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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, Pymont 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

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14 Abstract

15 Many developed countries have targeted landfill methane recovery among greenhouse gas mitigation strategies, since methane is the
16 second most important greenhouse gas after carbon dioxide. Major questions remain with respect to actual methane production rates in the
17 field settings and the relative mass of methane that is recovered, emitted, oxidized by methanotrophic bacteria, laterally migrated, or
18 temporarily stored within the landfill volume. This paper presents the results of extensive field campaigns at three landfill sites to eluci-
19 date the total methane balance and provide field measurements to quantify these pathways. We assessed the overall methane mass bal-
20 ance in field cells with a variety of designs, cover materials, and gas management strategies. Sites included different cell configurations,
21 including temporary clay cover, final clay cover, geosynthetic clay liners, and geomembrane composite covers, and cells with and without
22 gas collection systems. Methane emission rates ranged from -2.2 to $>10,000$ mg CH₄ m⁻² d⁻¹. Total methane oxidation rates ranged
23 from 4% to 50% of the methane flux through the cover at sites with positive emissions. Oxidation of atmospheric methane was occurring
24 in vegetated soils above a geomembrane. The results of these studies were used as the basis for guidelines by the French environment
25 agency (ADEME) for default values for percent recovery: 35% for an operating cell with an active landfill gas (LFG) recovery system,
26 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
27 90% for a cell with a geomembrane final cover and active LFG recovery.

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30 1. Introduction

31 When solid waste is buried in a landfill, the biodegrad-
32 able fractions decompose via a complex series of micro-
33 bial and abiotic reactions. Methane (CH₄), one of the
34 terminal products, is formed by methanogenic microor-
35 ganisms under anoxic conditions, either through the direct

36 cleavage of acetate into CH₄ and carbon dioxide (CO₂) or
37 the reduction of CO₂ with hydrogen. Large engineered
38 landfill sites containing more than a 1 million m³ of refuse
39 with high percentages of biodegradable municipal solid
40 waste can readily generate several hundred m³ of landfill
41 gas h⁻¹. Landfill gas, as recovered, is a water saturated
42 biogas that consists of 50–60% (v/v) CH₄, 40–50% (v/v)
43 carbon dioxide (CO₂), and numerous trace components.
44 Because of its CH₄ content, landfill gas has a fuel value
45 of 18–22 MJ m⁻³.

46 Technology to recover and utilize landfill CH₄ using
47 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).

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 CH₄ 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 CH₄ 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 CH₄ decreases a source of atmospheric CH₄, the second most important greenhouse gas after CO₂ and responsible for approximately 40% of global warming over the past 150 years (Hansen et al., 1998). Since CH₄ has an atmospheric lifetime of about 10 yr and a global warming potential (GWP) 23 times higher than CO₂ (mole basis, 100 yr timeframe), reductions in individual CH₄ sources can decrease atmospheric concentrations within a decade (Houghton et al., 2001). Annually, about 500–600 terragrams (Tg) (1 Terragram = 10¹² g) CH₄ are emitted to the atmosphere, with approximately 70% from several anthropogenic sources including rice cultivation, domesticated ruminant animals, biomass burning, natural gas leakages, coalbed CH₄, and landfills, with the remainder from natural wetlands (Matthews, 2000). Global landfill CH₄ 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 CH₄ contributions from landfill sources (Willumsen, 2003; Bogner and Matthews, 2003; Bogner, unpublished). Many countries are targeting landfill CH₄ recovery as a mitigation strategy to achieve national greenhouse gas reduction goals because landfills are concentrated sources of atmospheric CH₄ that are amenable to engineered gas extraction. In many developed countries, it is estimated that landfill CH₄ is the largest anthropogenic source of atmospheric CH₄. In developing countries, the annual volume of landfill CH₄ 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 CH₄ recovery at hundreds of sites worldwide, there have been virtually no field studies to better constrain the CH₄ 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). CH₄ generated in

landfills is partitioned into CH₄ recovered, emitted to the atmosphere, oxidized by methanotrophs, laterally migrated, and internally stored in the landfill volume (Bogner and Spokas, 1993), as follows:

$$\begin{aligned} \text{CH}_4 \text{ generated} = & \text{CH}_4 \text{ emitted} + \text{CH}_4 \text{ oxidized} \\ & + \text{CH}_4 \text{ recovered (flared)} \\ & + \text{CH}_4 \text{ migrated} + \Delta \text{CH}_4 \text{ storage} \end{aligned}$$

$$[\text{all units} = \text{mass time}^{-1}]. \quad (1)$$

Historically for commercial recovery projects, CH₄ 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 CH₄ 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 CH₄ 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

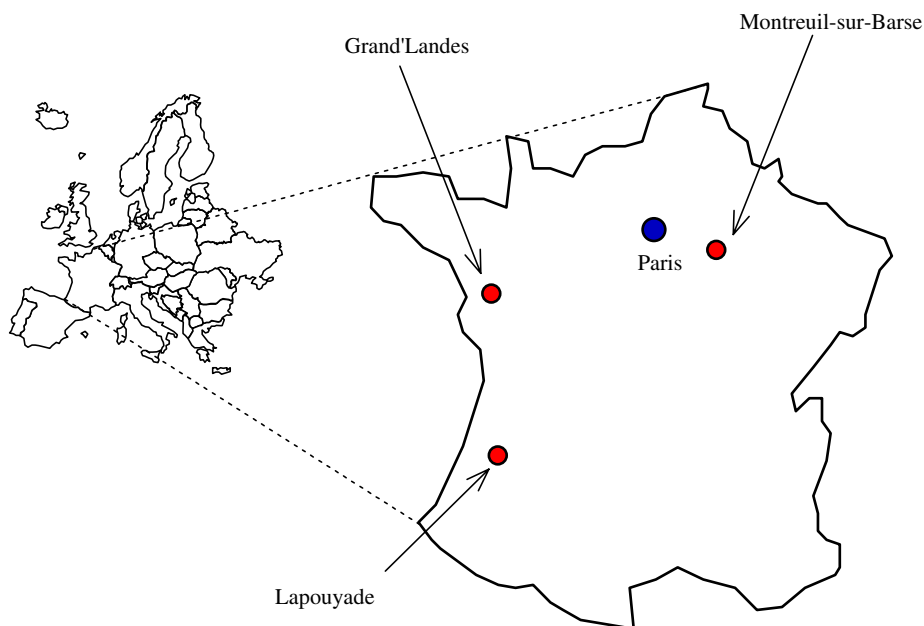


Fig. 1. Geographical location of investigated landfill sites in France.

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
<i>Montreuil-sur-Barse</i>						
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
<i>Lapouyade</i>						
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
<i>GrandLandes</i>						
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.

155 thickness was 130 cm). Field measurements were con-
156 ducted in February 2000.

157 2.2. Lapouyade landfill

158 Lapouyade is located near Bordeaux in southwestern
159 France (Fig. 1) and the landfill has been operating since
160 October 1996 receiving about 440 ton of waste d⁻¹. The
161 areas investigated at this site included a final soil cover
162 (with active gas recovery) and thin temporary soil cover

(with and without gas recovery) (Table 1). The cells with 163
gas recovery were investigated under different seasonal 164
conditions (December 2000 and September 2001). 165

2.3. Grandlandes landfill

The GrandLandes landfill is situated in western France 167
near Nantes (Fig. 1). This landfill has been operating since 168
1989 and was closed in 2001. CH₄ mass balance studies 169
were conducted on two experimental MSW cells with 170

171 different LFG recovery strategies. The first cell has conven- 211
 172 tional vertical wells with compacted clay as a top cover. 212
 173 The second cell has an innovative gas collection system 213
 174 consisting of horizontal pipes in a high permeability gas 214
 175 collection layer (gravel) at the top of the cell; the gravel 215
 176 layer is overlain by a high density polyethylene (HDPE) 216
 177 geomembrane (Aran et al., 2002). 217

178 3. Methods

179 For each field campaign, all of the measurements took 218
 180 place within 2 weeks under similar meteorological condi- 219
 181 tions. This was to avoid large fluctuations in barometric 220
 182 pressure caused by major storm events and other meteoro- 221
 183 logical conditions, which can greatly affect surface emis- 222
 184 sions (Galle et al., 2001; Czepiel et al., 2003). Below we 223
 185 describe the techniques used to quantify each CH₄ 224
 186 pathway. 225

187 3.1. CH₄ generation (landfill methane generation model: 188 LMGM)

189 The modeling of landfill gas production has primarily 231
 190 remained focused on empirical relationships derived from 232
 191 laboratory decomposition studies or test cell projects. 233
 192 The model developed for these sites is based on a first-order 234
 193 degradation model with multiple waste-types as inputs. 235
 194 This type of model has been successfully validated at land- 236
 195 fill sites with excellent agreement between measured and 237
 196 predicted gas generation rates (Coops et al., 1995). The 238
 197 amount of CH₄ generated was calculated from the follow- 239
 198 ing relationship (Oonk et al., 1993; Scheepers and van Zan-
 199 ten, 1994; Coops et al., 1995):

$$201 \quad V = G \sum_{n=1}^Z A_n C_{org,n} k_n e^{-k_n t}, \quad (2)$$

202 where V is the predicted amount of CH₄ produced (m³), G
 203 is the formation constant (0.94 m³ CH₄ kg⁻¹ organic car-
 204 bon), A_n is the total amount of waste stream n present in
 205 refuse (kg), $C_{org,n}$ is the amount of organic carbon present
 206 in waste stream n , k_n is the half-life of waste stream n
 207 (yr⁻¹), t is the elapsed years from start of filling, and Z is
 208 the number of waste categories. Default values were taken
 209 from the Dutch work on modeling landfill gas for Euro-
 210 pean landfills (Oonk et al., 1993; Coops et al., 1995). These

studies found that the multiphase production models and 211
 the simpler first order degradation models compared favor- 212
 ably at multiple sites. Where additional detailed waste in- 213
 put data are available, the prediction improves. Each cell 214
 investigated in this study had detailed waste data available 215
 for CH₄ generation modeling. This enabled an accurate 216
 model to be customized for each site. At the first site, Mon- 217
 treuil-sur-Barse, there was also a detailed characterization 218
 of MSW for each of the test cells (Opsomer and Jannink, 219
 1995; ADEME, 1992). It was assumed that this character- 220
 ization represented the average MSW composition for the 221
 other 2 landfills. Half-life decomposition times were di- 222
 vided into four categories: very slow (30 yr; textiles), slow 223
 (15 yr; paper), moderate (10 yr; fines), and fast (5 yr; labile 224
 organic material). The various waste categories and the or- 225
 ganic carbon and half-life assignments are summarized in 226
 Table 2. Fines is the category assigned to waste that was 227
 too small to physically identify and labile organic materials 228
 include food, vegetable, and green wastes. 229

3.2. CH₄ recovery 230

Where CH₄ is recovered by an active system of vertical 231
 wells or horizontal collectors, recovery is directly quantified 232
 by mass flow measurements. The mass of CH₄ recovered is 233
 the most accurately quantified pathway of the CH₄ balance. 234
 CH₄ recovered is directly calculated from measured flow 235
 rates and CH₄ concentration data, assuming that the CH₄ 236
 concentration is constant between measurements and after 237
 correction to standard temperature and pressure. 238

3.3. CH₄ emissions 239

Point measurements of landfill CH₄ emissions can vary 240
 over seven orders of magnitude (0.0004–4000 g m⁻² d⁻¹) 241
 (Bogner et al., 1997). CH₄ emissions from landfills can be 242
 controlled by active gas recovery systems and the natural 243
 methanotrophic attenuation potential of engineered soil 244
 covers (Christopherson et al., 2000; Bogner et al., 1997). 245
 Two different approaches were used in parallel to quantify 246
 surface emissions: an above-ground tracer method and flux 247
 chamber techniques. 248

3.3.1. Tracer method 249

Atmospheric tracer methods have been previously used 250
 to study whole landfill emissions (Trégourès et al., 1999; 251

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

252 Galle et al., 2001; Czepiel et al., 2003). The method in this
 253 study has been previously described by Trégourès et al.
 254 (1999). Briefly, this method relies on concurrent above-
 255 ground concentration measurements for CH₄ and an inert
 256 tracer gas (SF₆) released at a known rate upwind from the
 257 landfill. In this method the emissions from the landfill are
 258 obtained from measurements of methane and tracer con-
 259 centrations in transects across the plume downwind from
 260 the landfill. By knowing the concentration of both CH₄
 261 and SF₆, the flux rate of CH₄ from the landfill area can
 262 be determined using numerical dispersion modeling. This
 263 method averages fluxes across surface heterogeneities,
 264 but the exact footprint of the emission source may be dif-
 265 ficult to determine and is dependent on meteorological
 266 conditions. Therefore, the footprint may not always coin-
 267 cide exactly with the landfill cell under investigation.

268 3.3.2. Flux chambers

269 A static recirculating chamber was used to measure CH₄
 270 flux density from many small areas of the landfill surface
 271 (0.25 m²). This approach utilizes a pump with a flow rate
 272 of approximately 10 l min⁻¹ to circulate the enclosed
 273 chamber headspace to an outside loop. The rate of CH₄
 274 enrichment in the loop is measured periodically using a lab-
 275 oratory gas chromatograph that is transported in a utility
 276 vehicle around the site (Trégourès et al., 1999). Resulting
 277 surface flux density is calculated from the change in cham-
 278 ber CH₄ concentration with time ($\Delta C/\Delta t$), chamber vol-
 279 ume (V), and the area (A) by:

$$281 \text{ flux} = \frac{V}{A} \left(\frac{\Delta C}{\Delta t} \right). \quad (3)$$

282 CH₄ flux density is typically measured in units of
 283 g CH₄ m⁻² min⁻¹. Continuous measurement of CH₄
 284 allows rapid calculation of the CH₄ emission rate from
 285 the enclosed area. Measured emissions of CH₄ at cham-
 286 ber locations are the result of complex biological, chemi-
 287 cal, and physical processes occurring within landfill
 288 cover soils, and there can be difficulties in the extrapola-
 289 tion of chamber measurements for large landfill areas,
 290 which are detailed elsewhere (Spokas et al., 2003). Cham-
 291 ber fluxes were measured on a 20 × 20 m grid pattern and
 292 spatially interpolated using inverse distance weighing
 293 (IDW).

294 3.4. CH₄ oxidation

295 Microbial CH₄ oxidation is carried out by methano-
 296 trophic bacteria. Methanotrophs are a class of methyl-
 297 trophic bacteria that possess a specific enzyme (methane
 298 monooxygenase), which allows them to oxidize CH₄ to
 299 methanol (Anthony, 1982). Methanol is further degraded
 300 to CO₂. A major uncertainty in the CH₄ balance is deter-
 301 mining the attenuation of CH₄ emissions by methano-
 302 trophic bacteria in the aerobic outer portions of the cover
 303 soil. Previous field efforts have shown that a significant
 304 portion (10–100%) of the CH₄ present in the cover is oxi-

dized by indigenous methanotrophs (Whalen et al., 1990; 305
 Czepiel et al., 1996; Liptay et al., 1998; Chanton et al., 306
 1999; Chanton and Liptay, 2000; Börjesson et al., 2001; 307
 Christopherson et al., 2001; Barlaz et al., 2004). In addi- 308
 tion, field studies have shown that when methane supply 309
 from below is either blocked or consumed quantitatively, 310
 then these bacteria can oxidize atmospheric CH₄ as it dif- 311
 fuses into the soil from above, once the concentration of 312
 CH₄ is below atmospheric concentrations (Bogner et al., 313
 1995). It should be noted that atmospheric concentrations 314
 at landfill sites can be significantly greater than background 315
 atmospheric concentrations (≈ 1.8 ppmv) (Barlaz et al., 316
 2004). 317

In this study, methane oxidation was determined using a 318
 stable carbon isotope technique. Since there is a preferen- 319
 tial oxidation of ¹²C over ¹³C in CH₄ by the methanotrophs 320
 (Barker and Fritz, 1981; Coleman et al., 1981), it is possible 321
 to determine the oxidized fraction by quantifying the 322
 change in $\delta^{13}\text{C}$ of CH₄ between the anaerobic zone and 323
 the surface (Chanton and Liptay, 2000; Liptay et al., 324
 1998). It should be noted that this may be regarded as a 325
 conservative estimate (de Visscher et al., 2004). Anaerobic 326
 zone gas was collected from recovery headers and wells, 327
 while gas reflecting CH₄ oxidation was collected either in 328
 static flux chambers or through upwind/downwind air 329
 sampling along selected transects. These static flux cham- 330
 bers also provided additional point measurements of CH₄ 331
 emissions. 332

333 3.5. Change in CH₄ storage

Landfills can temporarily sequester or release CH₄. 334
 Change in CH₄ storage is perhaps the most difficult term 335
 of the balance to quantify. The term is a function of sever- 336
 al variables including the creation of additional void 337
 space by the decomposition of the waste, settlement that 338
 occurs due to the increased void space, fluctuating leach- 339
 ate levels within the landfill, changes in extraction efficien- 340
 cies, changes in the amount of dissolved CH₄ in leachate, 341
 as well as temperature and barometric pressure changes. 342
 All of these factors lead to changes in the amount of 343
 CH₄ gas contained within the air-filled porosity of the 344
 landfill. The CH₄ quantity within the landfill mass does 345
 change temporarily (Fig. 2). This is a significant factor 346
 in the dynamic CH₄ balance at a site and is often neg- 347
 lected in assessment studies (e.g., Peer et al., 1993). A 348
 reduction of 1% (v/v) in CH₄ concentration (assuming 349
 in this case the volume and pressure of the landfill re- 350
 mains constant) would require 14 g CH₄ m⁻³ moving out 351
 of the gas-filled porosity, or 6 kg CH₄ for a 20 × 20 × 352
 10 m cell (assuming a gas-filled porosity of 0.20). The val- 353
 ue of 0.2 for the air-filled porosity is the average of land- 354
 fill gas-filled porosities observed in other studies (e.g., 355
 Hutchigs et al., 2001; Cestaro et al., 2003). Change in 356
 storage is estimated from the temporal changes in CH₄ 357
 concentrations from gas sampling wells or, if they are 358
 not available, from the changing CH₄ concentrations in 359

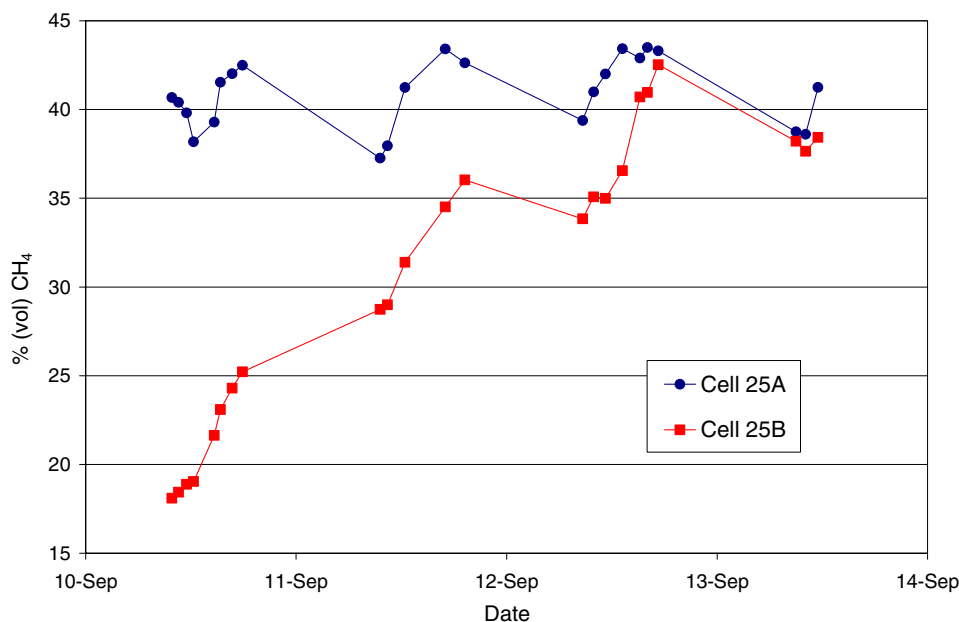


Fig. 2. Temporal variability of CH₄ concentration within the gas-filled pore space of the waste from Grand Landes site (September 2002). Cell 25A is the conventional landfill gas recovery system with vertical wells, and Cell 25B has an innovative horizontal system as described in this paper.

360 the main header from the gas recovery system. The
 361 change in storage (ΔS_{CH_4}) is calculated from the CH₄ con-
 362 centration changes within the waste (ΔA ; kg m⁻³), gas-
 363 filled porosity (ϕ_{air} ; 0.2), and the total landfill volume
 364 (V ; m³):

$$365 \Delta S_{\text{CH}_4} = (\Delta A)V\phi_{\text{air}}. \quad (4)$$

368 The accuracy of this term is highly dependent on the
 369 temporal sampling density of the landfill CH₄ gas concen-
 370 trations because higher sampling densities will lead to im-
 371 proved estimations of the change in storage term.

372 3.6. CH₄ migration

373 A variety of engineered controls, including geomem-
 374 branes (HDPE), compacted clay layers, and GCL liners,
 375 can be used to prevent subsurface lateral migration of
 376 leachate and landfill gas at the cell perimeter. Once a sys-
 377 tem reaches steady state, flow through a combination of
 378 barriers is controlled by the slowest diffusion rate. For
 379 the system investigated here, the HDPE geomembrane is
 380 the rate-limiting barrier, assuming no compromise in the
 381 integrity of the membrane. Therefore, only the diffusive
 382 flux through the geomembrane was used in this study.
 383 Transmission of gas molecules through a polymer structure
 384 is through permeation. For HDPE, the polymer liner will
 385 only allow 0.58 cm³ CH₄ m⁻² d⁻¹ at 1 atm pressure and
 386 1.5 mm membrane thickness to pass through (Pauly,
 387 1989; Lim, 1995). This translates into 400 μg CH₄ m⁻² d⁻¹
 388 per 1.5 mm of thickness. Cell shape was assumed to be cu-
 389 bic; then, the area of the four sides and bottom were
 390 summed and multiplied by the permeation rate above to
 391 estimate CH₄ migration. CH₄ losses through the top of

the cell were accounted for through the emission measure- 392
 ments discussed previously. 393

4. Results 394

4.1. Quantification of CH₄ emissions 395

In general, there was good agreement ($r^2 = 0.81$) be- 396
 tween the results of the tracer and chamber measurements 397
 extrapolated with IDW methodology. However, greater 398
 discrepancies existed when the measurement area was less 399
 than 1 ha. One of the potential problems with above- 400
 ground micrometeorological and tracer measurements is 401
 quantifying the footprint from which the measured flux is 402
 originating. This footprint is largely determined by the 403
 meteorological conditions and terrain considerations 404
 (Carpentieri et al., 2004). An additional problem with the 405
 tracer technique was illustrated during the winter (Decem- 406
 ber 2001 campaign) at the Lapouyade site, where a gas col- 407
 lection pipe was broken during landscaping activities, 408
 resulting in high emissions from this point source. This 409
 artificially biased the tracer measurements during this time 410
 period. We recommend caution when directly comparing 411
 chamber and tracer measurements to ensure that the basis 412
 of comparison (e.g., flux footprint areas) is valid. IDW spa- 413
 tial interpolation was used to process the chamber mea- 414
 surements into integrated area fluxes for reported surface 415
 emission values for the test cells. 416

Each landfill cell will be evaluated individually. The 417
 measured and modeled parameters were summed for the 418
 sources or sinks in the CH₄ balance. Then the CH₄ balance 419
 was solved by setting the change in storage term to the 420
 respective quantity needed to achieve mass balance. A 421

422 reasonable balance was assumed when the change in stor- 443
423 age was below the maximum calculated using Eq. (4). 444

424 4.2. Montreuil-sur-Barse

425 Differences in cover design between these cells of equal 446
426 size are apparent in the data (Table 3). There was a 6-fold 447
427 reduction in total emissions and 1.8-times increase in 448
428 recovery observed for the clay cover as opposed to the 449
429 GCL cover. Percentage recovery also varied considerably 450
430 between the two designs (clay: 94%; GCL: 41%). Thus, 451
431 the 1 m clay cover was much more efficient in terms of 452
432 containment as indicated by the lower emission and higher 453
433 recovery. Most of the emissions (94%) occurred on the 454
434 side slopes of both test cells. This could be due to prefer- 455
435 ential flow paths caused by erosion but was not investi- 456
436 gated further. The total oxidation was higher above the 457
437 GCL due to the higher emission rate. However, both cells 458
438 were oxidizing approximately 4% of the total emission 459
439 through the cover. This low value for CH₄ oxidation 460
440 was due to high soil moisture at the site during the field 461
441 investigation (Whalen et al., 1990). As shown in Table 462
442 3, both cells passed the CH₄ balance check because the 463
444 464

mass of CH₄ to achieve mass balance was less than the 443
maximum calculated using Eq. (4). 444

4.2.1. Lapouyade

445 There was an increase in CH₄ oxidation and emission at 446
447 both test sites (final and temporary covers) comparing the 448
449 summer and winter seasons. This seasonal increase in CH₄ 450
451 oxidation has been observed in other studies (Czepiel et al., 452
453 1996; de Visscher et al., 1999; Chanton and Liptay, 2000; 454
455 Börjesson et al., 2004). During the summer, a 5-fold in- 456
457 crease in the oxidation rate was observed in the temporary 458
459 cover cell and a 8.5-fold increase in the final cover cells. 460
461 However, the air temperatures were similar at both mea- 462
463 surement times. Thus, this increase hypothetically could 464
465 be linked to improved aeration status in the summer.

466 There was also a decrease in the observed efficiency of 467
468 the LFG recovery system at the site between the winter 469
470 and summer monitoring periods. However, the recovery 471
472 system in all cases still performed at a level exceeding 473
474 90% of the predicted generation rate. 475

476 Cell A10 was a temporary covered cell without an active 477
478 gas recovery system. This cell did not pass the CH₄ balance 479
480 criterion, mainly due to the biased CH₄ emission that was 481
482 483 484

Table 3
Summary of field CH₄ balance measurements^a

Field location	Production	Recovery	Emission	Oxidation (kg CH ₄ d ⁻¹)	Migration	Δ storage	Δ storage (maximum)	% Recovery
<i>Montreuil-sur-Barse</i>								
Final clay cover with LFG recovery	<i>109.0</i>	-102.0	-8.1	-0.3	<i>-1.1</i>	+2.5	±32.0	94.0
Final GCL cover with LFG recovery	<i>136.5</i>	-55.8	-49.4	-2.1	<i>-1.1</i>	-28.1	±32.0	40.9
<i>Lapouyade</i>								
Final clay cover with LFG recovery								
Summer	<i>4358.0</i>	-3935.0	-298.6	-83.5	<i>-20.0</i>	-20.9	±202.0	98.1
Winter	<i>3970.0</i>	-3893.0	-56.0	-9.8	<i>-20.0</i>	+8.8	±253.0	90.3
Thin temporary clay cover with LFG recovery								
Summer	<i>383.0</i>	-346.0	-287.0	-6.5	<i>-3.0</i>	259.5	±383.0	92.5
Winter	<i>317.0</i>	-293.2	-15.0	-2.3	<i>-4.0</i>	-2.5	±383.0	88.0
Thin temporary clay cover without LFG recovery								
Summer	<i>1100.0</i>	0	5369.0^c	-7.1	<i>3.0</i>	-6479.1	±448.0	0
<i>Grand Landes</i>								
Final clay cover with vertical LFG recovery	<i>1250.0</i>	-1101.0	-0.01	-4.0^b	<i>-5.1</i>	-0.0	±317.0^d	88.0
Final geomembrane cover with horizontal LFG recovery	<i>953.0</i>	-799.0	-6.2	-4.0^b	<i>-4.9</i>	-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 CH₄ by soils above geomembrane. Oxidation was calculated from the maximum CH₄ 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 ΔCH₄ storage were due to differences in the fluctuations in CH₄ gas concentration between cells (Fig. 2).

465 measured by the micrometeorological tracer technique.
466 There were no gridded dynamic chamber emission mea-
467 surements taken at this cell (only static chamber measure-
468 ments for quantifying methane oxidation using the
469 isotopic technique). This cell was adjacent to the active fill-
470 ing site of the landfill, and therefore some of the CH₄ cap-
471 tured in the tracer study originated from the active area.
472 This demonstrates the advantages of the CH₄ balance tech-
473 nique, since without this validation the tracer measure-
474 ments would be assumed to be valid.

475 4.2.2. Grand'landes

476 The CH₄ emissions from the geomembrane-covered cell
477 were negligible (Table 3). An additional transect using 6
478 static chambers across this cell, which had a horizontal col-
479 lection system in the gravel layer below the geomembrane,
480 indicated that the aerobic topsoil above the geomembrane
481 was actively oxidizing CH₄ out of the atmosphere at low
482 rates (-0.2 to -2.2 mg CH₄ m⁻² d⁻¹). The only observed
483 source of emissions was on the side-slope area at the inter-
484 face between the geomembrane cover and the side liner.
485 This was likely due to imperfect seaming between the two
486 liners.

487 The isotopic method to quantify CH₄ oxidation was not
488 applicable to Grand'Landes because of the negligible emis-
489 sions. In order for the isotopic method to be applicable,
490 there needs to be measurable emissions from the cover.
491 Therefore, the oxidation was calculated from the maximum
492 CH₄ that could diffuse through the geomembrane with the
493 assumption that this CH₄ was oxidised during transport.

494 4.2.3. Discussion

495 Total CH₄ recovery for the sites with active gas recovery
496 ranged from 41% to 94% of the theoretical modeled CH₄
497 production. However, this recovery rate was highly depen-
498 dent on the engineered cover design. The GCL did not per-
499 form well in reducing gaseous emissions with the lowest
500 recovery percentage observed in this study (41%). If the
501 GCL cover design is eliminated from the analysis, it should
502 be noted that all of the other cover designs with active gas
503 recovery systems exhibited recovery rates in excess of 90%.
504 This is a very important finding of this research: the final
505 soil cover and geomembrane composite covers all exceeded
506 85% recovery of the predicted CH₄ generation. This recov-
507 ery rate limits the environmental impact of landfills by re-
508 duced gas emissions. Another important result is that the
509 oxidation rates ranged from 4% to 50% of the total CH₄
510 flux through the various cover systems. By quantifying
511 the degradation of the emitted CH₄ by methanotrophs in
512 cover soils, it has been demonstrated that this is also an
513 important process to reduce emissions to the atmosphere.

514 This study also illustrated the significance of accounting
515 for the change in CH₄ storage within the landfill. The
516 change-in-storage term is actually larger than the emission
517 pathway at sites with engineered gas recovery systems. One
518 of the largest difficulties with measuring the CH₄ balance at
519 a site is the dynamic nature of temporal variations. Each of

the CH₄ pathways fluctuates with daily (Fig. 2), weekly, 520
and seasonal cycles; thus one must try to measure all the 521
pathways simultaneously. However, with chamber mea- 522
surements, these often require hours to days to capture 523
the spatial variability that exists at a particular site. There- 524
fore, the spatial distribution may change during the mea- 525
surement. Using the CH₄ balance increases the reliability 526
of landfill CH₄ assessment by constraining the result to 527
be within the logical balance presented in Eq. (1), resulting 528
in a higher confidence in the assessment. This balance is 529
also useful in constraining the modelling results for CH₄ 530
generation at a site because the sum of the measured path- 531
ways cannot exceed the total generation. 532

The results of this study were recently used by a working 533
group established by ADEME to develop default values for 534
landfill gas emissions from French landfills for the Euro- 535
pean Pollutant Emission Register (EPER), as part of the 536
directive for integrated pollution prevention and control 537
(IPPC). The chosen conservative values for CH₄ recovery 538
were 35% for an operating cell with an active LFG recov- 539
ery system, 65% for a temporary covered cell with an active 540
LFG recovery system, 85% for a final clay covered cell with 541
an active LFG recovery system, and 90% for a geomem- 542
brane final covered cell with an active LFG recovery cell. 543
These were strictly assigned default values and can be al- 544
tered if the landfill operator has actually measured alterna- 545
tive field values at individual sites. 546

5. Conclusions 547

Through intensive field measurements at 3 sites, the 548
METHAN project quantified all of the pathways for CH₄ 549
generated in 9 cells with a variety of cover designs and 550
gas management strategies. Because CH₄ production is 551
typically modeled from waste inputs and thus difficult to 552
validate at field scale, the sum of measured CH₄ pathways 553
(especially recovery, emissions, and oxidation) provides an 554
improved methodology to evaluate the actual CH₄ genera- 555
tion and percent recovery at field scale. The fraction of 556
CH₄ oxidized by methanotrophs in aerated cover soils rang- 557
ed from <10% to 50%; in addition, the soil above a geo- 558
membrane at Grand'Landes was actively oxidizing CH₄ 559
from the atmosphere. The fraction of CH₄ recovered rang- 560
ed from 41% to 98% of the predicted generation. Lateral 561
migration, modeled as a maximum, accounted for only 562
0.3–1.4% of the generation. Typically two techniques for 563
emissions (tracer and chambers) were deployed in parallel 564
since chambers quantify the spatial variability of emissions 565
while the tracer technique captures integrated emissions 566
from large surfaces. However, at specific sites, interpreta- 567
tion of tracer data can be problematical (e.g., edge of cell 568
leakages or contributions from the active tipping area). 569
Field measurements of CH₄ emissions ranged from 0% to 570
75% of the predicted generation, while the change-in-stor- 571
age within the available field pore space accounted for 572
0.2–589% of the generation. This high percentage indicates 573
that the change-in-storage can be almost 6 times more 574

575 important to the balance as the gas generation, depending
576 on site conditions. As illustrated through this study, the
577 change-in storage within the gas-filled pore space is a sig-
578 nificant contribution to the overall CH₄ balance at a site.
579 The major contribution of this work was to document that,
580 for landfills with active gas recovery systems, both engi-
581 neered geomembrane cover systems and final soil covers
582 are very effective (>84%) in recovering CH₄, thus signifi-
583 cantly reducing the environmental impact of landfill
584 emissions.

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