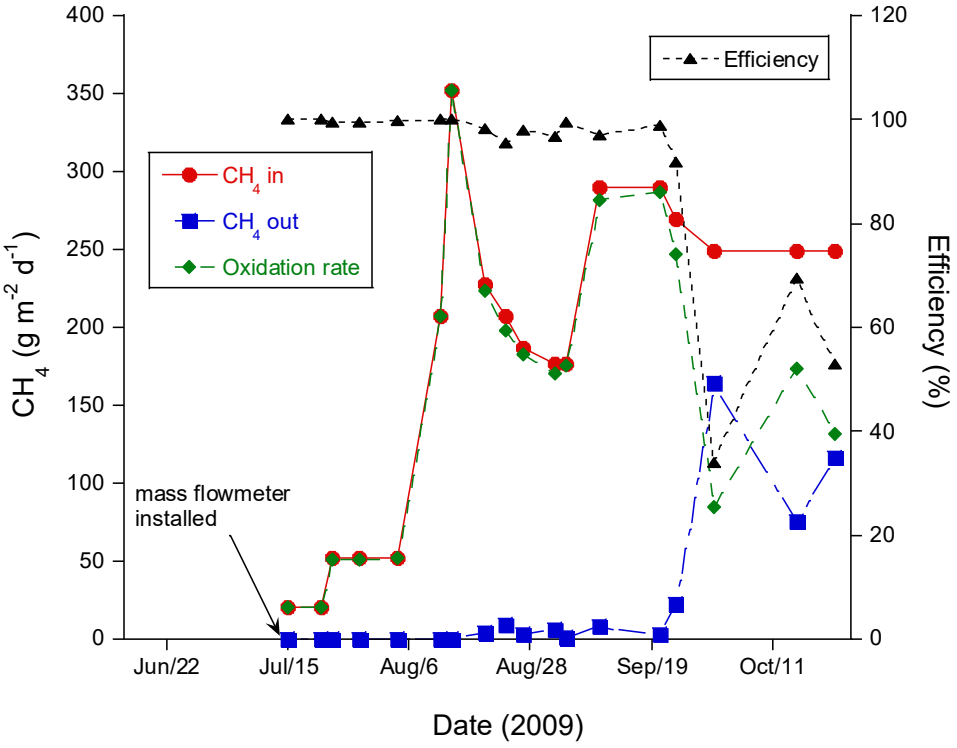

Figure 7 – Representative gas profiles for different CH_4 loadings of PMOB 3B: (a) low; (b) mid-range; and (c) high CH_4 loading

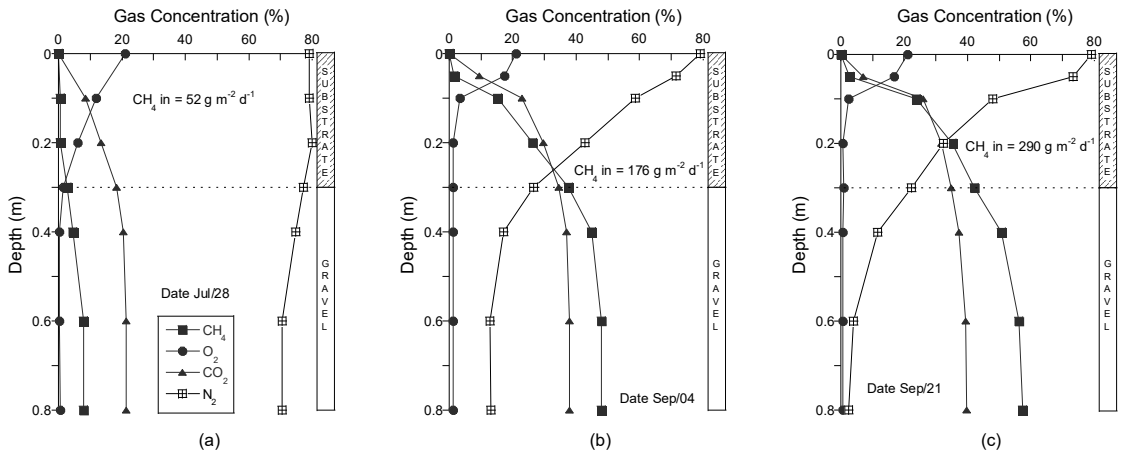

Figure 8 – Evolution of CH_4 loading, emissions and oxidation efficiencies for substrate of (a) PMOB 2 (“column PMOB 2”); and (b) PMOB 3B (“column PMOB 3B”)

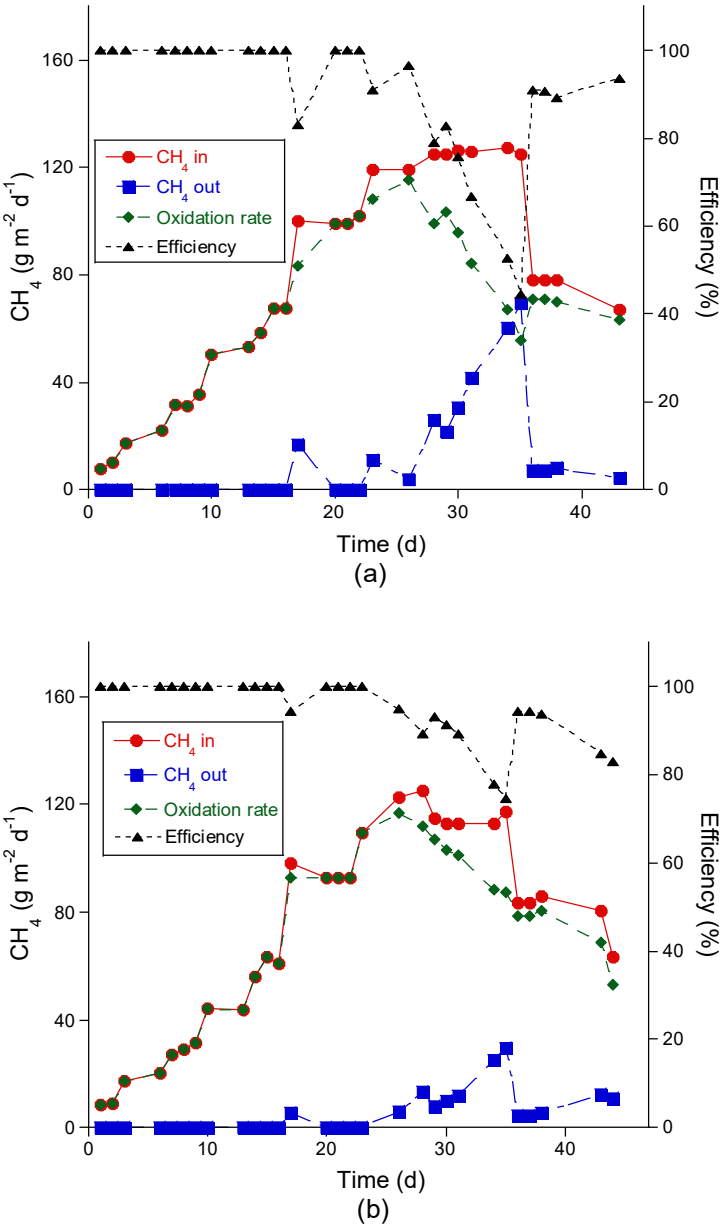

Figure 9 - Nitrogen profiles for column PMOB 2 (a) and column PMOB 3B (b)

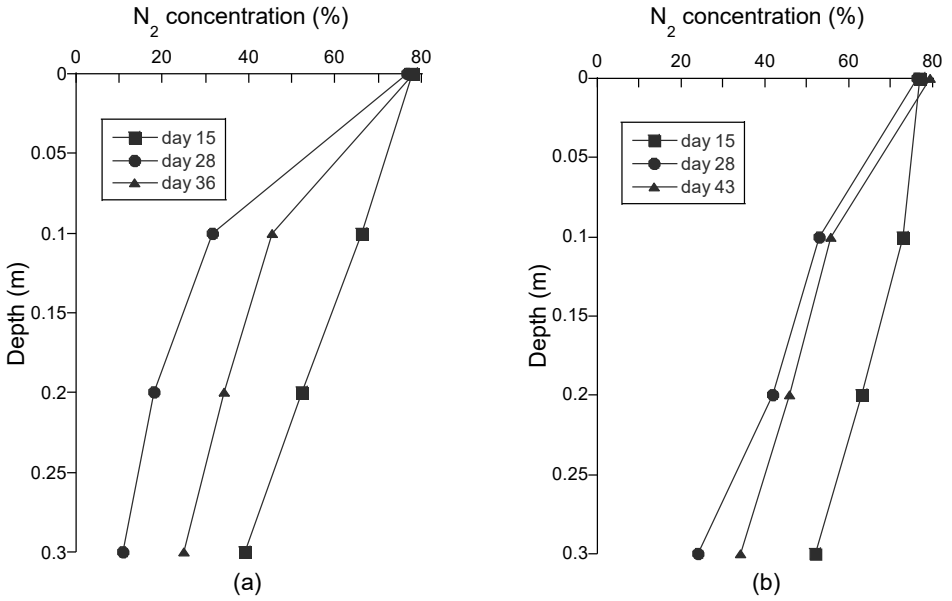

Figure 10 – Representative gas profiles for different CH_4 loadings of column PMOB 2 (a,b,c) and column PMOB 3B (d,e,f)

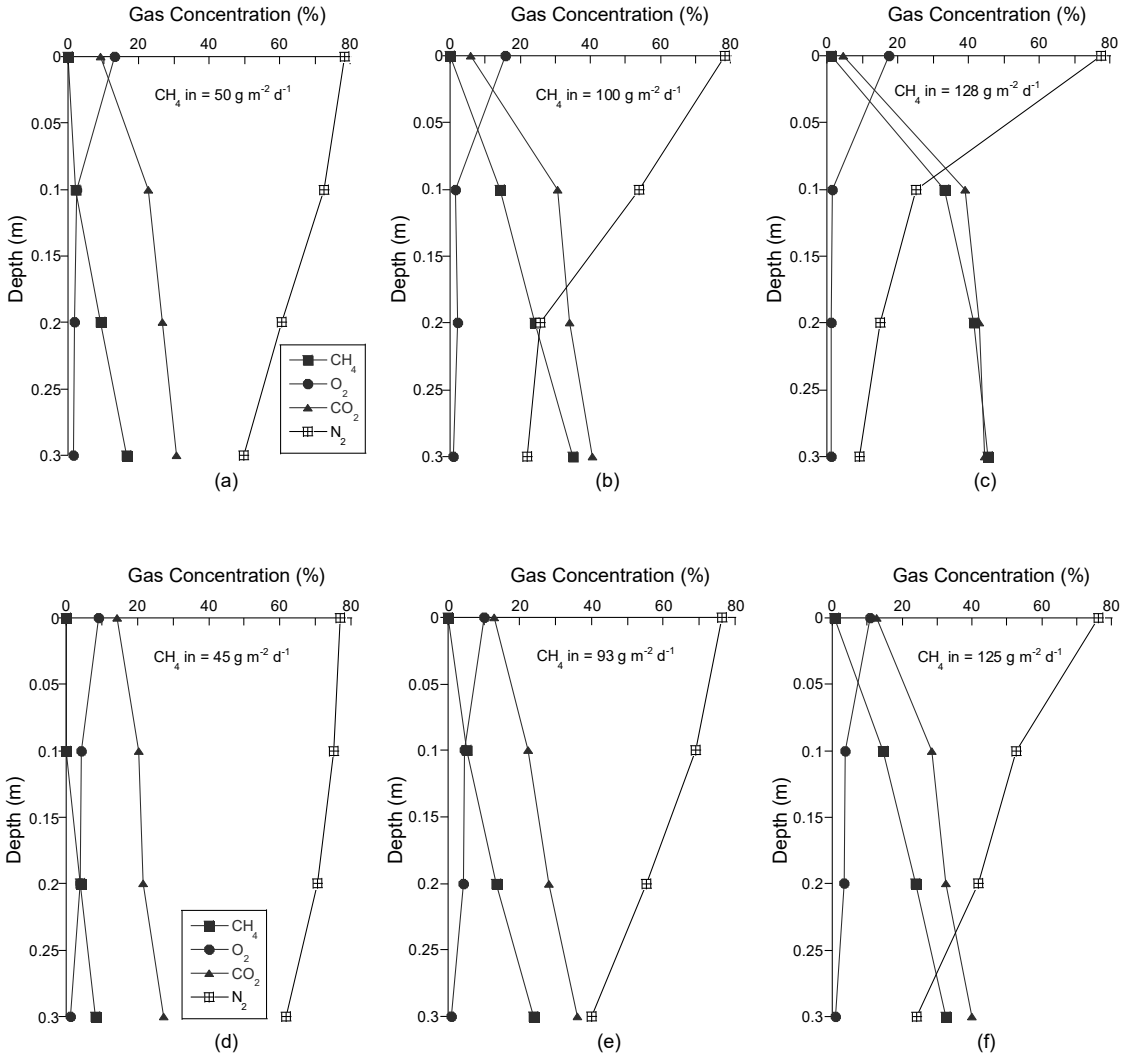

Table 1 – Oxidation efficiencies determined by stable isotopes for PMOB 2 and 3B

Date 2009	PMOB - Profile	Depth (m)	Oxidation f_0 (%)	Mass balance (%)
Aug. 24	P2 P1	0.00	Not determined	99.3
Aug. 24	P2 P1	0.10	94.4	
Aug. 24	P2 P4	0.10	68.9	
Aug. 24	P2 P4	0.30	53.8	Not Applicable
Aug. 24	P2 P4	0.60	40.6	
Aug. 24	P2 P4	0.82	00.0	
Aug. 20	P3B P3	0.00	Not determined	98.2
Aug. 20	P3B P3	0.10	(*)	
Aug. 20	P3B P3	0.30	(*)	Not Applicable
Aug. 20	P3B P3	0.82	75.8	
Aug. 27	P3B P3	0.00	Not determined	98.1
Aug. 27	P3B P3	0.10	67.0	Not Applicable
Aug. 27	P3B P3	0.82	40.5	
Sept. 21	P3B P3	0.00	Not determined	99.0
Sept. 21	P3B P3	0.10	93.2	Not Applicable
Sept. 21	P3B P3	0.30	50.5	
Sept. 24	P3B P3	0.00	Not determined	91.8
Sept. 24	P3B P3	0.05	96.0	
Sept. 24	P3B P3	0.10	83.6	Not Applicable
Sept. 24	P3B P3	0.30	47.4	

(*) The actual values are greater than 100%. This possibly results from: 1) errors during data collection (reading of concentrations); 2) uncertainties related to the exact values of the isotopic fractionation factors associated with microbial oxidation, α_{ox} and/or with gas transport, α_{trans}.

Table 2 – Methane and air loadings during column tests

Column	Day	CH_4 loading g CH_4 m^{-2} d^{-1} (ml/min)	Air loading g O_2 m^{-2} d^{-1} (ml/min)
PMOB 2	1	8 (0.2)	200 (9.2)
	15	68 (1.3)	890 (40.5)
	20	100 (1.9)	1244 (54.2)
	28	125 (2.4)	1848 (86.4)
	36	78 (1.5)	2734 (125.0)
PMOB 3B	1	8 (0.2)	200 (9.2)
	15	65 (1.2)	550 (25.5)
	17	98 (1.9)	670 (30.0)
	28	125 (2.4)	988 (45.2)
	43	75 (1.4)	1393 (65.9)

Table 3 – Initial and final degrees of saturation of column tests

Substrate	Depth (m)	Degree of saturation (S_r)	
		Initial (%)	Final (%)
PMOB 2	0 – 0.10	68.7	83.2
	0.10 – 0.20		74.9
	0.20 – 0.30		73.5
PMOB 3B	0 – 0.10	41.0	53.9
	0.10 – 0.20		41.8
	0.20 – 0.30		58.4