Available online at www.sciencedirect.com

Waste Management 26 (2006) 516–525

www.elsevier.com/locate/wasman

Methane mass balance at three landfill sites: What is the efficiency of capture by gas collection systems?

K. Spokas [a,*], J. Bogner [b], J.P. Chanton [c], M. Morcet [d], C. Aran [d], C. Graff [a], Y. Moreau-Le Golvan [e], I. Hebe [f]

[a] *University of Minnesota, Department of Soil, Water, and Climate, St. Paul, MN, USA*
[b] *Landfills+ Inc., Wheaton, Illinois and University of Illinois, Chicago, IL, USA*
[c] *Florida State University, Department of Oceanography, Tallahassee, FL, USA*
[d] *Centre de Recherches pour l'Environnement l'Energie et le Déchet (CReeD), Véolia Environnement, Limay, France*
[e] *COLLEX Pty Ltd., CReeD, Véolia Environnement, Pyrmont NSW, Australia*
[f] *Agence de l'Environnement et de la Maitrise de l'Energie (ADEME), French Agency for the Environment and Energy Management, Angers, France*

Accepted 29 July 2005
Available online 29 September 2005

Abstract

Many developed countries have targeted landfill methane recovery among greenhouse gas mitigation strategies, since methane is the second most important greenhouse gas after carbon dioxide. Major questions remain with respect to actual methane production rates in field settings and the relative mass of methane that is recovered, emitted, oxidized by methanotrophic bacteria, laterally migrated, or temporarily stored within the landfill volume. This paper presents the results of extensive field campaigns at three landfill sites to elucidate the total methane balance and provide field measurements to quantify these pathways. We assessed the overall methane mass balance in field cells with a variety of designs, cover materials, and gas management strategies. Sites included different cell configurations, including temporary clay cover, final clay cover, geosynthetic clay liners, and geomembrane composite covers, and cells with and without gas collection systems. Methane emission rates ranged from −2.2 to >10,000 mg CH_4 m^{-2} d^{-1}. Total methane oxidation rates ranged from 4% to 50% of the methane flux through the cover at sites with positive emissions. Oxidation of atmospheric methane was occurring in vegetated soils above a geomembrane. The results of these studies were used as the basis for guidelines by the French environment agency (ADEME) for default values for percent recovery: 35% for an operating cell with an active landfill gas (LFG) recovery system, 65% for a temporary covered cell with an active LFG recovery system, 85% for a cell with clay final cover and active LFG recovery, and 90% for a cell with a geomembrane final cover and active LFG recovery.

1. Introduction

When solid waste is buried in a landfill, the biodegradable fractions decompose via a complex series of microbial and abiotic reactions. Methane (CH_4), one of the terminal products, is formed by methanogenic microorganisms under anoxic conditions, either through the direct cleavage of acetate into CH_4 and carbon dioxide (CO_2) or the reduction of CO_2 with hydrogen. Large engineered landfill sites containing more than a 1 million m^3 of refuse with high percentages of biodegradable municipal solid waste can readily generate several hundred m^3 of landfill gas h^{-1}. Landfill gas, as recovered, is a water saturated biogas that consists of 50–60% (v/v) CH_4, 40–50% (v/v) carbon dioxide (CO_2), and numerous trace components. Because of its CH_4 content, landfill gas has a fuel value of 18–22 MJ m^{-3}.

Technology to recover and utilize landfill CH_4 using vertical wells or horizontal collection systems has been

---

[*] Corresponding author. Present address: Kurt Spokas, USDA-ARS, 803 Iowa Avenue, Morris, MN 56267, United States. Tel.: +1 320 589 3411x161; fax: +1 320 589 5787.
E-mail address: spokas@morris.ars.usda.gov (K. Spokas).

0956-053X/$ - see front matter © 2005 Elsevier Ltd. All rights reserved.
doi:10.1016/j.wasman.2005.07.021

commercialized for 30 yr, beginning with the first full-scale project at Palos Verdes, California (US) in 1975. Today there are more than 1100 full-scale landfill CH_4 recovery projects worldwide, including approximately 180 in Germany, 150 in the UK, 135 in Italy, and more than 350 in the US (Willumsen, 2003). Landfill CH_4 is used to fuel industrial or commercial boilers, to generate electricity on-site using internal combustion engines or gas turbines, and to produce a substitute natural gas suitable for compression or pipeline transport. Electrical projects range from smaller 30 kW microturbines to a large 50 MW steam turbine (located at Puente Hills Landfill, Whittier, California, US).

In addition to providing a local source of energy, the commercial recovery of landfill CH_4 decreases a source of atmospheric CH_4, the second most important greenhouse gas after CO_2 and responsible for approximately 40% of global warming over the past 150 years (Hansen et al., 1998). Since CH_4 has an atmospheric lifetime of about 10 yr and a global warming potential (GWP) 23 times higher than CO_2 (mole basis, 100 yr timeframe), reductions in individual CH_4 sources can decrease atmospheric concentrations within a decade (Houghton et al., 2001). Annually, about 500–600 terragrams (Tg) (1 Terragram = 10^{12} g) CH_4 are emitted to the atmosphere, with approximately 70% from several anthropogenic sources including rice cultivation, domesticated ruminant animals, biomass burning, natural gas leakages, coalbed CH_4, and landfills, with the remainder from natural wetlands (Matthews, 2000). Global landfill CH_4 emissions estimates have ranged from 9 to 70 Tg yr^{-1}, with the latest estimates in the range of 15–20 Tg yr^{-1} (Bingemer and Crutzen, 1987; Richards, 1989; Bogner and Matthews, 2003). Currently, estimates indicate that commercial landfill gas recovery projects recover more than 5 Tg worldwide, thus reducing atmospheric CH_4 contributions from landfill sources (Willumsen, 2003; Bogner and Matthews, 2003; Bogner, unpublished). Many countries are targeting landfill CH_4 recovery as a mitigation strategy to achieve national greenhouse gas reduction goals because landfills are concentrated sources of atmospheric CH_4 that are amenable to engineered gas extraction. In many developed countries, it is estimated that landfill CH_4 is the largest anthropogenic source of atmospheric CH_4. In developing countries, the annual volume of landfill CH_4 generated will increase significantly over the next decade as more controlled landfilling practices are expanded to deal with large quantities of solid waste in an environmentally acceptable manner, especially in rapidly growing mega-cities.

Even though there have been 30 yr of commercial landfill CH_4 recovery at hundreds of sites worldwide, there have been virtually no field studies to better constrain the CH_4 mass balance for landfill cells with differing designs and management practices. Typically, hydrologic investigations utilize water and contaminant mass balance to establish mass conservation and improve prediction of contaminant transport (e.g., Schroeder et al., 1994). CH_4 generated in landfills is partitioned into CH_4 recovered, emitted to the atmosphere, oxidized by methanotrophs, laterally migrated, and internally stored in the landfill volume (Bogner and Spokas, 1993), as follows:

$$\begin{aligned}\mathrm{CH_4\ generated} &= \mathrm{CH_4\ emitted} + \mathrm{CH_4\ oxidized}\\ &\quad + \mathrm{CH_4\ recovered(flared)}\\ &\quad + \mathrm{CH_4\ migrated} + \Delta\mathrm{CH_4\ storage}\\ &[\text{all units} = \text{mass time}^{-1}]. \qquad (1)\end{aligned}$$

Historically for commercial recovery projects, CH_4 generation at landfills has been modeled using a first order kinetic equation (Scheepers and van Zanten, 1994; Coops et al., 1995; Blaha et al., 1999) based on waste inputs, climate variables, and other factors. Modeling results are compared to actual recovery trends, and model parameters adjusted through the period of active gas extraction so that projections are consistent with actual recovery data. Through this process, gas recovery efficiencies have been typically estimated to be in the range of 50–75%, based on *measured* gas extraction rates divided by *modeled* gas generation rates. However, actual gas recovery efficiencies are dependent on quantification of all the pathways in Eq. (1).

The purpose of this paper is to summarize intensive field studies of the CH_4 mass balance for nine individual landfill cells at three landfills with well-defined waste inputs. The project cells were chosen to include a variety of designs, management practices, and meteorological conditions. Field measurements were conducted during short-term measurement campaigns to limit environmental variability and were supported by follow-up laboratory analyses and modeling.

2. Landfill methane mass balance study sites: project METAN

There were three ONYX (Veolia Environnement) landfill sites included in the METAN program (Fig. 1). The METAN program was established to study CH_4 mass balance in actual landfill settings and was conducted by CReeD (Veolia Environment). Partial financial support was provided by the French environment agency, Agence de l'Environnement et de la Maitrise de l'Energie (ADEME). Field sites are described below and site characteristics are summarized in Table 1.

2.1. Montreuil-sur-Barse landfill

This municipal solid waste (MSW) landfill is located near Troyes in northeast France (Fig. 1). Two experimental test cells filled from 1994 to 1999 were investigated as part of this program. The cover for the first cell consists of 30 cm of organic soil overlying a geosynthetic clay liner (GCL) (von Maubeuge, 2004) overlying a sand layer. The second cell has a clay cover consisting of 1 m of compacted clay overlain by a 30 cm organic soil layer (total cover

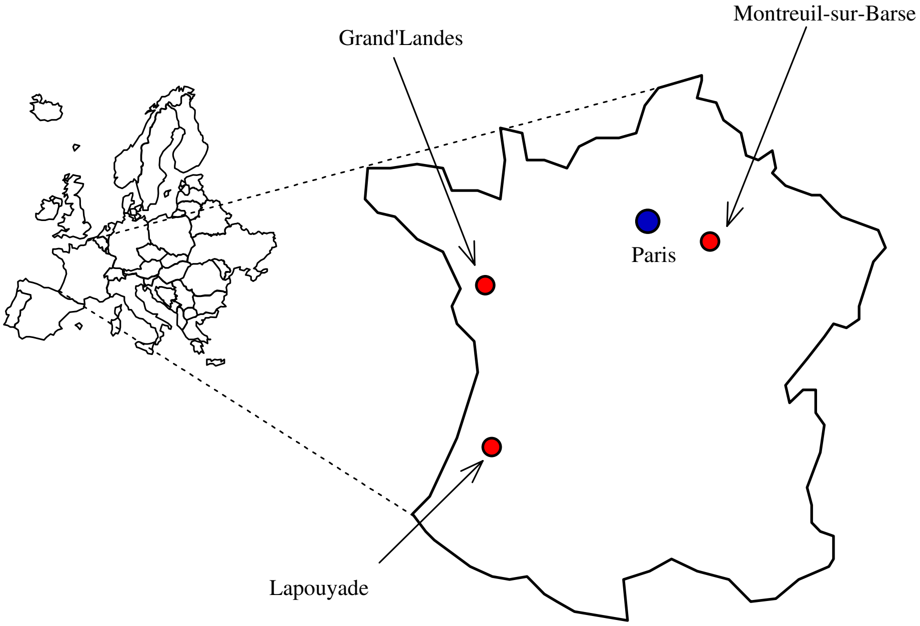

Fig. 1. Geographical location of investigated landfill sites in France.

Table 1
Summary of cells investigated in METHAN program

Field location	Cell area (m^2)	Cell volume (m^3)	Average waste thickness (m)	Waste in place (metric tons)	LFG recovery	Cover design specifics
Montreuil-sur-Barse						
Cell AS	2518	11,728	4.7	9542	✓	Final clay cover: 1 m clay, overlain by a 4 cm drainage geosynthetic fabric, capped with 30 cm of top soil
Cell BS	2516	10,853	4.3	9169	✓	Final GCL cover: GCL liner overlain by 4 cm drainage fabric and capped with 30 cm of top soil
Lapouyade						
Phase I Cells (A1–A8)	35,555	35,259,0	9.9	30,975,6	✓	Final clay cover: 1 m clay capped with 30 cm of top soil
Cell A8	6650	78,470[a]	11.8	46,686	✓	Temporary clay cover: 30 cm clay
Cell A10	4950	96,525[a]	15	68,124	No	Temporary clay cover: 30 cm clay
Grand'Landes						
Cell 25A	10,000	69,000	6.9	56,000	✓	Final clay cover: 1 m clay, overlain with 30 cm of top soil. Cell has vertical gas recovery wells
Cell 25B	10,000	59,000	5.9	52,000	✓	Final geomembrane with horizontal gas collection: 30 cm of gravel overlain by geomembrane cover with 30 cm of top soil. LFG recovery is achieved with a horizontal collection pipe in the gravel layer

[a] Denotes estimated since exact surveys were not conducted on the temporary cover cells.

thickness was 130 cm). Field measurements were conducted in February 2000.

2.2. Lapouyade landfill

Lapouyade is located near Bordeaux in southwestern France (Fig. 1) and the landfill has been operating since October 1996 receiving about 440 ton of waste d^{-1}. The areas investigated at this site included a final soil cover (with active gas recovery) and thin temporary soil cover (with and without gas recovery) (Table 1). The cells with gas recovery were investigated under different seasonal conditions (December 2000 and September 2001).

2.3. Grand'landes landfill

The Grand'Landes landfill is situated in western France near Nantes (Fig. 1). This landfill has been operating since 1989 and was closed in 2001. CH_4 mass balance studies were conducted on two experimental MSW cells with different LFG recovery strategies. The first cell has conventional vertical wells with compacted clay as a top cover. The second cell has an innovative gas collection system consisting of horizontal pipes in a high permeability gas collection layer (gravel) at the top of the cell; the gravel layer is overlain by a high density polyethylene (HDPE) geomembrane (Aran et al., 2002).

3. Methods

For each field campaign, all of the measurements took place within 2 weeks under similar meteorological conditions. This was to avoid large fluctuations in barometric pressure caused by major storm events and other meteorological conditions, which can greatly affect surface emissions (Galle et al., 2001; Czepiel et al., 2003). Below we describe the techniques used to quantify each CH_4 pathway.

3.1. CH_4 generation (landfill methane generation model: LMGM)

The modeling of landfill gas production has primarily remained focused on empirical relationships derived from laboratory decomposition studies or test cell projects. The model developed for these sites is based on a first-order degradation model with multiple waste-types as inputs. This type of model has been successfully validated at landfill sites with excellent agreement between measured and predicted gas generation rates (Coops et al., 1995). The amount of CH_4 generated was calculated from the following relationship (Oonk et al., 1993; Scheepers and van Zanten, 1994; Coops et al., 1995):

$$V = G\sum_{n=1}^{Z} A_n C_{org,n} k_n e^{-k_n t}, \tag{2}$$

where V is the predicted amount of CH_4 produced (m^3), G is the formation constant (0.94 m^3 CH_4 kg^{-1} organic carbon), A_n is the total amount of waste stream n present in refuse (kg), $C_{org,n}$ is the amount of organic carbon present in waste stream n, k_n is the half-life of waste stream n (yr^{-1}), t is the elapsed years from start of filling, and Z is the number of waste categories. Default values were taken from the Dutch work on modeling landfill gas for European landfills (Oonk et al., 1993; Coops et al., 1995). These studies found that the multiphase production models and the simpler first order degradation models compared favorably at multiple sites. Where additional detailed waste input data are available, the prediction improves. Each cell investigated in this study had detailed waste data available for CH_4 generation modeling. This enabled an accurate model to be customized for each site. At the first site, Montreuil-sur-Barse, there was also a detailed characterization of MSW for each of the test cells (Opsomer and Jannink, 1995). It was assumed that this characterization represented the average MSW composition for the other 2 landfills. Half-life decomposition times were divided into four categories: very slow (30 yr; textiles), slow (15 yr; paper), moderate (10 yr; fines), and fast (5 yr; labile organic material). The various waste categories and the organic carbon and half-life assignments are summarized in Table 2. Fines is the category assigned to waste that was too small to physically identify and labile organic materials include food, vegetable, and green wastes.

3.2. CH_4 recovery

Where CH_4 is recovered by an active system of vertical wells or horizontal collectors, recovery is directly quantified by mass flow measurements. The mass of CH_4 recovered is the most accurately quantified pathway of the CH_4 balance. CH_4 recovered is directly calculated from measured flow rates and CH_4 concentration data, assuming that the CH_4 concentration is constant between measurements and after correction to standard temperature and pressure.

3.3. CH_4 emissions

Point measurements of landfill CH_4 emissions can vary over seven orders of magnitude (0.0004–4000 g m^{-2} d^{-1}) (Bogner et al., 1997). CH_4 emissions from landfills can be controlled by active gas recovery systems and the natural methanotrophic attenuation potential of engineered soil covers (Christopherson et al., 2000; Bogner et al., 1997). Two different approaches were used in parallel to quantify surface emissions: an above-ground tracer method and flux chamber techniques.

3.3.1. Tracer method

Atmospheric tracer methods have been previously used to study whole landfill emissions (Trégourès et al., 1999;

Table 2
Waste characterization from Montreuil-sur-Barse

Waste type	Range	Value used (%)	Organic carbon	Half-life (yr)
Paper and cartons	13–26	19.5	40	15
Plastics	7–12	9.5	–	∞
Composites	3–4.5	3.75	–	∞
Textiles	3–18	10.5	20	30
Glass	6–12	9	–	∞
Labile organic material	12–24	18	80	5
Fines	16–20	18	60	10

Galle et al., 2001; Czepiel et al., 2003). The method in this study has been previously described by Trégourès et al. (1999). Briefly, this method relies on concurrent above-ground concentration measurements for CH_4 and an inert tracer gas (SF_6) released at a known rate upwind from the landfill. In this method the emissions from the landfill are obtained from measurements of methane and tracer concentrations in transects across the plume downwind from the landfill. By knowing the concentration of both CH_4 and SF_6, the flux rate of CH_4 from the landfill area can be determined using numerical dispersion modeling. This method averages fluxes across surface heterogeneities, but the exact footprint of the emission source may be difficult to determine and is dependent on meteorological conditions. Therefore, the footprint may not always coincide exactly with the landfill cell under investigation.

3.3.2. Flux chambers

A static recirculating chamber was used to measure CH_4 flux density from many small areas of the landfill surface ($0.25\ m^2$). This approach utilizes a pump with a flow rate of approximately $10\ l\ min^{-1}$ to circulate the enclosed chamber headspace to an outside loop. The rate of CH_4 enrichment in the loop is measured periodically using a laboratory gas chromatograph that is transported in a utility vehicle around the site (Trégourès et al., 1999). Resulting surface flux density is calculated from the change in chamber CH_4 concentration with time ($\Delta C/\Delta t$), chamber volume (V), and the area (A) by:

$$\text{flux} = \frac{V}{A}\left(\frac{\Delta C}{\Delta t}\right). \tag{3}$$

CH_4 flux density is typically measured in units of $g\ CH_4\ m^{-2}\ min^{-1}$. Continuous measurement of CH_4 allows rapid calculation of the CH_4 emission rate from the enclosed area. Measured emissions of CH_4 at chamber locations are the result of complex biological, chemical, and physical processes occurring within landfill cover soils, and there can be difficulties in the extrapolation of chamber measurements for large landfill areas, which are detailed elsewhere (Spokas et al., 2003). Chamber fluxes were measured on a 20×20 m grid pattern and spatially interpolated using inverse distance weighing (IDW).

3.4. CH_4 oxidation

Microbial CH_4 oxidation is carried out by methanotrophic bacteria. Methanotrophs are a class of methylotrophic bacteria that possess a specific enzyme (methane monooxygenase), which allows them to oxidize CH_4 to methanol (Anthony, 1982). Methanol is further degraded to CO_2. A major uncertainty in the CH_4 balance is determining the attenuation of CH_4 emissions by methanotrophic bacteria in the aerobic outer portions of the cover soil. Previous field efforts have shown that a significant portion (10–100%) of the CH_4 present in the cover is oxidized by indigenous methanotrophs (Whalen et al., 1990; Czepiel et al., 1996; Liptay et al., 1998; Chanton et al., 1999; Chanton and Liptay, 2000; Börjesson et al., 2001; Christopherson et al., 2001; Barlaz et al., 2004). In addition, field studies have shown that when methane supply from below is either blocked or consumed quantitatively, then these bacteria can oxidize atmospheric CH_4 as it diffuses into the soil from above, once the concentration of CH_4 is below atmospheric concentrations (Bogner et al., 1995). It should be noted that atmospheric concentrations at landfill sites can be significantly greater than background atmospheric concentrations (≅1.8 ppmv) (Barlaz et al., 2004).

In this study, methane oxidation was determined using a stable carbon isotope technique. Since there is a preferential oxidation of ^{12}C over ^{13}C in CH_4 by the methanotrophs (Barker and Fritz, 1981; Coleman et al., 1981), it is possible to determine the oxidized fraction by quantifying the change in $\delta^{13}C$ of CH_4 between the anaerobic zone and the surface (Chanton and Liptay, 2000; Liptay et al., 1998). It should be noted that this may be regarded as a conservative estimate (de Visscher et al., 2004). Anaerobic zone gas was collected from recovery headers and wells, while gas reflecting CH_4 oxidation was collected either in static flux chambers or through upwind/downwind air sampling along selected transects. These static flux chambers also provided additional point measurements of CH_4 emissions.

3.5. Change in CH_4 storage

Landfills can temporarily sequester or release CH_4. Change in CH_4 storage is perhaps the most difficult term of the balance to quantify. The term is a function of several variables including the creation of additional void space by the decomposition of the waste, settlement that occurs due to the increased void space, fluctuating leachate levels within the landfill, changes in extraction efficiencies, changes in the amount of dissolved CH_4 in leachate, as well as temperature and barometric pressure changes. All of these factors lead to changes in the amount of CH_4 gas contained within the air-filled porosity of the landfill. The CH_4 quantity within the landfill mass does change temporarily (Fig. 2). This is a significant factor in the dynamic CH_4 balance at a site and is often neglected in assessment studies (e.g., Peer et al., 1993). A reduction of 1% (v/v) in CH_4 concentration (assuming in this case the volume and pressure of the landfill remains constant) would require $14\ g\ CH_4\ m^{-3}$ moving out of the gas-filled porosity, or 6 kg CH_4 for a $20 \times 20 \times 10$ m cell (assuming a gas-filled porosity of 0.20). The value of 0.2 for the air-filled porosity is the average of landfill gas-filled porosities observed in other studies (e.g., Hutchigs et al., 2001; Cestaro et al., 2003). Change in storage is estimated from the temporal changes in CH_4 concentrations from gas sampling wells or, if they are not available, from the changing CH_4 concentrations in

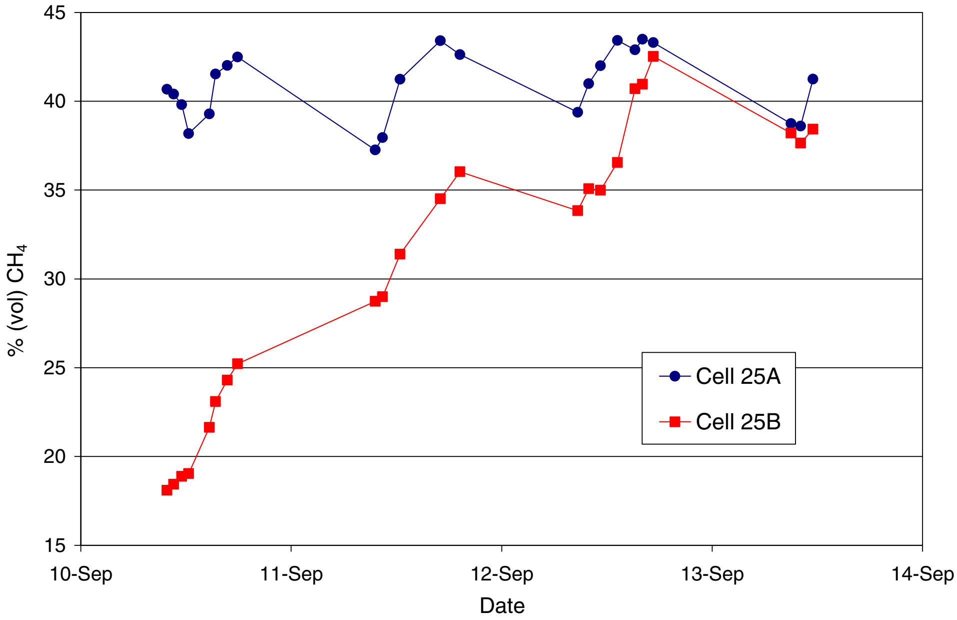

Fig. 2. Temporal variability of CH_4 concentration within the gas-filled pore space of the waste from Grand'Landes site (September 2002). Cell 25A is the conventional landfill gas recovery system with vertical wells, and Cell 25B has an innovative horizontal system as described in this paper.

the main header from the gas recovery system. The change in storage (ΔS_{CH_4}) is calculated from the CH_4 concentration changes within the waste (ΔA; kg m^{-3}), gas-filled porosity (ϕ_{air}; 0.2), and the total landfill volume (V; m^3):

$$\Delta S_{CH_4} = (\Delta A) V \varphi_{air}. \tag{4}$$

The accuracy of this term is highly dependent on the temporal sampling density of the landfill CH_4 gas concentrations because higher sampling densities will lead to improved estimations of the change in storage term.

3.6. CH_4 migration

A variety of engineered controls, including geomembranes (HDPE), compacted clay layers, and GCL liners, can be used to prevent subsurface lateral migration of leachate and landfill gas at the cell perimeter. Once a system reaches steady state, flow through a combination of barriers is controlled by the slowest diffusion rate. For the system investigated here, the HDPE geomembrane is the rate-limiting barrier, assuming no compromise in the integrity of the membrane. Therefore, only the diffusive flux through the geomembrane was used in this study. Transmission of gas molecules through a polymer structure is through permeation. For HDPE, the polymer liner will only allow 0.58 cm^3 CH_4 m^{-2} d^{-1} at 1 atm pressure and 1.5 mm membrane thickness to pass through (Pauly, 1989; Lim, 1995). This translates into 400 μg CH_4 m^{-2} d^{-1} per 1.5 mm of thickness. Cell shape was assumed to be cubic; then, the area of the four sides and bottom were summed and multiplied by the permeation rate above to estimate CH_4 migration. CH_4 losses through the top of the cell were accounted for through the emission measurements discussed previously.

4. Results

4.1. Quantification of CH_4 emissions

In general, there was good agreement ($r^2 = 0.81$) between the results of the tracer and chamber measurements extrapolated with IDW methodology. However, greater discrepancies existed when the measurement area was less than 1 ha. One of the potential problems with aboveground micrometeorological and tracer measurements is quantifying the footprint from which the measured flux is originating. This footprint is largely determined by the meteorological conditions and terrain considerations (Carpentieri et al., 2004). An additional problem with the tracer technique was illustrated during the winter (December 2001 campaign) at the Lapouyade site, where a gas collection pipe was broken during landscaping activities, resulting in high emissions from this point source. This artificially biased the tracer measurements during this time period. We recommend caution when directly comparing chamber and tracer measurements to ensure that the basis of comparison (e.g., flux footprint areas) is valid. IDW spatial interpolation was used to process the chamber measurements into integrated area fluxes for reported surface emission values for the test cells.

Each landfill cell will be evaluated individually. The measured and modeled parameters were summed for the sources or sinks in the CH_4 balance. Then the CH_4 balance was solved by setting the change in storage term to the respective quantity needed to achieve mass balance. A reasonable balance was assumed when the change in storage was below the maximum calculated using Eq. (4).

4.2. Montreuil-sur-Barse

Differences in cover design between these cells of equal size are apparent in the data (Table 3). There was a 6-fold reduction in total emissions and 1.8-times increase in recovery observed for the clay cover as opposed to the GCL cover. Percentage recovery also varied considerably between the two designs (clay: 94%; GCL: 41%). Thus, the 1 m clay cover was much more efficient in terms of containment as indicated by the lower emission and higher recovery. Most of the emissions (94%) occurred on the side slopes of both test cells. This could be due to preferential flow paths caused by erosion but was not investigated further. The total oxidation was higher above the GCL due to the higher emission rate. However, both cells were oxidizing approximately 4% of the total emission through the cover. This low value for CH_4 oxidation was due to high soil moisture at the site during the field investigation (Whalen et al., 1990). As shown in Table 3, both cells passed the CH_4 balance check because the mass of CH_4 to achieve mass balance was less than the maximum calculated using Eq. (4).

4.2.1. Lapouyade

There was an increase in CH_4 oxidation and emission at both test sites (final and temporary covers) comparing the summer and winter seasons. This seasonal increase in CH_4 oxidation has been observed in other studies (Czepiel et al., 1996; de Visscher et al., 1999; Chanton and Liptay, 2000; Börjesson et al., 2004). During the summer, a 5-fold increase in the oxidation rate was observed in the temporary cover cell and a 8.5-fold increase in the final cover cells. However, the air temperatures were similar at both measurement times. Thus, this increase hypothetically could be linked to improved aeration status in the summer.

There was also a decrease in the observed efficiency of the LFG recovery system at the site between the winter and summer monitoring periods. However, the recovery system in all cases still performed at a level exceeding 90% of the predicted generation rate.

Cell A10 was a temporary covered cell without an active gas recovery system. This cell did not pass the CH_4 balance criterion, mainly due to the biased CH_4 emission that was

Table 3
Summary of field CH_4 balance measurements[a]

Field location	Production	Recovery	Emission	Oxidation ($kg\ CH_4\ d^{-1}$)	Migration	Δ storage	Δ storage (maximum)	% Recovery
Montreuil-sur-Barse								
Final clay cover with LFG recovery	*109.0*	**−102.0**	**−8.1**	**−0.3**	*−1.1*	**+2.5**	**±32.0**	**94.0**
Final GCL cover with LFG recovery	*136.5*	**−55.8**	**−49.4**	**−2.1**	*−1.1*	**−28.1**	**±32.0**	**40.9**
Lapouyade								
Final clay cover with LFG recovery								
Summer	*4358.0*	**−3935.0**	**−298.6**	**−83.5**	*−20.0*	**−20.9**	**±202.0**	**98.1**
Winter	*3970.0*	**−3893.0**	**−56.0**	**−9.8**	*−20.0*	**+8.8**	**±253.0**	**90.3**
Thin temporary clay cover with LFG recovery								
Summer	*383.0*	**−346.0**	**−287.0**	**−6.5**	*−3.0*	**259.5**	**±383.0**	**92.5**
Winter	*317.0*	**−293.2**	**−15.0**	**−2.3**	*−4.0*	**−2.5**	**±383.0**	**88.0**
Thin temporary clay cover without LFG recovery								
Summer	*1100.0*	**0**	**5369.0**[c]	**−7.1**	*3.0*	**−6479.1**	**±448.0**	**0**
Grand'Landes								
Final clay cover with vertical LFG recovery	*1250.0*	**−1101.0**	**−0.01**	**−4.0**[b]	*−5.1*	**−0.0**	**±317.0**[d]	**88.0**
Final geomembrane cover with horizontal LFG recovery	*953.0*	**−799.0**	**−6.2**	**−4.0**[b]	*−4.9*	**−138.9**	**±561.4**[d]	**83.8**

[a] Numerically modeled terms in italics and field measurements are in bold. A valid methane balance was achieved when the solved methane balance term was within the change in storage calculated from the field data (Δ storage maximum).

[b] The isotopic method was not applicable due to uptake of atmospheric CH_4 by soils above geomembrane. Oxidation was calculated from the maximum CH_4 that could diffuse through the geomembrane and be oxidized, since no surface emissions were observed.

[c] Only tracer measurements were available for emission quantification for this site, no chamber measurements were conducted.

[d] Differences in ΔCH_4 storage were due to differences in the fluctuations in CH_4 gas concentration between cells (Fig. 2).

measured by the micrometeorological tracer technique. There were no gridded dynamic chamber emission measurements taken at this cell (only static chamber measurements for quantifying methane oxidation using the isotopic technique). This cell was adjacent to the active filling site of the landfill, and therefore some of the CH_4 captured in the tracer study originated from the active area. This demonstrates the advantages of the CH_4 balance technique, since without this validation the tracer measurements would be assumed to be valid.

4.2.2. Grand'landes

The CH_4 emissions from the geomembrane-covered cell were negligible (Table 3). An additional transect using 6 static chambers across this cell, which had a horizontal collection system in the gravel layer below the geomembrane, indicated that the aerobic topsoil above the geomembrane was actively oxidizing CH_4 out of the atmosphere at low rates (-0.2 to $-2.2\ \mathrm{mg\ CH_4\ m^{-2}\ d^{-1}}$). The only observed source of emissions was on the side-slope area at the interface between the geomembrane cover and the side liner. This was likely due to imperfect seaming between the two liners.

The isotopic method to quantify CH_4 oxidation was not applicable to Grand'Landes because of the negligible emissions. In order for the isotopic method to be applicable, there needs to be measurable emissions from the cover. Therefore, the oxidation was calculated from the maximum CH_4 that could diffuse through the geomembrane with the assumption that this CH_4 was oxidised during transport.

4.2.3. Discussion

Total CH_4 recovery for the sites with active gas recovery ranged from 41% to 94% of the theoretical modeled CH_4 production. However, this recovery rate was highly dependent on the engineered cover design. The GCL did not perform well in reducing gaseous emissions with the lowest recovery percentage observed in this study (41%). If the GCL cover design is eliminated from the analysis, it should be noted that all of the other cover designs with active gas recovery systems exhibited recovery rates in excess of 90%. This is a very important finding of this research: the final soil cover and geomembrane composite covers all exceeded 85% recovery of the predicted CH_4 generation. This recovery rate limits the environmental impact of landfills by reduced gas emissions. Another important result is that the oxidation rates ranged from 4% to 50% of the total CH_4 flux through the various cover systems. By quantifying the degradation of the emitted CH_4 by methanotrophs in cover soils, it has been demonstrated that this is also an important process to reduce emissions to the atmosphere.

This study also illustrated the significance of accounting for the change in CH_4 storage within the landfill. The change-in-storage term is actually larger than the emission pathway at sites with engineered gas recovery systems. One of the largest difficulties with measuring the CH_4 balance at a site is the dynamic nature of temporal variations. Each of the CH_4 pathways fluctuates with daily (Fig. 2), weekly, and seasonal cycles; thus one must try to measure all the pathways simultaneously. However, with chamber measurements, these often require hours to days to capture the spatial variability that exists at a particular site. Therefore, the spatial distribution may change during the measurement. Using the CH_4 balance increases the reliability of landfill CH_4 assessment by constraining the result to be within the logical balance presented in Eq. (1), resulting in a higher confidence in the assessment. This balance is also useful in constraining the modelling results for CH_4 generation at a site because the sum of the measured pathways cannot exceed the total generation.

The results of this study were recently used by a working group established by ADEME to develop default values for landfill gas emissions from French landfills for the European Pollutant Emission Register (EPER), as part of the directive for integrated pollution prevention and control (IPPC). The chosen conservative values for CH_4 recovery were 35% for an operating cell with an active LFG recovery system, 65% for a temporary covered cell with an active LFG recovery system, 85% for a final clay covered cell with an active LFG recovery system, and 90% for a geomembrane final covered cell with an active LFG recovery cell. These were strictly assigned default values and can be altered if the landfill operator has actually measured alternative field values at individual sites.

5. Conclusions

Through intensive field measurements at 3 sites, the METHAN project quantified all of the pathways for CH_4 generated in 9 cells with a variety of cover designs and gas management strategies. Because CH_4 production is typically modeled from waste inputs and thus difficult to validate at field scale, the sum of measured CH_4 pathways (especially recovery, emissions, and oxidation) provides an improved methodology to evaluate the actual CH_4 generation and percent recovery at field scale. The fraction of CH_4 oxidized by methanotrophs in aerated cover soils ranged from <10% to 50%; in addition, the soil above a geomembrane at Grand'Landes was actively oxidizing CH_4 from the atmosphere. The fraction of CH_4 recovered ranged from 41% to 98% of the predicted generation. Lateral migration, modeled as a maximum, accounted for only 0.3–1.4% of the generation. Typically two techniques for emissions (tracer and chambers) were deployed in parallel since chambers quantify the spatial variability of emissions while the tracer technique captures integrated emissions from large surfaces. However, at specific sites, interpretation of tracer data can be problematical (e.g., edge of cell leakages or contributions from the active tipping area). Field measurements of CH_4 emissions ranged from 0% to 75% of the predicted generation, while the change-in-storage within the available field pore space accounted for 0.2–589% of the generation. This high percentage indicates that the change-in-storage can be almost 6 times more important to the balance as the gas generation, depending on site conditions. As illustrated through this study, the change-in storage within the gas-filled pore space is a significant contribution to the overall CH_4 balance at a site. The major contribution of this work was to document that, for landfills with active gas recovery systems, both engineered geomembrane cover systems and final soil covers are very effective (>84%) in recovering CH_4, thus significantly reducing the environmental impact of landfill emissions.

Acknowledgements

The METHAN project was supported by CReeD with the partial financial support of ADEME. The authors also like to acknowledge partial support for the preparation of this paper by the US National Science Foundation under Grant No. 0332070. We also gratefully acknowledge the assistance of all the ONYX site managers and field personnel for making this project possible.

References

Anthony, C., 1982. The Biochemistry of Methylotrophs. Academic Press, London.

Aran, C., Diot-Morcet, M., Hébé, I., Têtu, A., Watermolen, T., Mianowski, V., 2002. Economic feasibility and performances of two LFG collection systems: optimization of the amount of biogas recovered. In: 25th Annual Landfill Gas Symposium of SWANA. pp. 233–238.

Barker, J.F., Fritz, P., 1981. Carbon isotope fractionation during microbial methane oxidation. Nature 293, 289–291.

Barlaz, M.A., Green, R., Chanton, J.P., Goldsmith, C.D., Hater, G.R., 2004. Evaluation of a biologically active cover for mitigation of landfill gas emissions. Environmental Science and Technology 38, 4891–4899.

Bingemer, H.G., Crutzen, P.J., 1987. The production of methane from solid wastes. Journal of Geophysical Research 92 (D2), 2182–2187.

Blaha, D., Bartlett, K., Czepiel, P., Harris, R., Crill, P., 1999. Natural and anthropogenic methane sources in new England. Atmospheric Environment 33, 243–255.

Bogner, J.E., Matthews, E., 2003. Global methane emissions from landfills: new methodology and annual estimates 1980–1996. Global Biogeochemical Cycles 17, 34-1–34-18.

Bogner, J., Spokas, K., 1993. Landfill CH_4: rates, fates, and role in global carbon cycle. Chemosphere 26, 366–386.

Bogner, J., Spokas, K., Burton, E., Sweeney, R., Corona, V., 1995. Landfills as atmospheric methane sources and sinks. Chemosphere 31, 4119–4130.

Bogner, J., Meadows, M., Czepiel, P., 1997. Fluxes of methane between landfills and the atmosphere: natural and engineered controls. Soil Use and Management 13, 268–277.

Börjesson, G., Chanton, J.P., Svensson, B.H., 2001. Methane oxidation in two swedish landfill cover soils determined with the use of $^{13}C/^{12}C$ isotope ratios. Journal of Environmental Quality 30, 369–376.

Börjesson, G., Sundh, I., Svensson, B., 2004. Microbial oxidation of CH_4 at different temperatures in landfill cover soils. FEMS Microbiology Ecology 48, 305–312.

Carpentieri, M., Corti, A., Zipoli, L., 2004. Wind tunnel experiments of tracer dispersion downwind from a small-scale physical model of a landfill. Environmental Modelling and Software 19, 881–885.

Cestaro, S., Cossu, R., Lanzoni, S., Raga, R., 2003. Analysis of pressure field in a landfill during in situ aeration for waste stabilization. In: Proceedings Sardinia 2003, Ninth International Waste Management and Landfill Symposium S. Margherita di Pula, Cagliari, Italy, 6–10 October.

Chanton, J., Liptay, K., 2000. Seasonal variation in methane oxidation in a landfill cover soil as determined by an in situ stable isotope technique. Global Biogeochemical Cycles 14, 51–60.

Chanton, J.P., Rutkowski, C.M., Mosher, B.M, 1999. Quantifying methane oxidation from landfills using stable isotope analysis of downwind plumes. Environmental Science and Technology 33, 3755–3760.

Christopherson, M., Linderod, L., Jensen, P.E., Kjeldsen, P., 2000. Methane oxidation at low temperatures in soil exposed to landfill gas. Journal of Environmental Quality 29 (6), 1989–1997.

Christophersen, M., Kjeldsen, P., Holst, H., Chanton, J., 2001. Lateral gas transport in soil adjacent to an old landfill: factors governing emissions and methane oxidation. Waste Management and Research 19, 126–143.

Coleman, D.D., Risatte, J.B., Schoell, M, 1981. Fractionation of carbon and hydrogen isotopes by methane-oxidizing bacteria. Geochimca et Cosmochimca Acta 45, 1033–1037.

Coops, O., Luning, L., Oonk, H., Weenk, A., 1995. Validation of landfill gas formation models. In: Proceedings from Sardinia '95, Fifth International Landfill Symposium, CISA, S. Margherita di Paula, Calgiari, Italy, 2–6 October 1995.

Czepiel, P.M., Mosher, B., Crill, P.M., Harriss, R.C., 1996. Quantifying the effect of oxidation on landfill methane emissions. Journal of Geophysical Research 101, 16721–16729.

Czepiel, P.M., Shorter, J.H., Mosher, B., Allwine, E., McManus, J.B., Harriss, R.C., Kolb, C.E., Lamb, B.K., 2003. The influence of atmospheric pressure on landfill methane emissions. Waste Management 23, 593–598.

de Visscher, A., Thomas, D., Boeckx, P., van Cleemput, O., 1999. Methane oxidation in simulated landfill cover soil environments. Environmental Science and Technology 33, 1854–1859.

de Visscher, A., DePourcq, I., Chanton, J., 2004. Isotope fractionation effects by diffusion and methane oxidation in landfill cover soils. Journal of Geophysical Research 109, D18111, doi:10.1029/2004JD004857.

Galle, B., Samuelsson, J., Svensson, B.H., Borjesson, G., 2001. Measurements of methane emissions from landfills using a time correlation tracer method based on FTIR absorption spectroscopy. Environmental Science and Technology 35, 21–25.

Hansen, J., Sato, M., Lacis, A., Ruedy., R., Tegen, I., Mathews, E., 1998. Climate forcings in the industrial era. In: Proceedings of the National Academy of Sciences USA, 95, 12753–12758.

Houghton, J.T., Ding, Y., Griggs, D.J., Noguer, M., vandeLinden, P.J., Xiaosu, D., 2001. Climate Change 2001: The Scientific Basis, Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change (IPCC), Cambridge University Press, UK, 944 pages. Available from: http://www.ipcc.ch/pub/reports.html].

Hutchigs, T.R., Moffat, A.J., Kemp, R.A., 2001. Effects of rooting and tree growth of selected woodland species on cap integrity in a mineral capped landfill site. Waste Management and Research 19, 194–200.

Lim, K.T., 1995. Mega-molecular dynamics on highly parallel computers: Methods and applications. PhD Dissertation. California Institute of Technology, Pasadena, California.

Liptay, K., Chanton, J., Czepiel, P., Mosher, B., 1998. Use of stable isotopes to determine methane oxidation in landfill cover soils. Journal of Geophysical Research 103, 8243–8250.

Matthews, E., 2000. Wetlands. In: Khalil, M.A.K. (Ed.), Atmospheric Methane. Springer, Berlin, pp. 202–233.

Oonk, J., Scheepers, M., Takke, J.W., 1993. Overzicht Stortgasprojecten in Nederland 1983–1991. Adviescentrum Stortgas. Apledoorn, Netherlands.

Opsomer, S., Jannink, E., 1995. Caracterisation O.M. cet de Montreuil/Barse CGEA-ONYX, 5/30-31/1995. Final Report to CReeD, July 1995.

Pauly, S., 1989, third ed.. In: Brandrup, J., Immergut, E.H. (Eds.), Permeability and Diffusion Data, Polymer Handbook Wiley, NY.

Peer, R.L., Thorneloe, S.A., Epperson, D.L., 1993. A comparison of methods for estimating global methane emissions from landfills. Chemosphere 26, 387–400.

Richards, K., 1989. Landfill gas: working with gaia. Biodeterioration Abstracts 3, 317–331.

Scheepers, M.J.J., van Zanten, B., 1994. Handleiding Stortgaswinning. Adviescentrum Stortgas, Utrecht.

Schroeder, P.R., Dozier, T.S., Zappi, P.A., McEnroe, B.M., Sjostrom, J.W., Peyton, R., 1994. The Hydrologic Evaluation of Landfill Performance (HELP) Model: Engineering documentation for Version 3, EPA/600/R-94/168b, US. Environmental Protection Agency Risk Reduction Engineering Laboratory, Cincinnati, OH.

Spokas, K., Graff, C., Morcet, M., Aran, C., 2003. Implications of the spatial variability of landfill emission rates on geospatial analyses. Waste Management 23, 599–607.

Trégourès, A., Beneito, A., Berne, P., Gonze, M.A., Sabroux, J.C., Pokryszka, Z., Savanne, D., Tauziede, C., Cellier, P., Laville, P., Milward, R., Arnaud, A., Levy, F., Burkhalter, R., 1999. Comparison of seven methods for measuring methane flux at a municipal solid waste landfill site. Waste Management and Research 17, 453–458.

von Maubeuge, K.P., 2004. Geosynthetic clay liners as environmental protection in infrastructure projects. In: Floss, R., Bräu, G., Nußbaumer, M., Laackmann, K. (Eds.), Geotechnical Engineering with Geosynthetics, Proceedings of the Third European Geosynthetics Conference. Deutsche Gesellschaft für Geotechnik (DGGt) und Technische Universität München, Zentrum für Geotechnik (TUM-ZG), München, pp. 203–205.

Whalen, S.C., Reeburgh, W.S., Sandbeck, K., 1990. Rapid methane oxidation in a landfill cover soil. Applied Environmental Microbiology 56, 3405–3411.

Willumsen, H., 2003. Landfill gas plants worldwide: number and types. In: Proceedings Sardinia '03 Waste Management and Landfill Symposium. CISA, University of Cagliari, Sardinia, Italy, 2003.