THE SHORT TERM INFLUENCE OF ATMOSPHERIC PRESSURE

AND LONG TERM INFLUENCE OF GAS RECOVERY ON

LANDFILL METHANE EMISSIONS

by

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ABSTRACT

Landfills are the largest sources of methane (CH_4) emissions to the atmosphere in the United States. However, few measurements of whole landfill CH_4 emissions have been reported. Here, we present the results of a multi-year study of whole landfill CH_4 emissions using atmospheric tracer methods at the Nashua, New Hampshire Municipal landfill in the northeastern United States. Emissions were measured in 1994, before the installation of a gas collection system, and again in 1996 and 1997 subsequent to the installation of a gas collection system. Both pre- and post-collection system emissions were negatively correlated with surface atmospheric pressure. Emissions decreased approximately 2-fold before gas collection over a pressure range of 1011 to 1028 mbar. The pressure effect was even more striking after the initiation of gas collection when emissions decreased more than 3-fold over a pressure range of 1007 to 1023 mbar. From these data we concluded that, at this site: 1) surface atmospheric pressure changes play a predominant role in determining the magnitude of emissions to the atmosphere, and 2) landfill gas extraction increased the rate of gas production by 40%.

INTRODUCTION

Methane (CH_4) is a radiatively active trace gas whose abundance in the atmosphere has more than doubled during the past several hundred years and continues to rise. (Cicerone and Oremland 1988; Dlugokencky et al. 1998; Shipham et al. 1998). This increase appears to be well correlated with increasing human population (Rasmussen and Khalil 1984). CH_4 contributes significantly to global warming as a result of its ability to trap heat, 21 times more effective than carbon dioxide over a 100-year period. The short lifetime of CH_4 in the atmosphere, 11 years compared to 120 years for carbon dioxide, coupled with its potency make it an ideal candidate for emissions reduction.

Reducing emissions would result in relatively rapid reductions of atmospheric CH_4 concentrations. A reduction in total emissions of approximately 10 percent would stabilize methane concentrations at current levels (IPCC 1995). Reductions of this magnitude can be accomplished by mitigating anthropogenic sources. Human activity contributes about 70 percent of the estimated 500 to 700 teragrams of CH_4 emitted each year to the atmosphere, and landfills are among the largest of the anthropogenic sources (IPCC 1995). Landfills are estimated to account for approximately 37 percent of annual anthropogenic CH_4 emissions in the United States and 3 to 19 percent of global anthropogenic emissions (EIA 1999; USEPA 1999; Stern and Kaufmann 1996). However, there remains significant uncertainty associated with U.S. and international estimates.

In December 1990, the United Nations formed the Intergovernmental Negotiating Committee (INC) for a Framework Convention on Climate Change to establish policies to stabilize and reduce the emissions of CH_4 and other greenhouse gases. This effort

resulted in the establishment of the Kyoto Protocol (KP) to the Framework Convention in December 1997 (Bodanzky 1994). The KP fixed a set of greenhouse gas emission reduction targets for each participating country. These limits vary among each of the countries and range from 90 to 110 percent of 1990 level emissions.

The KP offers a number of flexibility mechanisms to reduce emissions that can be broadly described as emissions trading programs. Under these programs, countries receive credits for the quantities of greenhouse gases eliminated within their borders. Credits can also be generated by a country through the funding of greenhouse gas emission reduction projects in other countries. These credits can then be bought and sold, permitting the funding countries to meet their reduction commitments.

The most logical initial approach to greenhouse gas reduction, and credit generation, is to mitigate the most concentrated sources with the highest global warming potential (GWP). Landfills present a unique opportunity because they are large, concentrated sources of high GWP gas, the associated recovery technology is relatively simple and mature, and the recovered product has economic value as a fuel gas. The concentration of waste in a relatively small number of large landfills further enhances their economic attractiveness. About 900 of the more than 6,000 landfills in the United States produce about 75 percent of all landfill CH_4, and 19 of the largest landfills account for about 20 percent of the total landfill CH_4 generated (USEPA 1993). These statistics indicate that a concentrated effort involving a relatively small number of sites could produce substantial reductions in landfill CH_4 emissions. Approximately 250 landfills in the U.S. are currently collecting their landfill gas for energy recovery (EIA 1999).

A critical aspect of any credit-trading program for participating landfills is the verifiability of the commodity. This requires that market participants monitor and track their emissions with a high degree of accuracy. Several methods have recently been demonstrated to measure whole landfill CH_4 emissions (Czepiel et al. 1996; Tregoures et al. 1999). The most successful, accurate, and least costly in terms of time and expense, is atmospheric tracer methods.

Here we report the results of whole landfill CH_4 emission measurements, using an atmospheric tracer method, conducted at an active landfill before and after the initiation of gas recovery. The purpose of these measurements was to observe the effect of landfill gas recovery on CH_4 emissions to the atmosphere and to determine the influence of environmental variables on emissions.

SITE DESCRIPTION

The Nashua municipal landfill is located in the northeastern United States in the state of New Hampshire. The MSW disposal area has been accepting waste since 1971 and is currently open. A total of 1.1×10^8 kg of waste was accepted for disposal in 1994. The waste disposed, at that time, consisted of 36% MSW, 36% commercial waste, 13% construction and demolition (C&D) waste, and 13% wastewater sludge. The disposal rate peaked at 1.3×10^8 kg of waste in 1988. There was a total of approximately 2.3×10^9 kg of waste-in-place in 1997 (Czepiel et al. 1996; Reid 2000). All waste values are by wet weight.

The site covers approximately 60 ha and consists of an unlined 25-ha MSW landfill and a separate closed and unlined 5.5-ha C&D waste landfill located directly

adjacent to the MSW landfill. The landfill surfaces are covered by 1-2 m of sandy-clay loam surface material with limited surface fissuring. No landfill gas control systems were in place at the time of the initial measurements in 1994, although exploratory wells were under construction.

The gas recovery system in place during the subsequent measurements in 1996 and 1997 consisted of 60 vertical and horizontal wells installed in a gridded pattern. The Landfill gas recovery rate was maintained within a narrow range during the measurement periods, from 25500 to 28300 l min^{-1}. The recovered gas was approximately 50% CH_4, 45% carbon dioxide, and 5% other trace gases. The resulting CH_4 recovery rate ranged from 12750 to 14150 l min^{-1}. The gas is used to generate electricity using conventional internal combustion generators or is flared when the generators are not operating due to maintenance or malfunction.

ATMOSPHERIC TRACER METHOD

An atmospheric tracer method was used to measure the total CH_4 emission rate from the landfill. Pure sulfur hexafluoride (SF_6) tracer gas was released from the emitting surface to simulate landfill gas emissions (Mosher et al. 1999; Czepiel et al. 1996; Howard et al. 1992). If the released tracer is well mixed in the source plume, then the CH_4 emission rate can be obtained directly by the ratio method as

(1) $$Q_m = Q_t * (C_m/C_t)$$

where Q_m is the CH_4 flux rate, Q_t is the SF_6 tracer release rate, C_m is the measured CH_4 mixing ratio above background, and C_t is the measured mixing ratio of the SF_6 tracer. This method is restricted to situations with no interfering sources, a sufficient signal to be

measured against the background, and a strong enough source to be measured far enough downwind to ensure adequate mixing with the tracer gas.

SF_6 tracer gas was released from three cylinders located along the upwind edge of the landfill on a line perpendicular to the direction of the prevailing wind. The cylinders were placed at locations 0.25, 0.5, and 0.75 times the landfill width. CH_4 and SF_6 were measured in the resulting plume 1.2-3.5 km from the downwind edge of the landfill.

Both the CH_4 and SF_6 plumes were located in real-time by repeated plume traverses. This method allowed field personnel to immediately identify the required condition of good CH_4 and SF_6 mixing. Having identified an adequately mixed plume, evacuated gas-sampling canisters were placed along a perpendicular line within the plume, and a canister within the mobile van was used to collect samples along the entire length of the plume. The canister samples, which provide an integrated result over the entire measurement period, were used to independently verify and supplement the "instantaneous" results of the plume traverses using the mobile instruments. Canister sampling and real-time analyses also determined atmospheric background mixing ratios.

The real-time infrared (RTI) CH_4 analyzer, developed at Aerodyne Research, Inc., is based on the absorption by CH_4 of infrared radiation (3.39 μm wavelength) produced by a HeNe laser (McManus et al. 1989; McManus et al. 1992). The instrument is sensitive to changes in ambient mixing ratio of about 0.3% or 5 parts per billion by volume (ppbv) CH_4 (RMS at 1-s averaging). Response time is 6 s because of gas flow time through the sampling cell. The instrument was calibrated hourly by comparison to standard gases (Scott Specialty Gases) calibrated with NIST certified standards.

A continuous SF_6 gas analyzer was also van-mounted for measurement of the

tracer concurrent with the measurement of CH_4 (Benner and Lamb 1985). This custom instrument, developed by the Laboratory for Atmospheric Research at the Washington State University (WSU), utilizes an electron capture detector (ECD) with a response time of 0.4 s and a detection limit of less than 10 parts per trillion by volume (pptv). The instrument was calibrated hourly using SF_6 in air standards (Scott-Marrin, Inc.) over the range 25 pptv to 10 ppbv.

An infrared CO_2 analyzer (Li-Cor, Inc., model 6262) was used to measure ambient CO_2 mixing ratios. The CO_2 instrument was used to identify anomalous CH_4 sources. Significant CH_4 mixing ratios accompanied by high ambient CO_2 are characteristic of combustion processes and were neglected in the subsequent analyses.

Whole air samples were collected in evacuated canisters using portable samplers. Ambient air was drawn into each sampler using a battery-powered pump fitted with a Teflon-aluminum head. The 6 L stainless steel electro-polished canisters were pressurized with zero air for shipment and evacuated immediately prior to use in the field. The canisters were pressurized to approximately 138 kPa at a steady rate by pumping against a stainless steel capillary flow restrictor during a 30-min. sampling period. Approximately four to six canister samples were collected during each tracer test.

Canister samples were analyzed after return to WSU. CH_4 was quantified using a HP5880 gas chromatograph (GC) with a flame ionization detector after constituent separation using columns packed with HayeSep Q or Poropak Q. Mixing ratios were determined by comparison to standard gases traceable to NIST standards. Canister samples were analyzed for SF_6 using a HP5880 GC with an ECD after constituent separation using columns packed with HayeSep Q or Poropak Q. Mixing ratios were

determined by comparison to standard gases of varying mixing ratios of SF_6 in air (Scott-Marrin, Inc.).

The tracer release system consisted of a gas cylinder pressure regulator and a length of stainless steel capillary restrictor tubing. The length of the tubing was sized to yield approximately 2 L min^{-1} flow rate. The release system was calibrated using a dry gas meter prior to field deployment. Periodic observation of the release system was made using a calibrated mass flow meter during the tracer release period.

The emission rate from each sampling canister and plume traverse was calculated using equation 1 and the results were pooled. Each emission rate presented is the average of the pooled values and the coefficient of variation (CV) is the variability among these data. The variability is an indication of the agreement of results among the canisters and traverses during each test.

RESULTS AND DISCUSSION

CH_4 emissions were measured during field experiments in October and November 1994 with no operational gas collection system in place (Czepiel et al. 1996). Emission measurements were also conducted in August 1996, February, March, and April 1997 subsequent to the installation of a gas collection system in 1995. The resulting whole landfill emission data and CV are presented along with the surface atmospheric pressure in Table 1. Emissions before gas collection ranged from 11080 to 21575 l CH_4 min^{-1}, while emissions after the installation of a gas collection system ranged from 7317 to 26542 l CH_4 min^{-1}. Atmospheric pressure was determined by averaging hourly data

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recorded at a nearby weather station. The length of individual tests typically ranged from 3 to 5 hours and the change in pressure during the tests was no greater than 2 mbars.

Figure 1 presents measured CH_4 emissions as a function of atmospheric pressure measured during each test. An inverse relationship was observed between emissions and atmospheric pressure both before and after the installation of gas collection. The data were modeled by linear regression. The correlation coefficients, or r^2, of the linear regressions of the data before gas recovery and after gas recovery were 0.96 and 0.95, respectively. The slopes of the linear regressions were significantly dissimilar indicating a difference in sensitivity of emissions to atmospheric pressure before and after gas recovery.

The effect of methane oxidation on the emissions-pressure relationship after the installation of gas recovery is presented in Figure 2. Stable isotopes of carbon in the CH_4 plumes were used to determine whole landfill oxidation rates over a full range of seasonal cover soil temperatures at a depth of 10 cm (Chanton et al. 1999; Liptay et al. 1998). The calculated oxidation rates ranged from 5% at the low temperature of 4°C in March 1997 to 13% at the high temperature of 22°C in August 1996. Figure 2 shows the measured CH_4 emission values and the data adjusted to reflect the expected emissions before the effect of oxidation. While oxidation does change the magnitude of emissions and is used later in our estimate of annual emissions, it does not alter the sensitivity of emissions to atmospheric pressure based on the statistical similarity of the slopes of the regression lines.

The spatial heterogeneity of emissions may explain the difference in emissions sensitivity. Before the installation of a gas recovery system, internal gas and atmospheric

pressures were in a balance maintained by the heterogeneous resistive characteristics of the landfill body and surface. The extreme variability of surface emissions was reported by Czepiel et al. (1996) and is shown in Figure 2. Introducing a negative pressure in regularly spaced recovery wells significantly alters the former balance, particularly if the vacuum in the gas recovery system is not properly adjusted to account for variable gas flows in different areas of the site. The sensitivity of landfill emissions to atmospheric pressure before and after gas recovery would be similar if the distribution of gas recovery matched the distribution of emissions. However, the landfill gas collection system was not optimized for maximum recovery at the time of our measurements and therefore out of balance. This condition was the result of a contractual agreement to collect a constant quantity of gas for electricity production that was well below the gas generation capacity of the landfill.

A significant enhancement of gas generation after the installation of the recovery system was also apparent from our data. We used the relationship between emissions and atmospheric pressure from Figure 1 to determine the magnitude of the enhancement by estimating annual emissions during 1994 (pre-recovery) and 1997 (post-recovery). The full range of surface atmospheric pressure recorded in the Nashua area during 1994 and 1997 is represented by the frequency diagram in Figure 3.   These data can be used as the independent variable in a model based on the linear regression of our measured emission values. But, because our emissions measurements encompass a pressure range smaller than the full range of measured pressure over the 1-year periods, it is necessary to make some simplifying assumptions.

In the post-gas recovery data, our measurements at pressures greater than 1020

mbar suggest that the observed decrease in emissions does not continue linearly to zero as indicated by the regression. But, maintaining a constant emission of 7000 l min^{-1} in the model at pressures exceeding 1023 mbar (our highest measurement pressure) increased the average annual emissions estimate by only 2.5% when compared to extrapolating the regression model to zero. So, the choice of extrapolation was selected for high pressures. A conservative approach was also adopted for low pressures. No emissions measurements were performed at pressures less than 1007 mbar and we do not know whether the observed linear response of emissions would continue in the lower pressure range. Therefore, the linear regression was extrapolated to 1005 mbar and held constant at 27000 l min^{-1} at lower pressures.

A similar approach was used to model the pre-recovery data. Emissions at atmospheric pressures higher than our maximum measurement pressure, 1028 mbar, were estimated by extrapolating the regression line. Also, no emissions measurements were performed at pressures less than 1011 mbar. So, the linear regression was extrapolated to 1005 mbar and held constant at 26000 l min^{-1} for lower atmospheric pressures.

CH_4 emissions were calculated for each hour using the resulting model and the atmospheric pressure data. These values were summed to calculate total annual emissions. The estimated annual CH_4 emission rate during 1994 was 10.1×10^6 m^3 CH_4 year^{-1}. The estimated annual CH_4 emission rate during 1997 was 7.6×10^6 m^3 CH_4 year^{-1}.

We can also estimate gross CH_4 production using the previously discussed whole landfill CH_4 oxidation measurements. The reported oxidation rates ranged from 5% in March 1997 to 13% in August 1996. If we accept these values as CH_4 oxidation extremes, then the average of 9% can be assumed to be reasonably representative of the

annual oxidation rate.

Increasing the estimated emission rates by 9% yields a production rate of 11.1 x 10^6 m^3 CH_4 $year^{-1}$ in 1994, before the initiation of gas recovery. Applying the 9% increase to 1997 emissions yields a rate of 8.4 x 10^6 m^3 CH_4 $year^{-1}$. Including an average 1997 gas recovery rate of 7.1 x 10^6 m^3 CH_4 $year^{-1}$ (13450 l CH_4 min^{-1}) yields a total CH_4 production rate in 1997 of 15.5 x 10^6 m^3 CH_4 $year^{-1}$, or a 40% increase in CH_4 production after the initiation of gas recovery.

CONCLUSION

Two conclusions can be drawn from our data. First, removing gas from the body of the landfill increases gas production. This is intuitively reasonable from the aspect of biochemical kinetics; reducing the concentration of reaction products from the extracellular environment increases the reaction rate. Second, any future credit-trading program that requires the net accounting of CH_4 through all potential pathways must require the characterization of fugitive emissions under the full range of atmospheric variables. While atmospheric pressure appears to be the primary determinant of emissions, other factors that were not included in our study may play important roles. For example, the rate of change of atmospheric pressure, the infiltration of precipitation, and different landfill cover characteristics may significantly effect short-term gas emissions. Basing an assessment of landfill emissions on a single day of measurements could result in a large monetary loss. Determining the losing party in the transaction would depend on the day.

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Captions

Table 1 - Landfill CH_4 emissions collected in 1994, 1996, and 1997. The CV is presented in parentheses next to each emission value. Gas Recovery Status indicates whether a measurement was performed before or after the installation of the gas recovery system.

Figure 1 - CH_4 emissions as a function of atmospheric pressure measured during each test. The open circles (○) represent emissions measured in 1994 (pre-recovery). Solid squares (■) are data from August 1996, solid circles (●) are data from February 1997, and solid triangles (▲) are data from March and April 1997; all after the installation of a gas recovery system. The error bars represent the CV of each test. The solid lines are the linear regressions of these data.

Figure 2 - CH_4 emissions as a function of pressure measured in 1996 and 1997 after the initiation of gas recovery. Solid circles (●) are emission values as measured, open circles (○) are estimated emission values before cover soil oxidation. The solid line is the linear regression of the measured emission values and the dashed line is the linear regression of the pre-oxidation estimated emission values.

Figure 3 - Map of emissions from the landfill surface based on linear interpolation of the surface enclosure measurements from Czepiel et al. (1996). CH_4 emissions are represented on the vertical axis in grams of CH_4 per square meter per day.

Figure 4 – A frequency diagram of surface atmospheric pressure in the Nashua area during 1994 (solid bars) and 1997 (open bars). The diagram consists of hourly averaged pressure values binned in 2 mbar increments. The 1994 and 1997 data are normally distributed with means of 1015 and 1014 mbar, and standard deviations of 8.3 and 9.2, respectively.

Table 1

Test Date	Methane Emissions ($l\,CH_4\,min^{-1}$)	Atmospheric Pressure (mbar)	Gas Recovery Status
10/07/94	17835 (11%)[1]	1020	None
10/12/94	11080 (24%)[1]	1028	None
10/21/94	13920 (12%)[1]	1025	None
11/06/94	21575 (9%)[1]	1011	None
11/12/94	15120 (11%)[1]	1024	None
8/22/96	7924 (10%)	1019	Active
8/24/96	7317 (14%)	1020	Active
8/27/96	7436 (7%)	1021	Active
2/25/97	13588 (12%)	1016	Active
2/27/97	11576 (15%)	1018	Active
3/01/97	17103 (10%)	1012	Active
3/04/97	20970 (9%)	1010	Active
4/15/97	21937 (11%)	1008	Active
4/17/97	26542 (9%)	1007	Active
4/19/97	7749 (17%)	1023	Active
4/22/97	12696 (18%)	1017	Active
4/24/97	13028 (4%)	1015	Active

1: data from Czepiel et al., 1996.

Figure 1
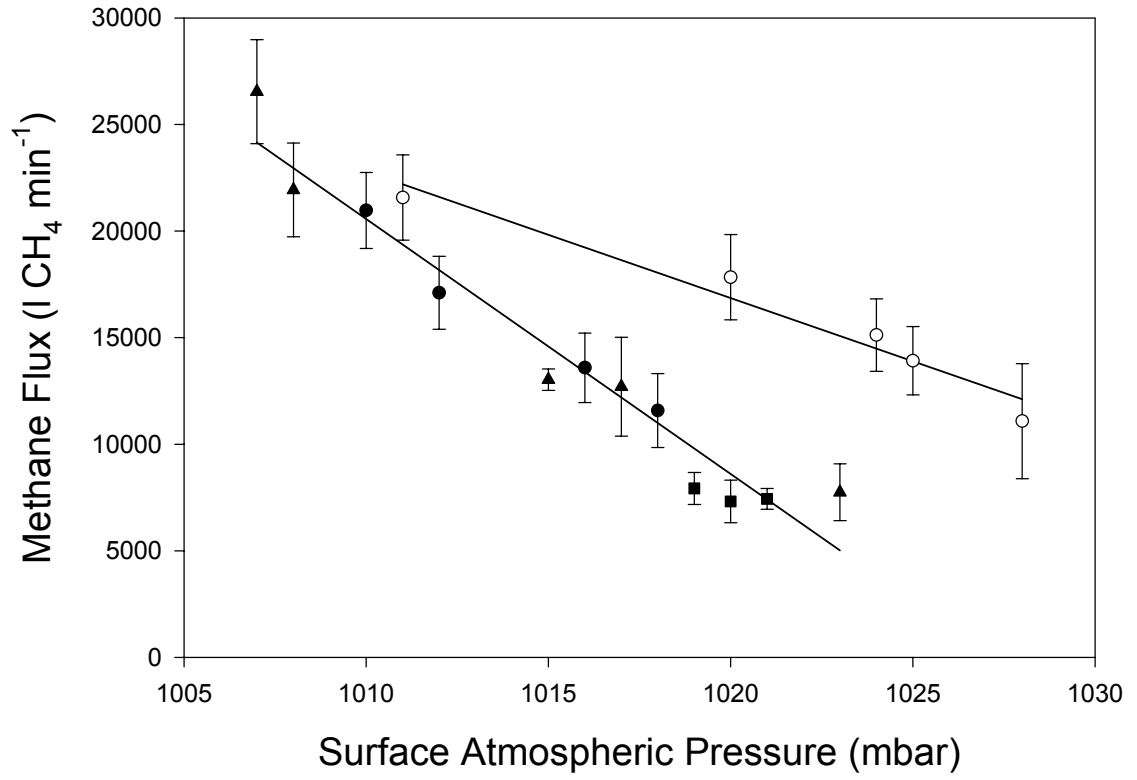
Figure 2

Figure 3

Figure 4