FINAL REPORT

Methane Oxidation: Field-Scale Test Sections Experiment
Riverbend Landfill, Oregon

By

Tarek Abichou, Ph.D., P.E.
Jeff Chanton, Ph.D.
Florida State University

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EXECUTIVE SUMMARY AND MAJOR FINDINGS

The U.S. EPA greenhouse reporting rules provide an option for site-specific methane oxidation values other than the default value of ten (10) percent. Methane oxidation values are now assigned to be 0, 10, 25, or 35 % depending on the methane flux in grams per square meter per day (g/m²/d) and on the extent and type of landfill cover. A few important issues in the new rule, however, have made it difficult for landfills to actually use these higher than the default 10% oxidation values.

Florida State University, in collaboration with Waste Management Inc., performed a field study to measure methane oxidation in soil covers at the Riverbend landfill in Oregon. The study provided a unique opportunity to monitor methane flow and methane oxidation through different soil profiles at a large scale and under field conditions. To the best of our knowledge, this study is also the only study in which LFG was introduced below soil profiles at a controlled rate. Test pads were constructed as lysimeters with a geomembrane at their bottom. LFG was continuously introduced at the bottom of the 10 x 20 m test pads and through a gas distribution layer placed above the geomembrane. The gas distribution layer consisted of a 15 cm thick gravel layer covered with a non-woven geotextile. A perforated PVC pipe network was also embedded in the gravel layer to distribute LFG across the bottom of the test pads.

The soil profile in Test Pad 1 consisted of a 45 cm (18 inches) thick layer of local soil (silt loam) to simulate an interim cover. This thickness is widely used in most states. The soil profile in Test Pad 2 consisted of a layer of 15 cm (6 inches) of local soil overlain with 30 cm (12 inches) of fresh mulch. Test Pad 2 was constructed to simulate a different type of interim cover (a daily cover overlain by 30 cm of mulch) for erosion control and possible additional oxidation of methane. In Test Pad 3, 15 cm (6 inches) of local soil was placed to simulate a daily cover. Note
that due to excessive soil erosion, Test Pad 3 was abandoned 3 months after construction. After completion of test pad construction, LFG was obtained from a gas header and continuously introduced into the bottom of each test pad. The gas inflow rate was maintained at a low rate so that methane loadings into the soil profile of the test pads were in the range of three methane loadings listed in the U.S.EPA GHG Reporting Rule (0-10 g/m²/d, 10 -70 g/m²/d, more than 70 g/m²/d).

An extensive program was designed to monitor the mass balance of methane through each test pad. Site visits and methane emissions and oxidation monitoring started on July 15th 2015 and continued until March 2017. Weekly and then monthly surveys of the composition of the gas below each test section was performed using a field LFG analyzer, and the concentrations of CH₄, CO₂, and O₂ were recorded. The flow rate and composition of the landfill gas entering each test pad, was also monitored on a weekly and then monthly basis. Three flux measurements were performed during each monitoring event. Flow into the test pads was regulated through the use of one valves and estimated by measuring the pressure drop across the orifice plates. The flow rate was then combined with methane concentration at the orifice plate to obtain the mass flux of methane entering each Test Pad. A dynamic chamber technique was used to determine the methane loading into the soil profile (from the gravel layer) at five locations in each test pad, using chambers permanently installed at the soil-gravel interface. Methane surface emission rates from the test pad surfaces were measured using static chambers located on the surface, right above the buried dynamic chambers. Gas samples from the inflow, the gravel layer, and the emitted gas were also obtained and shipped to the laboratory for composition and stable isotopic analysis.

The methane, oxygen and carbon dioxide concentrations in the injected LFG varied from 23.4% to 62.9%, for methane, 0.2% to 13.1% for oxygen and 14.2% to 38.1% for carbon dioxide.
Even though a gas distribution layer was placed at the bottom of each test pad, uneven distribution of methane concentration was observed in the gravel layer. The LFG tended to stay at the elevated end of the test pads and near the point where it was injected.

Methane mass balance monitoring was used to assess and delineate the extent of methane oxidation (in the different vertical layers of the test pads)

- The oxidation occurring in the soil cover layer is referred to as “in-soil oxidation. The in-soil oxidation was determined by comparing the surface (soil to air) fluxes measured by the static chambers to the bottom fluxes (from the gravel to the soil layer) measured by the dynamic chambers. Two approaches were then used to determine the in-soil oxidation.
  - The first approach was to determine the in-soil oxidation at each of the five discrete monitoring stations by comparing the individual measured discrete bottom and top fluxes at that station (using the buried and the surface flux chamber data). The five calculated levels of oxidation at each station were then averaged for that monitoring event, as an estimate of the average oxidation for the test pad during that event. This approach is referred as the “discrete approach”. The stable isotopes samples obtained from the dynamic chamber and the static chamber at the same monitoring station were then used to provide another estimate of methane oxidation at that discrete location.
  - The second approach consisted of using the geometric mean of the five measured bottom and top fluxes first, and then calculating the level of oxidation by comparing the geometric mean of the bottom fluxes and the geometric mean of the top fluxes. This approach is referred to as the “geospatial approach”. The isotope analysis for this approach is not applicable.
The discrete and geospatial approaches can be considered as the possible range (upper and lower limits) of actual methane field oxidation.

- The oxidation occurring in the gravel layer is referred to as in-gravel layer oxidation. The in-gravel methane oxidation was determined by comparing the overall bottom flux of methane out of the gravel layer into the soil profile (determined by the geometric mean of five dynamic flux chambers during each monitoring event) multiplied by the area of the test pad (10 by 20 m) and the mass flow rate of methane into the test pad (calculated by multiplying the measured flow rate by methane concentration at the orifice plate of the injected gas). The stable isotopes from each dynamic chamber are also compared to that of the sample from the injected gas to obtain five estimates of in-gravel methane oxidation during each monitoring event. The in-gravel stable isotope based oxidations estimates are averaged as an independent estimate of oxidation in the gravel layer.

- The total oxidation is the oxidation that occurred within the entire test pad and was obtained independently by comparing the geometric mean of the surface emission fluxes multiplied by the area of the test pad and the mass flow rate of methane into the test pad.

**In-Soil Methane Oxidation**

During the entire monitoring period and using a cumulative approach, 4.3 tons of methane were loaded into the bottom of the soil profile in Test Pad 1. Some 1.3 tons of methane were emitted from the surface of Test Pad 1. The soil profile in Test Pad 2 received 4.4 tons of methane of which 2.0 tons were emitted from the surface. Using these overall numbers, the 30 cm thick soil in Test Pad 1 was able to oxidize 3.0 tons of methane which corresponds to an overall in-soil oxidation of 70%. The 15 cm soil overlain with 30 cm mulch, in Test Pad 2 oxidized 2.4 tons of methane which corresponds to 56%.
Using the discrete approach mass balance, the in-soil percent methane oxidation during the monitoring period varied from 50 to 99% and averaged 76% for Test Pad 1. In Test Pad 2 the discrete mass balance approach yielded percent oxidation varying from 52 and 98% and an average of 76%. Using the discrete approach, the in-soil oxidation rate varied from 0.75 to 52 g/m²/d and averaged 22.7 g/m²/d for Test Pad 1. The oxidation rate in Test Pad 2 varied from 1 to 39 g/m²/d and averaged 27 g/m²/d. Using the stable isotopes analysis, percent in-soil methane oxidation varied from 20 to 65% and averaged 38%, for Test Pad 1. The in-soil percent methane oxidation during the monitoring period varied from 15 to 70% and averaged 34%, in Test Pad 2. Consistent with the literature and with the EPA rules, percent methane oxidation in the soil layer was found to decrease with increasing methane loading flux. The in-soil methane oxidation, measured during this study, was higher than the EPA GHG rule values of 10, 25, and 35% for the corresponding levels of methane loading in the rule.

In-Gravel Methane Oxidation

During the entire monitoring period, 19 tons of methane were loaded into the gravel layer of Test Pad 1. Some 4.3 tons of methane were delivered by the gravel layer to the soil above it. The gravel layer in Test Pad 2 also received 19 tons of methane of which 4.4 tons were transferred to the soil profile. Using these overall numbers, the gravel layer and the geotextile in Test Pad 1 oxidized 14.7 tons of methane which corresponds to an overall in-soil oxidation of 77% of the injected gas. The gas distribution layer in Test Pad 2 oxidized 14.6 tons of methane which corresponds to 76% of the injected gas.

The discrete in-gravel percent oxidation varied from 56 to 100% for Test Pad 1 and from 37 to 100% for Test Pad 2. The average in-gravel methane oxidation during the entire monitoring period was 71 and 79% for Test 1 and Test Pad 2, respectively. The deep or in-gravel oxidation
rate varied from 24 to 264 g/m²/d for Test Pad 1 and from 25 to 288 g/m²/d for Test Pad 2. Using the stable isotopes analysis, the in-gravel percent methane oxidation during the monitoring period varied from 14 to 76% and averaged 33% for Test Pad 1. The isotopes analysis from Test Pad 2 indicated that in-gravel methane oxidation varied from 9 to 48% and averaged 27%.

**Total Methane Oxidation**

During the entire monitoring period, 19 tons of methane were loaded into Test Pad 1. Some 1.3 tons of methane were emitted. Test Pad 2 also received 19 tons of methane of which 2 tons were emitted. Using these overall numbers, Test Pad 1 oxidized 17.7 tons of methane which corresponds to an overall in-soil oxidation of 92% of the injected gas. Test Pad 2 oxidized 17 tons of methane which corresponds to 89% of the injected gas. Using the discrete approach (average of 5 monitoring stations), the total percent oxidation varied from 63 to 100% for Test Pad 1 and from 71 to 100% for Test Pad 2. The total methane oxidation rate ranged from 42 to 296 g/m²/d for Test Pad 1 and from 56 to 298 g/m²/d for Test Pad 2.

**Incubation Experiments**

The highest measured value of maximum methane oxidation capacity, $V_{\text{max,Lab}}$, was 170 nmoles /kg dry soils/s for Test Pad samples. The minimum value of $V_{\text{max,Lab}}$ was around 80 nmoles /kg dry soils/s for Test Pad 1. Test Pad 2 had lower oxidation capacity in the lab. Samples from Test Pad 2 had $V_{\text{max,Lab}}$ from 100 to 20 nmoles/kg dry soil/s. These results underscore the spatial variability of methane oxidation capacity measurement even at a relatively small distance (chambers are less than 5 m apart). All samples had a methane half saturation constant, $K_{m,Lab}$, of around 1%.
SECTION 1

BACKGROUND

Previously, the principal investigators on this study reported methane oxidation in landfill covers measured by stable isotope analyses for over 37 seasonal sampling events at 20 different landfills which had intermediate covers (Chanton et al., 2011). The study spanned four years and five climate types. These data showed an inverse relationship between methane loading and fractional CH₄ oxidation. The U.S. EPA used these data and other values from the literature to support a change in HGH rule to allow methane oxidation to vary as a function of methane loading. The three test sections, constructed in this study, represented a unique opportunity to evaluate these new guidelines where LFG is being introduced into soil profiles at a controlled rate (Field Scale).

The EPA GHG reporting rule provides an option for site-specific methane oxidation values other than the default value of ten (10) percent. Under this option, methane oxidation values are now assigned to be 0, 10, 25, or 35% depending on the methane flux in grams per square meter per day (g/m²/d) and on the landfill cover type. A few important issues in the new rule, however, make it difficult for landfills to actually use these higher than the default 10% oxidation value. First, the EPA new rule assigns a ZERO oxidation value for “landfills that have a geomembrane (synthetic) cover with less than 30 cm (12 inches) of cover soil for the majority of the landfill area containing waste.” Second, The higher oxidation values of 35% can only be used for landfills with a methane flux rate less than 10 grams per square meter per day (g/m²/d). The higher value of 25% can only be used when the methane flux rate is between 10 and 70 grams per square meter per day (g/m²/d). A value of 10% oxidation is assigned for landfills with a methane flux rate higher than 70 grams per square meter per day (g/m²/d).
Florida State University teamed up with Waste Management Inc. for a field study to measure field-scale methane oxidation in soil covers at the Riverbend landfill in Oregon to address the limitations of the newly published EPA GHG reporting rule.

The methane oxidation capacities of the soils in the three test sections were also evaluated in incubation experiments in our laboratories. The methane loadings to the test sections were controlled to cover the range of methane loading associated with the recently published EPA rule on methane oxidation as follows:

- Below 10 g/m²/day
- 10 to 70 g/m²/day
- Above 70 g/m²/day
SECTION 2

METHODS AND MATERIALS

2.1 Site Description and Testing Set-Up

Three 20 meter by 10 meter (65 ft x 35 ft) test pads, shown in Figure 2.1, were constructed at Riverbend Landfill near the city of McMinnville, Oregon. The landfill provides waste disposal and recycling services for all of Yamhill County and neighboring areas, including metropolitan Portland. The three test beds were constructed similarly. The only variations in construction were the type of cover soils used in each test pad, and the thickness of each test pad. Figure 2.2 shows a cross-section of the installed test pads, and Figure 2.3 shows a plan view of the installed test pads.

Figure 2.1. View of the Testing Site
Figure 2.2. Test Pad Cross-Section View

Figure 2.3. Test Pad Layout Plan View
Construction of each test pad occurred in five steps. Step 1 involved properly preparing the subgrade underneath each test pad. Step 2 consisted of constructing two berms at the upper and lower sides of each test pad. In Step 3, a 60 mil high density polyethylene (HDPE) liner was placed at each test pad’s base (Figure 2.4). Step 4 consisted of overlying three inches of gravel on top of the HDPE liner, and then installing a 3.8 centimeter polyvinyl chloride (PVC) gas pipe on top of the gravel that runs the full length of the test pads. This was done to allow for LFG transport throughout the test pads (Figure 2.5). This middle pipe was also connected to a condensate and leachate drain at the low end of each test pad (Figure 2.3). In addition to the middle pipe, four 7.6 meter-long horizontal perforated pipes were then attached to the middle pipe, equally spaced (4.6 meters apart laterally) to allow for enhanced LFG dispersion (Figure 2.2). In Step 5, three more inches of gravel were placed above the gas pipes. The gravel was then covered by a geotextile filter fabric (Figure 2.6). Finally, Soil was then placed on top of the geotextile in accordance with the thickness of each test pad, as shown in Figure 2.7, which represents each of the three completed test pads. Note that, on the sides of each test pad, the soil extended 1 m beyond the edge of the geomebrane, geotextile and gravel layers and then slopes back to original ground level at 2:1 slope. The soil on the sides of the test pads was placed to contain flow through the sides and force the flow of gas vertically. A field FID was used to verify that no LFG is escaping from the edges of each test pad.
Figure 2.4. HDPE Liner and Gravel Layer Construction

Figure 2.5. Installation of PVC for Gas Distribution in the Gravel Layer.
Test Pad 1 consisted of 45 cm (18 inches) of local soil, silt loam, (above the geotextile), which was placed in one lift. The soil was lightly compacted with small dozer to simulate placement of soil cover in an operation landfill. Test Pad 1 was constructed to simulate a long term interim cover, widely used in most states. In Test Pad 2, 15 cm (6 inches) of local soil, as daily cover, was overlain with 30 cm (12 inches) of fresh mulch. Test Pad 2 was also constructed to
simulate a long term interim cover, a daily cover overlain by 30 cm (12 inches) of organics for possible additional oxidation of methane. In Test Pad 3, 15 cm (6 inches) of local soil was placed. Test Pad 3 simulated a daily cover. Note that due to excessive soil erosion (due to heavy rainfall) of the 15 cm of soil layer, Test Pad 3 was abandoned after 3 months of service. As mentioned earlier, all three test pads contained a gravel layer, installed as a gas distribution layer below the soil cover.

After completion of test pad construction, LFG was obtained from a gas header (Figure 2.8) and continuously introduced and pumped by a regenerative blower (GAST R6P155Q-50) into the bottom of each test pad (Figure 2.9). The gas inflow rate was maintained low so that methane loadings into each test pad stays in the same range as discussed in the U.S.EPA GHG rule (0-10, 10-70, and higher than 70 g/m²/d). Flow into each test pad was regulated through the use on-way valve and estimated using an orifice plates (both installed at the entrance of gas into each test pad).
2.2. Field Monitoring of Test Pads

An extensive monitoring program was designed to determine the mass balance of methane through each test pad. Site visits and methane emissions and oxidation monitoring of the test sections started on July 15th 2015. Weekly and then monthly survey of the composition of the gas below each test section was performed using a field LFG analyzer (GEM 5000, Lantec), where the concentrations of CH$_4$, CO$_2$, and O$_2$ were recorded. The flow rate of landfill gas, along with the composition of the landfill gas entering each test section, was also monitored on a weekly and then monthly basis. Flow into the test pads was regulated using a one-way valve estimated through the use of orifice plates by measuring the pressure drop across the orifice plate. The dynamic chamber technique was used to determine the methane loading into the bottom of each test section (from the gravel to the soil profile above it) at five stations in each test pad. Methane surface emission rates from the test pad surface is being measured using static chambers. LFG samples
were also obtained and shipped to the laboratory for gas composition and Stable Isotopes analysis. Figure 2.10. shows a sketch of the different components of the methane mass balance monitoring component and techniques used in this study.

![Figure 2.10. Sketch of Methane Mass Balance Monitoring](image)

2.2.1. Total Methane Loading into Test Pads

The total flow of landfill gas into the bottom of the test pad was estimated by measuring the pressure drop across the orifice plate (Figure 2.11). Flow into the test pads was regulated through a one-way valve and estimated by measuring the pressure drop across an orifice plate installed at the entrance pint of LFG into the test pads using a field LFG analyzer (GEM 5000, Lantec). The LFG flow rate into the bottom of the test pad was combined with the methane content to provide a volumetric flux. Gas temperature was used to convert the volumetric flux into a mass flux of methane into each test pad.
2.2.2. Gas Concentration in Gravel Layer

Monthly surveys of the concentrations of CH₄, CO₂, and O₂ in the gas below each test section were performed using a field gas analyzer (GEM 5000, Landtec) shown in Figure 2.12. Gas was also collected through a collection connected to a buried chamber sitting on top of the nonwoven geotextile installed on top of the gravel layer. Gas was collected using a plastic syringe and transferred to a vacuumed vial and sent to the laboratory (Florida State University) for analysis.
2.2.3. Bottom Flux Measurements

The dynamic chamber technique was used to determine the methane loading from the bottom of each test section (the gravel layer) into the soil profile above it at the five stations in each test pad shown in Figure 2.13 and Figure 2.14. Two surface ports were connected to each buried chamber. Nitrogen was swept through each buried chamber using one of two surface ports. The other surface port was left open and used as the outflow for the swept nitrogen/gas from the buried chamber. The flow rate of nitrogen was measured using a flow meter. Each chamber was swept with nitrogen for at least 30 minutes at a flow rate of at least 4 to 8 L/min. The volume of swept nitrogen corresponded to at least 30 to 60 times the volume of the buried chamber. Once the concentration of methane (in the outflow port from the buried chamber) is determined, it was combined with the flow rate of the swept nitrogen (measured by the flow meter) to determine the methane loading flux in g/m$^2$/day into the bottom of the soil profile using Eq. 2.1.

$$f = C \cdot F / A \quad (2.1)$$

Where $C$ is the measured methane concentration (g/m$^3$), $F$ is the measured flow rate of injected nitrogen (L/d) and $A$ is the area of the dynamic chamber (m$^2$).
Figure 2.13. Installation of Dynamic Chamber

Figure 2.14. Sweeping of Nitrogen into Dynamic Chamber Through One Surface Port and Sampling of Outflow from the Buried Chamber from the Other Surface Port
2.2.4. Surface Flux Measurements

Methane surface emission rates from the test pad surface were measured using static chambers as shown in Figure 2.15. The chamber dimensions are 0.63 x 0.63 x 0.2 m (covering an area of 0.4 m²) and contain a small fan to circulate air inside the chamber. Chambers were sealed to the ground by firming soil around the outside or by clamping them to pre-installed collars. Methane samples were collected from each chamber immediately after sealing it to the ground (time = 0) and after 5, 10, 15, 20, and 25 minutes using 60 mL disposable syringes (Becton, Dickinson, and Co.) fitted with plastic stopcocks (Cole Parmer Instrument Co.). Samples were injected into sealed glass vials and transported to our laboratory for further analyses.

Figure 2.15. Methane Surface Emissions Monitoring Using Static Chambers
The methane top flux was determined from the slope of concentration data (C in ppmv) plotted versus elapsed time (t in minutes). A linear fit was used to determine the slope as shown in Figure 2.16, in which dC/dt is the slope of the fitted line.

![Figure 2.16. Top Flux Development](image)

Once the slope is determined, CH₄ fluxes are calculated using Equation 2.2 (Barlaz et al., 2004).

$$f = \frac{V dC}{Adt} \times \frac{273}{T + 273}$$  \hspace{1cm} (2.2)

Where $f$ is the methane flux (g/m²/d), $V$ is the chamber headspace (m³), $A$ is the cross-sectional area of the chamber (m²), $\frac{dC}{dt}$ is the slope defined by Figure 2.17 converted into g/m³/d and $T$ is the temperature in the chamber.
2.3. Laboratory Analysis

All samples taken in the field were transported to the laboratory where the LFG samples were analyzed on a gas chromatograph equipped with a flame ionization detector as shown in Figure 2.17.

![Lab Analysis with Gas Chromatograph](image)

Figure 2.17. Lab Analysis with Gas Chromatograph

Stable isotope tracing was used to quantify the oxidation of methane in the soil profile. Stable isotopic ratios were determined using a Hewlett Packard gas chromatograph coupled via a combustion interface to a Finngan Mat Delta S isotope ratio mass spectrometer (GC-IRMS). Stable isotopic ratios for the anoxic gases and soil gas profile were determined using direct injection on the GC-IRMS. For methane concentrations below 1%, concentrations were determined on a Shimadzu 14A gas chromatograph with a flame ionization detector and a methanizer, a 1 mL sampling loop, and a 2-m 0.32 cm diameter stainless steel column packed with carbosphere.

2.4. Incubation Study to determine $V_{\text{max}}$ and $K_{\text{m}}$

An incubation study was performed to determine the maximum reaction rate ($V_{\text{max}}$, nmol g$^{-1}$ h$^{-1}$) and the Michaelis constant ($K_{\text{m}}$, nM) for soil samples from each test pad, as shown in Eq. 2.3:
\[ v = \frac{V_{\text{max}} S}{K_m + S} \]  

(2.3)

where \( v \) is the reaction rate (nmol g\(^{-1}\) h\(^{-1}\)) and \( S \) is the dissolved methane concentration (nM).

Four 1-L flasks of the soil and mulch samples delivered to our laboratory were incubated (three flasks from each soil). The flasks were dosed with 6% \( \text{CH}_4 \) in air weekly for a month before the experiment to increase the population of active methanotrophs. At the beginning of the experiment air in the flasks was replaced with 24% \( \text{CH}_4 \) in air. The flasks were incubated at 22\(^\circ\)C and six samples were taken over 23 hours after restoring the flasks to atmospheric pressure with oxygen before each sampling. Methane concentrations were determined by injecting a subsample from the vials into a TCD-GC (Shimadzu GC-8A with CRT 1 column). Methane gas concentrations were converted to molar concentration using Henry’s law and plotted against \( v \), the reaction rate. Equation 2.3 was fitted to the data by the least-squares-method.

### 2.5. Data Analysis

Eventhough a gravel layer and geo-composite were used as a gas distribution layer at the bottom of each test pad, methane concentrations in the gravel layer varied greatly, which indicated uneven distribution of landfill gas underneath the soil layer. Additionally, oxygen was present (measured) in the gravel/geo-composite layer and geotextile below the test pads, and therefore methane oxidation was possible in the soil profile and in the gravel-geocomposite layer. Therefore, the collected monitoring data was used to estimate different methane oxidation. The oxidation occurring in the gravel layer is referred to as “in-gravel oxidation”. The deep methane oxidation was determined by comparing the bottom methane loading flux of methane from the gravel into the soil (determined by the dynamic flux chamber) and the inflow of the injected gas (measured by the orifice plate at the entrance of gas into the test section). The oxidation occurring in the soil...
cover layer is referred to as “in-soil oxidation”. The in-soil oxidation was determined by comparing the top fluxes measured by the static chambers to the bottom methane loading fluxes measured by the dynamic chambers. Finally, an estimate of total oxidation occurring within the test pads was obtained independently by comparing the top emission fluxes to the inflow of methane into the test pads (using the orifice plate measurement). Figure 2.18 shows more details about the different methane oxidations estimated using our monitoring data. Stable Isotopes from the injected gas, gas within the gravel layer, and gas from the surface of the test pads were also used to provide another independent estimate of the in-gravel, in-soil, and total methane oxidation, in accordance with Chanton and Liptay (2000).

2.5.1. In-Gravel Oxidation

The in-gravel methane oxidation occurring within the gravel layer could be determined based on the bottom flux given by the dynamic flux chamber and the inflow of the injected gas. The oxidation rate (g/m²/d) is given by Equation 2.4. The oxidation capacity (%) is given by Equation 2.5.

\[
OX_{RATE,DEEP} = F_{INFLOW} - F_{BOTTOM} \tag{2.4}
\]

\[
OX_{PERCENT,DEEP} = \frac{F_{INFLOW} - F_{BOTTOM}}{F_{INFLOW}} \tag{2.5}
\]

Where \( F_{INFLOW} \) is the methane inflow (g/m²/d) and \( F_{BOTTOM} \) is the bottom methane flux (g/m²/d).
2.5.2. In-Soil Oxidation

Similar to the in-gravel oxidation term, the in-soil oxidation was determined by comparing the top fluxes measured by the static chambers to the bottom flux measured by the dynamic chambers. The top oxidation rate (g/m²/d) is given by Equation 2.6 and the top oxidation potential (%) is given by Equation 2.7.

\[
OX_{RATETOP} = F_{BOTTOM} - F_{TOP}
\]  

(2.6)
\[ OX_{\text{PERCENT\_TOP}} = \frac{F_{\text{BOTTOM}} - F_{\text{TOP}}}{F_{\text{BOTTOM}}} \]  

Where \( F_{\text{TOP}} \) is the methane top flux (g/m\(^2\)/d) and \( F_{\text{BOTTOM}} \) is the bottom methane flux (g/m\(^2\)/d).

### 2.5.3. Total Oxidation

The total oxidation is the combination of the top and deep oxidation that occurred within the test pads. The total oxidation rate (g/m\(^2\)/d) is given by Equation 2.8 and the total oxidation potential (%) is given by Equation 2.9.

\[ OX_{\text{RATE\_TOTAL}} = F_{\text{INFLOW}} + F_{\text{TOP}} \]  

\[ OX_{\text{PERCENT\_TOTAL}} = \frac{F_{\text{BINFLOW}} - F_{\text{TOP}}}{F_{\text{INFLOW}}} \]  

### 2.5.4. Stable Isotopes

The \( \text{CH}_4 \) stable isotope ratio (\( \delta \)) of gas collected from the gravel layer and the surface gas is compared to that of the gas being pumped into the bottom of the test pads (IN FLOW) using the following general equation:

\[ \delta = 1000 \left[ \frac{R_{\text{SAM}}}{R_{\text{STD}}} - 1 \right] \]  

Where \( R_{\text{SAM}} \) is the \( ^{13}\text{C}/^{12}\text{C} \) ratio of the sample, and \( R_{\text{STD}} \) is the corresponding ratio for standard Vienna Peedee Belemnite (Chanton et al. 2008).

The fraction (\( f_{\text{ox}} \)) of the \( \text{CH}_4 \) that is oxidized by the gravel layer was calculated using the following general equations:

\[ f_{\text{oxGravel}} = \frac{\delta_{\text{BOTTOM}} - \delta_{\text{INFLOW}}}{1000(\alpha_{\text{ox}} - \alpha_{\text{trans}})} \]  

and in the soil layer, the fraction oxidized was calculated using the following general equation:

\[ f_{\text{oxSoil}} = \frac{\delta_{\text{TOP}} - \delta_{\text{BOTTOM}}}{1000(\alpha_{\text{ox}} - \alpha_{\text{trans}})} \]
and in the whole system, total oxidation was calculated using the following general equation:

\[
\delta_{\text{TOP}} - \delta_{\text{INFLOW}} \\
\frac{1000(\alpha_{\text{ox}} - \alpha_{\text{trans}})}{
\]

(2.13)

Where \( \delta \) is the \( \delta^{13}C \) value of the CH\(_4\) injected, in the gravel, or emitted from the surface and \( \alpha_{\text{trans}} \) is the isotope fraction factor due to transport (\( \alpha_{\text{trans}}=1 \), since the LFG is being pumped into the test pads and the flow is mainly advective, De Visscher et al., 2003). \( \alpha_{\text{ox}} \) was assumed to be soil temperature dependent in accordance with Chanton and Liptay (2000) who incubated landfill soil and mulch, and found that for both materials, \( \alpha_{\text{ox}} \) decreased linearly with increasing temperature from an average of 1.039 at 8\(^{\circ}\)C to 1.027 at 35\(^{\circ}\)C. The same relationship was used for our study.
SECTION 3
RESULTS AND ANALYSIS

3.1. MONITORING RESULTS

Figure 3.1 shows the gas composition of the inflow LFG into Test Pads 1 and 2. Methane, oxygen and carbon dioxide concentrations vary respectively from 23.8 to 61.9, 0.2 to 12.9 and 14.5 to 36.3 % for Test Pad 1 and from 23.4 to 62.9, 0.5 to 13.1 and 14.2 to 38.1 % for Test Pad 2. Oxygen concentrations varied from 0.2 to 13.1% during the monitoring period. These findings indicate that enough oxygen is present in the pumped LFG that one should expect methane oxidation to occur in the gravel and geotextile layer below the test pads (distribution layer).

Figure 3.2 shows the resulting methane concentration below the soil profiles in Test Pads 1 and 2 at the five monitoring stations. Note that even though a gas distribution layer was placed at the bottom of each test pad, uneven distribution of methane is evident in Figure 3.2. Figure 3.3 shows an example of methane concentrations below Test Pad 1 during a monitoring event. It is clear that the methane was not evenly distributed below the test pad. The LFG tended to stay at the elevated upper end of the test pad. This uneven distribution was also caused by the fact that we wanted to keep the methane loading low to simulate low fluxes of methane into the soil profile (per the objectives of the study). Since the flow of gas was low, the injected gas stayed in the upper portions of the test pads.
Figure 3.1. LFG Composition of Inflow into Test Pads 1 (a) and 2 (b)
Figure 3.2. CH₄ Composition Below Test Pads 1 (a) and 2 (b) as Measured in Five Locations
As mentioned earlier, the methane mass balance analysis involved monitoring methane oxidation in the soil profile, in the gravel layer, and the total methane oxidation through the whole system (from the inflow to the surface emissions). Therefore, three methane oxidation terms were determined. The oxidation in the gravel layer is referred to as “deep oxidation” or in-gravel oxidation. The oxidation occurring in the soil cover layer is referred to as “top oxidation” or in-soil oxidation. The total oxidation between the inflow and the surface emissions from the test pads is referred to as “total oxidation”.

3.1.1. In-Soil Methane Oxidation

First, we present the results of methane oxidation in the soil layers of each test pad because the main study objective was to assess the oxidation of the two soil profiles of Test
Pad 1 and Test Pad 2 (45 cm of local soil for Test Pad 1 and 15 cm of soil + 30 cm of mulch for Test Pad 2). The measured in-soil methane oxidation should represent the minimum possible methane oxidation by the soil profile.

Figures 3.4 and 3.5 show the results obtained from the buried dynamic chamber and the surface static chambers for the monitoring period at one monitoring station of Test Pad 1. Figure 3.4 shows the measured top and bottom fluxes using static and dynamic chambers respectively at Test Pad 1, Station 2 (at the center of the test pad). During the entire monitoring period, the top methane flux varied from 0 to 12.18 g/m²/d and averaged 3.3 g/m²/d. The bottom flux varied from 0.85 to 26.12 g/m²/d and averaged 12.1 g/m²/d. Note that the top flux is lower than the bottom flux due to the oxidation process. Figure 3.5 shows the methane oxidation rate as determined by subtracting the surface emissions from the methane loading flux. Based on the mass-balance approach, percent oxidation and oxidation rate are then calculated and reported at that same monitoring station (Figure 3.5). The oxidation rate varied from 0.16 to 20 g/m²/d and averaged 8.8 g/m²/d while the percent oxidation varied from 19 to 100% and averaged 71% (Test Pad 1, Station 2). The stable isotope analysis on the gas samples from the bottom flux and the top flux at Test Pad 1, Station 2 yielded percent oxidation varying from 6 to 65% and averaging 32%. The results of the mass balance approach and the stable isotopes show that both techniques were able to assess methane oxidation at one station, knowing that the stable isotopes tend to underestimate methane oxidation (Chanton et al., 2011).
Figure 3.4. Methane Loading and Surface Emission Fluxes in Test Pad 1 – Station 2

Figure 3.5. Top Oxidation Rate and Percent Oxidation in Test Pad 1 – Station 2
The discrete approach was then used to determine the in-soil percent oxidation at each of the five discrete monitoring stations by comparing the individual measured discrete bottom and top fluxes at that station.

For the geospatial approach, used to determine the methane mass balance for each monitoring event, the geometric means of the five bottom methane loading fluxes and the five surface emission fluxes were calculated. The results of this approach are also shown in Figure 3.6. The geometric mean of the bottom and top fluxes were assumed to represent the average methane loading flux and surface emissions flux for the test pads. Abichou et al. (2011) and others used geospatial analysis of field measured surface emissions and suggested that the geometric mean is the best representation of central tendencies of measured methane fluxes. Figure 3.6 shows the geometric mean (average) top and bottom flux for the Test Pad 1. The average top and bottom fluxes vary from 0.01 to 30 g/m²/d and 0.76 to 70 g/m²/d respectively for Test Pad 1 and from 0.02 to 46 g/m²/d and 1 to 66 g/m²/d for Test Pad 2. The difference between the bottom and the top fluxes shown in Figure 3.6 is the methane oxidation rate within the soil profile. Using this approach, the in-soil oxidation rate varied from 0.75 to 52 g/m²/d for Test Pad 1 and from 1 to 39 g/m²/d for Test Pad 2 during the monitoring period (Figure 3.7). The average oxidation rate in Test Pad 1 is 24.9 g/m²/d. In Test Pad 2, the average oxidation rate was 22.8 g/m²/d.

The discrete approach and the geospatial approach provide the range or the maximum and minimum possible estimate of actual field methane oxidation. The maximum and minimum estimate of percent oxidation obtained using the mass balance approach are shown in Figure 3.7 for Test Pad 1 and Figure 3.8 for Test Pad 2, along with the results obtained using the stable isotope analysis. The in-soil percent oxidation varied from 42 to 99% for Test Pad 1 and from 5 to 98% for Test Pad 2. The in-soil average percent oxidation was 72% for Test Pad 1 and 63% for Test
Pad 2. These values are very similar to the results obtained with the discrete analysis and those obtained from stable isotope technique. Using the stable isotopes analysis, percent in-soil methane oxidation varied from 20 to 65% and averaged 38%, for Test Pad 1. The in-soil percent methane oxidation during the monitoring period varied from 15 to 70% and averaged 34%. As expected the stable isotope technique yields lower methane oxidation than the mass balance approach.

Finally, the total mass released from the gravel layers to the soil profile, and the mass of methane emitted from the surfaces of Test Pad 1 and Test Pad 2 were calculated to provide an overall estimate of methane oxidation in the soil profile. Over the entire monitoring period, 4.2 tons of methane were loaded into the bottom of the soil profile Test Pad 1. Some 1.3 tons of methane were emitted from the surface of Test Pad 1. The soil profile in Test Pad 2 received 4.2 tons of methane of which 2 tons were emitted from the surface. Using these overall numbers, the 30 cm thick soil in Test Pad 1 was able to oxidize 2.9 tons of methane which corresponds to an overall in-soil oxidation of 68%. The 15 cm soil overlain with 30 cm mulch, in Test Pad 2 oxidized 0.6 tons of methane which corresponds to 56%.
Figure 3.6. Geospatial Oxidation Rate and Percent Oxidation for Test 1(a) and 2(b).
Figure 3.7. Percent Oxidation Using Discrete and Geospatial Approach and Stable Isotopes for Test Pad 1

Figure 3.8. Percent Oxidation Using the Discrete and Geospatial Approach and Stable Isotopes for Test Pad 2
3.1.2. In-Gravel Methane Oxidation

As discussed earlier, the in-gravel oxidation that occurs in the gravel and geotextile gas distribution layer was separately estimated using the mass balance approach. This oxidation was also monitored and assessed by comparing the isotopes from the inflow and from the gas sampled from the gravel layer. Therefore, we have two independent measurements of in-gravel oxidation. This oxidation occurred due to O₂ being present in the landfill gas supplied to the test sites.

The in-gravel percent oxidation varied from 32 to 100% for Test Pad 1 (Figure 3.9) and from 35 to 100% for Test Pad 2 (Figure 3.10), using the mass balance approach. For the entire monitoring period, the average percent oxidation was 72% of the injected methane for both test pads. The mass balance approach also yielded deep or in-gravel oxidation rates varying from 24 to 264 g/m²/d (average = 137 g/m²/d) for Test Pad 1 and from 25 to 288 g/m²/d (average = 138 g/m²/d) for Test Pad 2.

Using the stable isotopes analysis, the in-gravel percent methane oxidation during the monitoring period varied from 14 to 76% and averaged 33% for Test Pad 1 (Figure 3.9). The isotopes analysis from Test Pad 2 indicated that in-gravel methane oxidation to vary from 9 to 48% and averaged 27% (Figure 3.10).

During the entire monitoring period, 19 tons of methane were loaded into the gravel layer of Test Pad 1. Some 4.3 tons of methane were delivered by the gravel layer to the soil above it. The gravel layer in Test Pad 2 also received 19 tons of methane of which 4.4 tons were transferred to the soil profile. Using these overall numbers, the gravel layer and the geotextile in Test Pad 1 oxidized 14.7 tons of methane which corresponds to an overall in-soil oxidation of 77% of the injected gas. The gas distribution layer in Test Pad 2 oxidized 14.6 tons of methane which corresponds to 76% of the injected gas.
Figure 3.9. Mass Balance and Isotope In-Gravel Percent Oxidation in Test Pad 1

Figure 3.10. Mass Balance and Isotope In-Gravel Percent Oxidation in Test Pad 2
3.1.3. Total Test Pad Oxidation

The total mass balance derived methane oxidation rate within these test pads was calculated by comparing the inflow to the average of the five measured surface emission fluxes and represents the whole methane oxidation throughout the gas distribution layers and the soil profile. The total mass balance percent oxidation varied from 63 to 100% and averaged 90% for Test Pad 1 and varied from 71 to 100% and averaged 90% for Test Pad 2 (Figure 3.11). The total mass balance methane oxidation rate ranged from 42 to 296 g/m²/d and averaged 163 g/m²/d for Test Pad 1. For Test Pad 2, the total mass balance methane oxidation rate varied from 56 to 298 g/m²/d and averaged 161 g/m²/d (Figure 3.11).

During the entire monitoring period, 19 tons of methane were loaded into Test Pad 1. 1.3 tons of methane were emitted. Test Pad 2 also received 19 tons of methane of which 2 tons were emitted. Using these overall numbers, Test Pad 1 oxidized 17.7 tons of methane which corresponds to an overall in-soil oxidation of 92% of the injected gas. Test Pad 2 oxidized 17 tons of methane which corresponds to 89% of the injected gas.
Figure 3.11. Average Total Percent Oxidation and Rate for Test Pad 1 (a) and Test pad 2 (b)
3.1.4. Incubation Results

One soil cover specimen from each static chamber location was tested. As described earlier, these samples were homogenized in the laboratory and their methane oxidation capacity was measured. The main objective of this test is to assess the spatial variability of the methane oxidation capacity ($V_{\text{max},\text{Lab}}$) and the methane half saturation constant ($K_{m,\text{Lab}}$) from one chamber to the next. The oxygen half-saturation constant was not measured in this study. The highest measured value of maximum methane oxidation capacity, ($V_{\text{max},\text{Lab}}$), was 170 nmoles/kg dry soils/s. The minimum value of ($V_{\text{max},\text{Lab}}$) was around 80 nmoles/kg dry soils/s for Test Pad 1. Test Pad 2 had lower oxidation capacity in the lab. Samples from Test Pad 2 had ($V_{\text{max},\text{Lab}}$) from 100 to 20 nmoles/kg dry soil/s. These results underscore the spatial variability of methane oxidation capacity measurement even at relatively small distances (about 5 meters apart). All samples had a methane half saturation constant, ($K_{m,\text{Lab}}$), of around 1%.

3.2. DISCUSSION OF RESULTS

The most important quantity monitored in this study is the in-soil methane oxidation since the EPA GHG rule concerns only landfill cover soil oxidation. The in-soil oxidation data from each of the mass balance approaches can be considered to bracket the range of actual field methane oxidation. In one approach (discrete approach), the in-soil oxidation rate and percent in-soil oxidation were calculated using the measured bottom and surface fluxes for each of the five monitoring stations and then averaged. In the other approach (geospatial approach), the measured five bottom and five top fluxes during a monitoring event are first averaged (geometric mean). The geometric mean of fluxes is then used to assign an oxidation rate and percent oxidation for the test pad for that day of monitoring. The two estimates of methane oxidation rate and percent oxidations
along with the one obtained using the stable isotope technique are shown in Figure 3.11 for Test Pad 1 and Figure 3.12 for Test Pad 2. These results indicate that the flux based estimates of methane oxidation are in the same range as those estimated using the stable isotope technique. The two independent field assessments of methane oxidation yielded evidence of methane oxidation higher than the ranges provided in U.S. EPA GHG reporting rule. Figure 3.12 and Figure 3.13 show the relationship between methane loading flux and percent oxidation as obtained from our study. Both soil profiles have oxidized more than the percent methane oxidation suggested by the EPA rules.

Figure 3.12. Average In-Soil Percent Oxidation versus Methane Loading Flux as Determined by Mass Balance and Stable Isotopes for Test Pad 1
Figure 3.13. Average In-Soil Percent Oxidation versus Methane Loading Flux as Determined by Mass Balance and Stable Isotopes for Test Pad 2
An extensive program was designed to monitor the mass balance of methane through each test pad. Site visits and methane emissions and oxidation monitoring started on July 15th 2015 and continued until March 2017. Weekly and then monthly surveys of the composition of the gas below each test section were performed using a GEM2000, and the concentrations of CH$_4$, CO$_2$, and O$_2$ were recorded. The flow rate and composition of the landfill gas entering each test pad, was also monitored on a weekly and then monthly basis. Three flux measurements were performed during each monitoring event. Flow into the test pads was regulated through the use of orifice plates and estimated by measuring the pressure drop across the orifice plate. The flow rate was then combined with methane concentration at the orifice plate to obtain the mass flux of methane entering each Test Pad. A dynamic chamber technique was used to determine the methane loading into the soil profile (from the gravel layer) at five locations in each test pad, using chambers permanently installed at the Soil-Gravel interface. Methane surface emission rates from the test pad surfaces were measured using static chambers located on the surface, right above the buried dynamic chambers. Gas samples from the inflow, the gravel layer, and the emitted gas were also obtained and shipped to the laboratory for composition and stable isotopic analysis.

**In-Soil Methane Oxidation**

During the entire monitoring period, and using a cumulative approach, 4.3 tons of methane were loaded into the bottom of the soil profile in Test Pad 1. Some 1.3 tons of methane were emitted from the surface of Test Pad 1. The soil profile in Test Pad 2 received 4.4 tons of methane of which 2.0 tons were emitted from the surface. Using these overall numbers, the 30 cm thick soil in Test Pad 1 was able to oxidize 3.0 tons of methane which corresponds to an overall in-soil
oxidation of 70%. The 15 cm soil overlain with 30 cm mulch, in Test Pad 2 oxidized 2.4 tons of methane which corresponds to 56%.

Using the discrete approach mass balance, the in-soil percent methane oxidation during the monitoring period varied from 50 to 99% and averaged 76% for Test Pad 1. In Test Pad 2 the discrete mass balance approach yielded percent oxidation varying from 52 to 98% and an average of 76%. Using the discrete approach, the in-soil oxidation rate varied from 0.75 to 52 g/m²/d and averaged 22.7 g/m²/d for Test Pad 1. The oxidation rate in Test Pad 2 varied from 1 to 39 g/m²/d and averaged 27 g/m²/d. Using the stable isotopes analysis, percent in-soil methane oxidation varied from 20 to 65% and averaged 38%, for Test Pad 1. The in-soil percent methane oxidation during the monitoring period varied from 15 to 70% and averaged 34%, in Test Pad 2. Consistent with the literature and with the EPA rules, percent methane oxidation in the soil layer was found to decrease with increasing methane loading flux. The in-soil methane oxidation, measured during this study, was higher than the EPA GHG rule values of 10, 25, and 35% for the corresponding levels of methane loading in the rule.

**In-Gravel Methane Oxidation**

During the entire monitoring period, 19 tons of methane were loaded into the gravel layer of Test Pad 1. Some 4.3 tons of methane were delivered by the gravel layer to the soil above it. The gravel layer in Test Pad 2 also received 19 tons of methane of which 4.4 tons were transferred to the soil profile. Using these overall numbers, the gravel layer and the geotextile in Test Pad 1 oxidized 14.7 tons of methane which corresponds to an overall in-soil oxidation of 77% of the injected gas. The gas distribution layer in Test Pad 2 oxidized 14.6 tons of methane which corresponds to 76% of the injected gas.
The discrete deep or in-gravel percent oxidation varied from 56 to 100% for Test Pad 1 and from 37 to 100% for Test Pad 2. The average in-gravel methane oxidation during the entire monitoring period was 71 and 79% for Test 1 and Test Pad 2, respectively. The deep or in-gravel oxidation rate varied from 24 to 264 g/m²/d for Test Pad 1 and from 25 to 288 g/m²/d for Test Pad 2. Using the stable isotopes analysis, the in-gravel percent methane oxidation during the monitoring period varied from 14 to 76% and averaged 33% for Test Pad 1. The isotopes analysis from Test Pad 2 indicated that in-gravel methane oxidation to vary from 9 to 48% and averaged 27%.

Total Methane Oxidation

During the entire monitoring period, 19 tons of methane were loaded into Test Pad 1. Some 1.3 tons of methane were emitted. Test Pad 2 also received 19 tons of methane of which 2 tons were emitted. Using these oval numbers, Test Pad 1 oxidized 17.7 tons of methane which corresponds to an overall in-soil oxidation of 92% of the injected gas. Test Pad 2 oxidized 17 tons of methane which correspond to 89% of the injected gas. The total percent oxidation varied from 63 to 100% for Test Pad 1 and from 71 to 100% for Test Pad 2. The total methane oxidation rate ranged from 42 to 296 g/m²/d for Test Pad 1 and from 56 to 298 g/m²/d for Test Pad 2.

Incubation Experiments

The highest measured value of maximum methane oxidation capacity, \( V_{\text{max,Lab}} \), was 170 nmoles /kg dry soils/s for Test Pad samples. The minimum value of \( V_{\text{max,Lab}} \) was around 80 nmoles /kg dry soils/s for Test Pad 1. Test Pad 2 had a lower oxidation capacity in the lab. Samples from Test Pad 2 had \( V_{\text{max,Lab}} \) from 100 to 20 nmoles/kg dry soil/s. These results underscore the spatial variability of methane oxidation capacity measurement even at relatively small distances.
(chambers are less than 5 m apart). All samples had a methane half saturation constant, $K_{m,Lab}$, of around 1%.
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