



LANDFILLS AS ATMOSPHERIC METHANE SOURCES AND SINKS

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ABSTRACT

Sanitary landfills are recognized as globally significant sources of atmospheric methane, but field measurements are rare. Existing country-specific landfill emissions have been estimated from solid waste statistics and a series of assumptions regarding methane generation and emission rates. There has been no attempt to reconcile the national and global estimates with limited field data on landfill methane emissions which range over six orders of magnitude (Bogner and Scott, 1995).

This paper addresses controlled field measurements of methane emissions at sites in Illinois and California (USA) using a closed chamber technique. Overall, observed rates from various controlled monitoring experiments during 1988-1994 ranged from 0.003 to more than 1000 g CH₄ m⁻² d⁻¹. Rates were related to the presence or absence of gas recovery wells, physical properties of cover soils (texture, moisture, and temperature) relating to their aeration status for diffusional flux, and rates of methane oxidation by indigenous methanotrophs. Surprisingly, at the Illinois site during spring, 1994, the landfill surface was consuming atmospheric methane rather than emitting landfill methane. This was attributed to high capacities for methane oxidation in well-aerated soils which had reduced landfill methane compared to 1993, the result of an effective pumped gas recovery system. Three independent methods confirmed that the landfill cover soils were functioning as a methane sink: (a) static closed chamber measurements yielding negative flux rates (uptake of atmospheric methane); (b) rates of methane oxidation similar to chamber results from *in vitro* field incubation studies using ambient methane; and (c) a reversal in the soil gas methane concentration gradient at the 25 cm depth. Field verification of landfill cover soils functioning as methane sinks has profound implications for revision of landfill contributions to global methane budgets; furthermore, it should be feasible to develop mitigation strategies incorporating a combination of engineered and natural methanotrophic controls.

1. BACKGROUND

Sanitary landfills are sites of controlled burial of refuse with surface areas ranging from tens to hundreds of hectares. An evolution in landfill design among developed countries over the last 30 years has resulted in highly-engineered modern facilities with systematic containment of solids, liquids, and gases. Filling is typically accomplished by a sectional approach where, at the completion of a given portion of a site, a final compacted soil cover 1-2 m thick is placed on the surface, seeded, and revegetated. There is also daily

application of soil cover (few cm) or synthetic alternatives. Because landfills are often the least expensive, environmentally-acceptable waste disposal alternative, they have been a prevalent method of waste disposal worldwide. In the U.S., 62% of municipal solid waste was landfilled in 1993 (U.S. EPA, 1994). Increasingly, more controlled landfilling practices are evolving in developing countries.

After refuse burial, anaerobic conditions are quickly established with depletion of free oxygen and oxygenated species. Landfills function as small dispersed anoxic basins with high rates of microbially-mediated methane generation at temperatures and pressures slightly above ambient. This setting is similar to early diagenetic conditions for anaerobically buried organic matter in geologic settings (Bogner, 1992; Bogner and Spokas, 1993; Bogner and Spokas, 1995). At a landfill, however, the transition between aerobic conditions at the top and anaerobic conditions in the thick refuse sequence below (zone of methanogenesis) occurs over short vertical distances. Aerobic soils at the top may be characterized by high capacities for methanotrophic methane oxidation as a natural biomediation mechanism (Mancinelli et al., 1981; Mancinelli and McKay, 1985; Whalen et al., 1990; Knightley et al., 1995).

In many developed countries, since the biogas extracted from landfilled refuse contains 50-60 V% methane, pumped gas recovery systems have been installed to reduce emissions and recover methane for energy use. Commercial methane recovery via vertical wells or horizontal collectors now occurs at more than 100 sites in the U.S (Thorneloe and Pacey, 1994) and more than 400 sites worldwide (M. Meadows, Harwell, U.K., personal communication).

Landfill methane has been implicated in global warming scenarios linked to historic increases in atmospheric methane. Typically, however, a "top-down" approach has been applied in which landfill emissions have been estimated from country-specific solid waste statistics and a series of assumptions regarding methane generation and emission rates. Resulting compilations for worldwide landfill methane emissions have ranged from 9-70 Tg year⁻¹ (U.S. EPA, 1994; Hogan et al., 1991; Bingemer and Crutzen, 1987; Richards, 1989), with the most recent estimates suggesting that landfills contribute 22-36 Tg CH₄ year⁻¹ to an annual atmospheric burden of approximately 500 Tg CH₄ year⁻¹ (Subak et al., 1992; Khalil and Shearer, 1993; Doorn and Barlaz, 1995). It has been estimated that landfills are the largest anthropogenic source of methane in the U.S. (U.S. EPA, 1993).

There is a strong need to improve landfill emissions estimates through process-level understanding of major controlling variables. This is accomplished by field measurements at representative sites using techniques well-verified for methane emissions from other area sources such as wetlands. At sites with existing pumped gas recovery systems, there is rarely any field measurement of the residual methane emissions, precluding field-based determination of the methane recovery efficiency. In the U.S., regulations are being put into place under the Clean Air Act Amendments to estimate site-specific emissions from modeled methane generation (assuming first order kinetics) minus any measured methane recovery via pumping wells; this approach also requires field validation through parallel measurement of emissions at selected sites. Since the

worldwide number of pumped gas recovery systems is increasing, field quantification of their effect on emissions is thus relevant to global methane budgets.

A compilation of existing worldwide field studies (Bogner and Scott, 1995), mostly using closed chamber techniques, has indicated that landfill methane emissions may range over six orders of magnitude (0.003 to $3000 \text{ g m}^{-2} \text{ day}^{-1}$). To date, there has been no attempt to more rigorously examine process-level controls which account for the high variability in measured landfill methane emissions. The purpose of this paper is to relate landfill methane emissions to major controlling variables, including both natural controls (soil aeration status, rate of methanotrophic methane oxidation) and engineered controls (presence or absence of a pumped gas recovery system).

Herein we report results from two contrasting field sites (northern Illinois and southern California). Both sites are active landfills that receive conventional municipal solid waste; the experimental areas at both sites had compacted cover soil overlying the refuse. This paper compares methane emissions at the Mallard Lake Landfill, DuPage County, Illinois, and the Brea-Olinda Landfill, Orange County, California, during 1988-1994. In particular, 1994 emissions data at the humid Mallard Lake site are contrasted to: (1) 1992-1993 data from the same site; and (2) 1988 and 1994 data from the semi-arid Brea-Olinda Landfill Orange County, California). The pre-1994 data have been previously published (Bogner, 1992; Bogner and Spokas, 1993; Bogner et al., 1993). The data sets were derived from various controlled monitoring experiments where methane emissions relative to soil type, soil moisture content, and status of pumped gas recovery were the major variables. This paper addresses order-of-magnitude differences and the high variability of landfill methane emissions between various settings, with major emphasis on the effect of the pumped gas recovery system. We also present recent field data from the Illinois site indicating that landfill cover soils may seasonally function as sinks for atmospheric methane.

2. PREVIOUS WORK: OVERVIEW OF LANDFILL GAS FIELD MEASUREMENTS PRIOR TO 1994

During 1985-1987, we examined vertical soil gas concentration and pressure profiles at the Mallard North Landfill, DuPage County, Illinois (Bogner, 1986; Bogner and Moore, 1986; Bogner et al., 1987). No gas recovery wells had been installed at this site; rather, landfill gas was being passively vented using vertical perforated pipes, each with a simple flare at the top. Soil gas concentrations were typically 50 V% methane at the refuse-cover interface, decreasing to atmospheric concentrations (low ppm's) through approximately 1.5 m. of compacted clay cover. The highest methane concentrations at shallow depth in the cover materials were observed in probes located on side slopes, in some cases associated with mappable vertical fractures. During dry soil periods, continuous monitoring of soil gas pressures indicated equilibration with atmospheric pressure. However, during times of surface saturation, elevated soil gas pressures were observed in the cover materials and top of refuse (up to 2 KPa above atmospheric). During such times, shallow soil gas pressures were typically in-phase with barometric pressure but at a constant offset.

During 1987-1988, periodic soil gas studies, including field measurement of methane emissions using a static closed chamber technique, were conducted at the Brea-Olinda Landfill, Orange Co., California (Bogner, 1990; Bogner and Spokas, 1993). These studies were concentrated in a portion of the lower Alpha Canyon fill prior to installation of vertical gas recovery wells. Refuse had been placed to a depth of about 30 m during the mid-1980's. The cover consisted of 1 m sandy silt with sparse to negligible surface vegetation. Annual average precipitation at this site is approximately 36 cm. Methane emissions were extremely high, exceeding $1,000 \text{ g m}^{-2} \text{ day}^{-1}$. The high emissions were attributed to the lack of a pumped gas recovery system and the relatively coarse-grained dry cover materials. Average gravimetric moisture content of surface soils was <5% (wt water/wt dry soil).

During 1992-1993, we monitored methane emissions (using a static closed chamber technique) and soil gas concentrations from permanent probe clusters at the Mallard Lake Landfill, DuPage County, Illinois (Bogner et al., 1993). This site lies adjacent to the Mallard North site discussed above. Measurements were conducted on the south face of the south hill where the refuse depth is approximately 40 m. Mallard Lake is a mounded site, with the south hill rising a maximum of 60 m above grade. Cover materials consist of 1.5 m silty clay overlain by topsoil. Average annual precipitation is 89 cm. Gravimetric moisture contents of surface soils averaged 10.5% during the dry summer (1992) and 28.5% in the autumn-winter (1992-1993). This area was filled from the middle 1970's with final cover placement in the late 1980's. There is abundant grass cover and a well-integrated system of vertical gas recovery wells. Methane is currently flared; however, commercial electrical generation is planned since about $100,000 \text{ Nm}^3$ methane are recovered daily. In the 1992-1993 studies, emissions were monitored at 24 locations along a transect between two gas recovery wells to investigate proximal (near well) vs. distal (between well) differences (Bogner et al., 1993). Both the emissions measurements (using static closed chambers) and soil gas concentration data confirmed proximal/distal differences, with lower proximal values extending to a radius of about 20 m from each well. Methane emissions (measured by closed chamber techniques) were reduced by more than 10^3 at the proximal locations.

3. SITE DESCRIPTIONS AND METHODS FOR 1994 MONITORING

In 1994, we conducted additional monitoring at both the Mallard Lake and Brea-Olinda sites. At Brea-Olinda, vertical gas recovery wells had been installed in lower Alpha Canyon after our original monitoring in 1988. These wells contribute to an integrated vertical and horizontal gas recovery system drawing about $140,000 \text{ Nm}^3$ methane per day for on-site electrical generation. Probe clusters and chamber collars were placed in a proximal area between three gas recovery wells. This area also encompassed two contrasting surficial soil types in a nested grided design. One soil type was the same tan sandy silt cover material in the same area in which methane emissions had been measured in 1988. The second surficial soil type was a finer-grained gray clayey silt which formed a thin (few cm) veneer over the tan sandy silt.

At Mallard Lake, gas extraction had been optimized since 1993 by wellhead adjustments. We established semi-permanent monitoring locations in a proximal location immediately next to well EW-37 and in

a distal location approximately midway between well EW-37 and EW-40. These locations were chosen to maximize proximal-distal differences as determined from the 1992-1993 transect study.

At both sites, static closed chamber methods were supplemented by clusters of soil gas probes at the chamber locations to observe vertical concentration profiles. Chambers consisted of 9 L stainless steel hemispheres. For total measurement periods of 30 minutes or less, chambers were tightly sealed onto semi-permanent steel collars with water-filled upper trenches; the collars were embedded in the landfill surface. Samples (4-6) were withdrawn at 5-minute intervals. Fluxes were calculated from the product of the change in methane concentration over time (slope of linear regression for plot of concentration vs. time) and the chamber volume/chamber area ratio (Rolston, 1986). Permanent probes consisted of stainless steel or PVC (polyvinyl chloride) tubing at depths ranging from 10-100 cm. All probes had gas-tight top fittings. Continuous monitoring of soil gas pressure and temperature at various depths was accomplished by dedicated data acquisition systems coupled to additional permanent probes within the probe clusters. Thermister probes with sensitivity exceeding 0.1 °C. and differential electronic pressure transducers with sensitivity exceeding 4 Pa were used, respectively, for *in situ* temperature and pressure measurements (Spokas and Bogner, 1995). Soil moisture was determined both gravimetrically and *in situ* (Troxler moisture-density gauge with ²⁴¹Am and ¹³⁷Cs sources).

For the Mallard Lake site, landfill gases were analyzed at Argonne National Laboratory using a system of three HP 5890 (Hewlett-Packard) gas chromatographs (GC's) configured together in a 5-column, 5-detector arrangement (dual TCD/dual FID/ECD); this permitted routine analysis for gases of interest (CH₄, CO₂, O₂, N₂, N₂O) over multiple ranges with a single injection. Methane was analyzed by GC/FID. At Brea-Olinda (1994), methane was analyzed by GC/FID on an HP5890 and redundantly by TCD on an MTI P200 (Bogner et al., 1994); these systems were located in the Geochemistry Laboratory of the Unocal Hartley Research Center, Brea.

In 1994 at the Mallard Lake site, the methane oxidation potential of cover soils was investigated using a field incubation technique (method adapted from Ryden et al., 1987). Triplicate soil cores with a volume of approximately 200 mL were incubated *in vitro* for two hours in depressions in the landfill cover (24-25 °C) to examine headspace methane loss. Trials included ambient atmospheric methane at 1.6 ppmv and 454 ppmv CO₂, plus trials with added gas standards to yield headspace concentrations of (a) 135 ppmv CH₄ [391 ppmv CO₂]; (b) 135 ppmv CH₄ [520 ppmv CO₂]; and (c) 3.5 V% CH₄ [3.3 V% CO₂].

4. RESULTS

Methane emissions from 1994 and historic monitoring at Mallard Lake and Brea-Olinda are summarized in Table 1. For the Brea-Olinda site, under semi-arid conditions with negligible to sparse surface vegetation, a decrease of more than two orders of magnitude through the tan sandy silty cover materials was observed

Table 1. Summary of Measured Methane Emissions at Landfills in Illinois (Mallard Lake) and California (Brea-Olinda). All data using static closed chambers. Arithmetic means in $\text{g m}^{-2} \text{day}^{-1}$ (standard deviation in parentheses following value). N is number of values averaged, weighted according to number of daily replicates for each value.

<u>Site</u>	<u>Year</u>	<u>Recovery Wells</u>	<u>Experimental Area</u>	<u>Value</u>	<u>N</u>
Mallard Lake	1992-1993	Yes	Proximal-silty clay cover	0.0032 (0.0024)	12
			Distal-silty clay cover	19.7 (42.2)	29
	1994	Yes (optimized)	Proximal-silty clay cover	-0.00098 (0.0008)	5
			Distal-silty clay cover	-0.00154 (0.0018)	6
Brea-Olinda	1988	No	Sandy silt cover	1119 (804)	4
	1994	Yes	Proximal-Sandy silt cover	4.03 (3.4)	18
			Proximal-Clayey silt cover	0.0032 (0.0016)	9

following the installation of a gas recovery system between 1988 and 1994. Soil gas methane concentrations in the refuse at depths of 5 m or more, sampled periodically during 1994, were consistently higher than 45 V%, suggesting that there was no loss of methane generation capacity since 1988. However, a major difference from 1988 was that the depth of high methane concentrations in soil gas (45-50 V%) had been depressed to 5 meters. In 1988, soil gas methane concentrations of 50 V% occurred at the cover-refuse interface (1 m depth).

The second set of Brea-Olinda data for 1994 indicated variations in methane emissions relative to contrasting soil types: an area with a veneer of finer-grained clayey silt overlying sandy silt had substantially reduced emissions (about 10^3) over an adjacent area with sandy silt alone. Contrasting gravimetric moisture contents for the two areas averaged 7.4% for the tan sandy silt and 12.2% for the gray clayey silt. Continuous pressure monitoring during 1994 data acquisition indicated negligible differential pressures (<0.1 kPa), suggesting that there was no direct observable convective influence of the gas recovery system within the cover soils; rather, the net effect of the gas recovery system was to reduce methane concentrations at the base of the cover, resulting in reduced rates of diffusive methane flux toward the atmosphere due to smaller gradients.

The Brea-Olinda emissions can be contrasted with the Mallard Lake results. The Mallard Lake data document emissions that may be typical for a humid Midwestern site with compacted clay cover materials and abundant vegetation. The 1992-1993 data for Mallard Lake, when methane emissions were monitored at 24 locations along a transect between two gas recovery wells, indicated emissions reduced by more than three orders of magnitude at the proximal locations over the distal locations (Bogner et al., 1993). These results indicated the strong influence of the gas recovery system. As at Brea-Olinda, concurrent monitoring of soil gas pressures in cover materials indicated that the direct convective influence of wells on soil gas transport in cover soils and methane emissions was negligible. Diffusion remained the major mechanism for bi-directional vertical transport of landfill gases to the atmosphere and atmospheric gases into the landfill. Again, the reduced methane concentrations at the base of the cover resulted in reduced rates of diffusive methane flux toward the atmosphere due to smaller gradients. It is important to note that net emissions with wells installed at Mallard

Lake (1992-1993 averages) were lower than the Brea-Olinda average for the sandy silt cover area after well installation (1994). This was attributed to thicker finer-grained cover soils at Mallard Lake with lower gas-filled porosity due to higher moisture contents. However, 1994 emissions for the finer-grained clayey silt cover area at Brea-Olinda were approximately the same as the proximal Mallard values (1992-1993). Average moisture contents for the silty clay cover at Mallard Lake (1992-1993) and the clayey silt cover at Brea-Olinda (1994) overlapped, approximately 12 % at Brea-Olinda and a range of 11-24% at Mallard Lake. Methane oxidation and proximal/distal soil gas concentration data will be addressed in more detail below.

At Mallard Lake in 1994, guided by the 1992-1993 monitoring, semipermanent distal and proximal sites were established to observe temporal variations. Figure 1 (a) indicates resulting fluxes for methane at proximal and distal locations in the spring and early summer, 1994. For this period, a negative slope was obtained for plots of chamber methane concentrations vs. time, indicating "negative" fluxes. These fluxes are a measure of the rate of atmospheric methane oxidation by soil methanotrophs under the chambers (Whalen and Reeburgh, 1990). Rates ranged from -0.0003 to $-0.005 \text{ g m}^{-2} \text{ day}^{-1}$. Methane-oxidizing rather than methane-emitting conditions during spring, 1994, can be contrasted with the 1992-1993 data in Table 1 when methane emissions varied between 0.003 and $21 \text{ g m}^{-2} \text{ day}^{-1}$. Sporadic negative fluxes had also been observed at proximal locations during 1992-1993; these averaged $-0.0075 \text{ g m}^{-2} \text{ day}^{-1}$, in absolute value similar to the lowest positive proximal fluxes (Bogner et al., 1993). One Swedish study has also reported negative methane fluxes averaging $-0.0027 \text{ g m}^{-2} \text{ day}^{-1}$ at a landfill with specialized cover materials consisting of clay mixed with sewage sludge (Borgesson and Svensson, 1993). Given temporal cross-overs at landfills from net methane-emitting to net methane-oxidizing conditions, it is also possible that a dynamic equilibrium where rate of methane supply is equal to rate of methanotrophic methane oxidation may persist for some period of time.

Figures 1b and 1c show temporal changes in shallow subsurface soil moisture and soil temperature at Mallard Lake during spring, 1994. The temperature of surface soils increased during spring warming while soil moisture concurrently decreased. Differences between the predominately positive fluxes in 1992-1993 and the low negative fluxes in 1994 can be explained by a combination of natural and anthropogenic controls, namely: (1) dry surface soils in 1994, facilitating diffusion of atmospheric oxygen into the soil profile; (2) a more efficient gas recovery system (additional wellfield adjustments), which lowered soil gas methane concentrations at the base of the cover to decrease diffusive flux of methane toward the atmosphere; and to be addressed below, (3) high capacities for methane oxidation in cover soils which had significantly reduced methane concentrations in 1994 compared to 1992-1993.

Figure 2 compares composite distal and proximal methane profiles (from shallow probes) for 1994 to profiles for 1992-1993. These profiles plot the geometric means of probe values composited by depth. Previous work (Bogner et al., 1993) has shown that this is a useful representation of the central tendency for probe values. The shapes of the 1992-1993 profiles were fairly typical for landfills characterized by net methane emissions in previous studies (Bogner, 1992; Bogner and Spokas, 1993). As discussed above, the proximal/distal contrast resulted in reduced rates of diffusive methane flux toward the atmosphere at the proximal sites due to smaller gradients. The 1994 profiles were distinctively different and indicated: (1) at

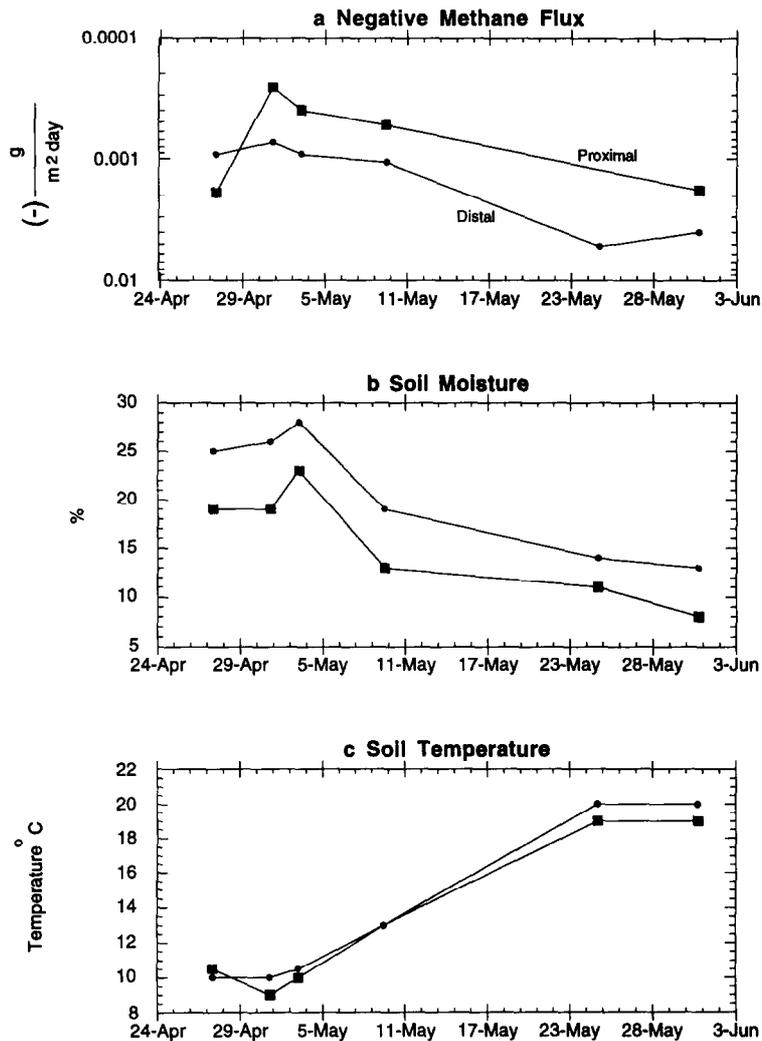


Figure 1. Temporal variations in methane fluxes at Mallard Lake during spring and early summer, 1994. Average flux of replicate static closed chambers; spline curves fitted to data. (Legend : - ■ - Proximal Location, - ● - Distal Location)
a) Negative methane fluxes. All values had $r^2 > 0.8$ for linear regression on dc/dt .
b) Gravimetric moisture content of surface soil (0-7 cm). (wt/dry wt).
c) Soil temperature at 7 cm.

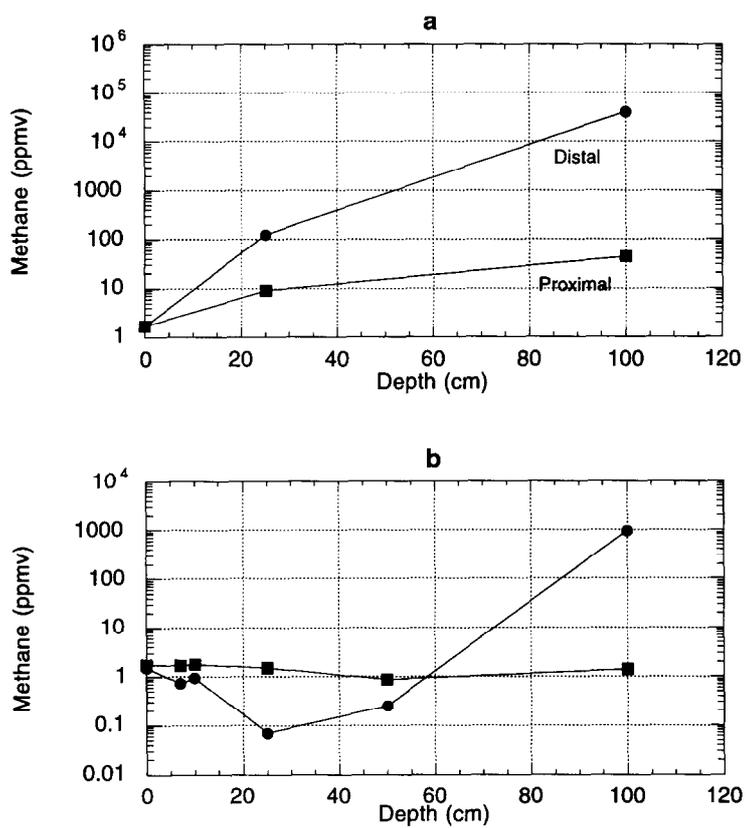


Figure 2. Soil gas methane profiles at Mallard Lake. Geometric means of probe values composited by depth. (Legend: - ■ - Proximal Location, - ● - Distal location)
a) 1992-1993.

b) 1994 (same period as Figure 1). Recovered gas at nearest wells had 43-49% methane with 0.2% oxygen.

proximal locations, atmospheric concentrations of methane (approximately 1.5 ppmv) to a depth of 100 cm; and (2) at distal locations, decreasing methane concentrations with depth to values below 0.1 ppm at 25 cm, where a reversal in the concentration gradient occurred. This reversal might be inferred to occur near the optimum depth of methane oxidation. At 100 cm depth, the mean methane concentration approached 1000 ppmv for the distal probes while it was below 2 ppmv for the proximal locations. Contrasting the proximal and distal chamber data for 1994 (Table 1), one must conclude that the distal rate of methane oxidation was higher than the proximal, suggesting a limitation of methane supply at proximal locations. Correspondingly, oxygen appeared not to be limiting since oxygen concentration profiles (not shown) indicated >19 V% at all locations and depths except at the distal 100 cm probes (approximately 17 V%).

The supporting *in vitro* field incubation studies demonstrated the potential of Mallard Lake soils to oxidize methane at rates several orders of magnitude above observed rates. When headspace methane at Mallard Lake was at ambient levels of 1.6 ppmv, the methane oxidation rate averaged $0.0004 \text{ g m}^{-2} \text{ day}^{-1}$. This rate lies within the range of both the negative chamber fluxes reported in this study and rates for non-landfill soil environments (approximately $0.0002\text{-}0.0065 \text{ g m}^{-2} \text{ day}^{-1}$) (Whalen and Reeburgh, 1990; Adamsen and King, 1993). The maximum measured rate of methane oxidation for the field incubations was $8.1 \text{ g m}^{-2} \text{ day}^{-1}$ when headspace methane was 3.5 V%. Previous laboratory studies of the capacity of landfill soils to oxidize methane have reported very high rates of $45\text{-}166 \text{ g m}^{-2} \text{ day}^{-1}$ (Whalen et al., 1990; Knightley et al., 1995).

5. CONCLUSIONS

Controlled field monitoring experiments indicate that it is possible to constrain the 10^6 range of observed landfill methane emissions on the basis of: (1) the presence or absence of pumped gas recovery wells; (2) the relative proximity of pumping wells to the point of monitoring; (3) physical properties of cover soils (texture, moisture, and temperature) relating to their aeration status; and (4) rates of methane oxidation by indigenous methanotrophs. In a current study at a northern Illinois (USA) landfill, there were no net methane emissions during the spring and early summer, 1994. The possibility of a landfill as a sink rather than a source for atmospheric methane has not been previously considered and was in direct contrast to 1992-1993 data for the same site which indicated methane emission rates up to $20 \text{ g m}^{-2} \text{ day}^{-1}$ (Bogner et al., 1993). The change from net methane-emitting to net methane-oxidizing conditions resulted from a combination of engineering and natural control mechanisms: more efficient gas recovery which lowered soil gas methane in the cover materials; high capacities for methanotrophic methane oxidation in cover soils; and high concentrations of soil gas oxygen from atmospheric diffusion due to seasonally dry soils with higher gas-filled porosity.

The data discussed herein indicate that future estimates for global landfill methane emissions should incorporate algorithms which recognize both: 1) the natural variability of measured emissions relative to major controlling variables; and 2) the possibility of landfill cover soils functioning as methane sinks. For the present, landfills require further application of field techniques to improve understanding of seasonal emissions at specific sites and better constrain national estimates for policy decisions.

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