Geotechnics Of Methane Oxidation In Landfill Cover Soils

Jeffrey P. Chanton[1*], Tarek Abichou[2], Gary Hater3 and Roger Green[3] and Jean Bogner[4]

1. Department of Oceanography, Florida State University, Tallahassee, Fl 32306-4320. corresponding author, jchanton@fsu.edu
2. FSU-FAMU School of Engineering, Department of Civil Engineering, Pottsdammer Road, Tallahassee, Fl 32310.
3. Waste Management Inc. 2956 Montana Ave Cincinnati, Ohio 45211, rgreen2@wm.com
4. Jean Bogner, Landfills +, Inc, Wheaton, Illinois

ABSTRACT

Microbial methane oxidation in landfill cover soil is a very effective approach for reducing emissions from landfills. Oxidation of methane may be enhanced by the application of materials present on site, such as yard waste or compost. Engineers require an method to quantify methane oxidation in different types of covers. In this paper we will present a simple, effective stable isotope technique for the evaluation of cover soil methane oxidation. The approach exploits systematic variations in the ratio of $^{13}C/^{12}C$ in CH_4 prior to and following exposure to methane oxidizing microbes in the soil. The action of the bacteria increases this ratio, due to their preference for utilizing $^{12}CH_4$ rather than $^{13}CH_4$. The shift in the ratio following oxidation is proportional to the amount of CH_4 oxidized.

INTRODUCTION

Sources of CH_4 to the atmosphere include wetlands, rice agriculture, coal and gas mining, landfills, termites, and ruminants. Most atmospheric CH_4 sources are associated with human activity and could be attenuated by proper management. The imbalance between sources and sinks in the global CH_4 budget is less than 6% of the total global source (Dlugokencky et al., 1994a; Dlugokencky et al., 1994b; Etheridge et al., 1998) so a small decrease in methane emissions could result in stabilization of atmospheric CH_4 concentrations or even better, a reduction (Lelieveld et al. 1998). As CH_4 is a more potent greenhouse agent than CO_2, lowering the atmospheric CH_4 concentration may be a very realistic and worthwhile goal. The relatively short residence time of CH_4 in the atmosphere (7-10 years) relative to CO_2 and N_2O means that the effects of mitigation efforts would be rapidly observed Thompson et al., 1992).

Landfills are responsible for about 3-7% of global CH4 emissions (Lelieveld et al., 1998; Bogner and Matthews, 2003) and are among the largest anthropogenic CH_4 sources in the United States (US-EPA, 2007). Landfills may be thought of as point sources of CH_4 to the atmosphere and therefore they make good targets for mitigation. At older and smaller landfills without gas collection systems a considerable fraction of CH_4 emissions pass through the soil where they can be reduced by soil methanotrophic bacteria (Chanton and Liptay, 2000; Stern et al., 2006; Abichou et al., 2006a,b; Barlaz et al., 2004). Passive vents at these sites can be treated with biofilters (Powelson et al., 2006, 2007, Gebert and Groengroeft, 2006). At large modern landfills, gas capture for power generation or flaring reduces methane emissions considerably. But some CH_4 also escapes these landfills through the soil and through leaks in the gas collection system. Recently, the technique of using methanotrohic bacterial to reduce methane release has received considerable attention (Huber-Humer, 2004) including recognition from environmental

agencies in Finland and Germany. Currently, the default value for landfill cover CH_4 oxidation is set at a much relatively value, between 0 and 10% of emitted CH_4 (IPCC, 2006, USEPA, 2004). This value was based on seasonal results for a New Hampshire landfill as determined by the studies of Czepiel et al (1996a,b). Recently Chanton et al. (2009) reviewed the literature and compiled methane oxidation results for 42 determinations of the fraction of methane oxidized from the literature following and including Czepiel's landmark study and reported a mean value of 36 ± 6% for this parameter. Fifteen seasonal studies ranging from latitude 30^o to 55^o N yielded a similar value of 35 ± 6%.

We report here a stable isotope approach for the determination of methane oxidation in landfill cover soils. There are two stable isotopes of carbon, ^{13}C which is about 1% abundant and ^{12}C which comprises 99% of carbon atoms. Stable isotopes are useful for determining CH_4 oxidation because as it occurs, the remaining CH_4 becomes ^{13}C enriched due to preferential utilization of the lighter ^{12}C isotope by bacteria (Coleman et al, 1983). Carbon isotopic composition is expressed in the δ notation, which is defined as follows:

$$\delta^{13}C‰ = ((R_{sample}/R_{standard})-1)*1000 \quad (1)$$

where R_{sample} is the $^{13}C/^{12}C$ ratio of the sample and $R_{standard}$ is the $^{13}C/^{12}C$ ratio of the marine carbonate standard (PDB, 0 ‰). Typical biogenic CH_4 is produced at values below -50‰. Following oxidation, CH_4 may exhibit ^{13}C enriched values of -30 to -50‰. Typical organic matter is ^{13}C enriched relative to CH_4 with a $\delta^{13}C$ value of -25‰. The negative δ value indicates that the sample is ^{13}C depleted relative to the carbonate standard. The more negative the value, the more ^{13}C depletion is indicated.

Recent publications which quantify landfill cover soil oxidation using stable isotopes include Bergamaschi et al., 1998; Liptay et al., 1998; Chanton et al., 1999; Chanton and Liptay, 2000; Borjesson et al., 2001; 2007; Christophersen et al., 2001, Abichou et al., 2006a,b; Stern et al 2007; Chanton et al 2008a,b). Significant isotopic fractionation occurs when methane is oxidized. Microbial culture studies have shown that methanotrophic organisms preferentially consume lighter isotopes, leaving residual CH_4 enriched in ^{13}C (Coleman et al., 1981; Barker and Fritz, 1981; Powelson et al., 2007). If one knows the preference of the bacteria for the lighter isotope $^{12}CH_4$ then one may estimate the extent of oxidation from the isotopic difference between the unaffected and the residual (or left over) methane which has been exposed to oxidation but not itself oxidized.

This method can be applied to evaluate methane oxidation in landfill covers, and to contrast differing cover materials with respect to their ability to oxidize methane, or for biofilters (Powelson et al., 2006, 2007). For example, Chanton and Liptay (2000) compared methane oxidation between two treatments. One was a clay cover soil and the other included 6 inches of additional mulch/topsoil which was applied over the clay. They found that the mulch/topsoil oxidized 55 +/- 14% of methane while the clay alone averaged 33% +/- 13% oxidation (Fig. 1). We suggest that this method may find broad application in the evaluation of methane oxidation in landfills and in the design of cover soils to attenuate methane emissions.

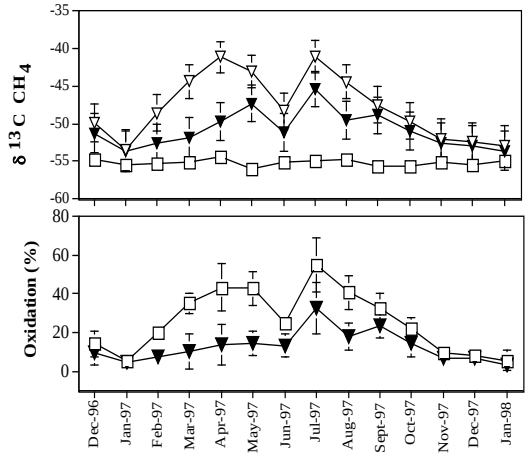

Figure 1. In the upper panel are $\delta^{13}C$ values of methane emitted from mulch soil (open triangles), emitted from clay soil (filled triangles) and collected from within the landfill deep anoxic zone (open squares) as a function of time of year. In this paper we will explain our approach for calculating the % oxidation of methane from the difference between anoxic zone and emitted methane. Percent oxidation is plotted in the lower graph, where the open symbols represent mulch soil and the closed symbols represent clay soil. The presence of the mulch fostered methane oxidation (redrawn from Chanton and Liptay, 2000).

APPROACH & METHODS

Our technique for the in situ determination of methane oxidation is based upon measuring the difference in $\delta^{13}C$ between deep, anoxic zone methane which is not affected by oxidation and that emitted from the landfill cover soil which has been subjected to oxidation (Figure 2). Combined with measurement of the preference of the bacteria for $^{12}CH_4$ relative to $^{13}CH_4$, α (see Chanton et al., 2008b), we can offer a quantitative estimate of the fraction of methane oxidized as it passes through the landfill cover soil. Emitted methane can be captured in chambers (Liptay et al., 1998; Chanton and Liptay, 2000; Borjesson et al., 2001; Christophersen et al., 2001) or in downwind plumes, which integrate the activity of the entire landfill (Chanton et al., 1999, Bergamaschi et al., 1998). Alternatively, methane oxidation can be measured in the soil by collecting soil gas profiles (Bergamaschi et al., 1998, Chanton et al., 2008a).

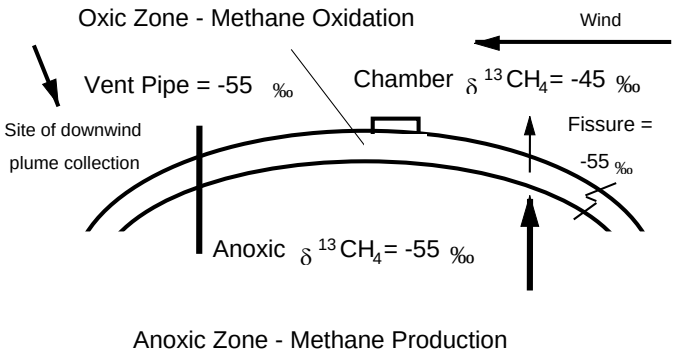

Figure 2: Diagram showing the main means of CH_4 escape from landfills: 1) escape through fissures and vents, which is measured through downwind plume sampling, and 2) transport through the soil cap, which is measured utilizing the chamber technique and the downwind plume sampling method. Methane is produced in the landfill interior. Methane oxidation, however, occurs in the outer rind of the landfill where O_2 penetrates. Values are for the $\delta^{13}C$ of CH_4 in ‰. The diminishing vertical arrows on the right hand side of the figure indicate the attenuation of the methane flux from the landfill by CH_4 oxidation as the gas passes from the anoxic zone through the gauntlet of methanotrophic bacteria in the oxic soil layer. Methane is produced with a d13C value of –55‰. This is the signature of unoxidized methane. As themethane becues oxidized it becomes more positive (e.g. -45‰, Redrawn from Chanton et al., 1999)

Oxidation percentage is determined by the following equation (2) (Chanton et al., 1999), which describes isotopic fractionation in an open system:

(2) $f_o\% = [(\delta E-\delta A)/(\alpha_{ox}-\alpha_{trans})]*1000*100$

where f_o is the % of CH_4 oxidized in transit through the cover soil

$\delta E = \delta^{13}C$ value of emitted CH_4

$\delta A = \delta^{13}C$ value of anoxic zone CH_4

α_{ox} is the isotopic fractionation factor for bacterial oxidation

α_{trans} is the isotopic fractionation factor associated with gas transport.

Liptay et al. (1998) and Bergamashi et al. (1998) have argued that gas transport across the soil cap is dominated by advection, so $\alpha_{trans} = 1$.

The bacterial fractionation factor (α_{ox}) associated with methanotrophy is determined by incubating soils samples at *in situ* temperatures. The fractionation factor is determined with the equation 3 (Chanton et al., 1999, Chanton and Liptay, 2000; Chanton et al., 2008b):

(3) $\delta^{13}C_t = 1000 * (1/\alpha -1)\ln(m/m_o) + \delta^{13}C_{t=0}$

where m/m_o is the fraction of methane remaining at time t

$\delta^{13}C_{t=0}$ is the $\delta^{13}C$ value of the methane at the initial time

and α ($=\alpha_{ox}$) is defined as

(4) $\alpha = k_l/k_h$

where k_l and k_h refer to the rate constants of the light and heavy isotopes respectively.

A time series of analysis is performed to determine the fractionation factor α. Landfill cover soil is placed in a flask and a known concentration of methane is added. These flasks are incubated at outside ambient temperature and two gas samples are taken roughly every day over seven days. The determination of the isotopic composition of these samples permits us to calculate α from equation (3), the fractionation factor inherent to the soil and to its specific microbial flora.

Anoxic zone methane (δA, equation 2) can be captured in several ways: from pipes used to capture methane, from gas ventilation pipes, and from bubble streams which may be found near the edges of cells.

Emitted methane (δE, equation 2) is collected from the air over the landfill at night, downwind of the landfill or from the headspace of chambers placed over the landfill soil. The measured CH_4 $\delta^{13}C$ value of such samples are corrected for the presence of background, or ambient, CH_4 through mass balance to obtain the $\delta^{13}C$ of excess CH_4 ($[CH_4]_{xs}$, that methane added to ambient air by landfill processes, using the following equation:

(5) $[\delta CH_4]_{xs}=(([CH_4]_{(meas)} *[\delta CH_4]_{(meas)})-([CH_4]_{amb}*[\delta CH_4]_{amb}))/ ([CH_4]_{(meas)}- [CH_4]_{amb})$

Where $[CH_4]_{(meas)}$ and $[\delta CH_4]_{(meas)}$ represent the concentration and $\delta^{13}C$ values of CH_4 in the downwind plume or chamber CH_4 and $[CH_4]_{amb}$ and $[\delta CH_4]_{amb}$ represent the concentrations and $\delta^{13}C$ values of background air measured upwind of the landfill. These ambient values (collected upwind) represent the concentration and isotopic composition of CH_4 in air at ground level in the region. Also, $([CH_4]_{(meas)}- [CH_4]_{amb}) = [CH_4]_{xs}$.

When chambers are used to collect methane, it is useful to determine the rate of methane emission into the chambers. At a minimum, one must be sure that methane is accumulating within the chambers over time. To do this, gas samples are taken every 5 minutes over an 20 minutes, using syringes. Following gas analysis in the laboratory, a plot of concentration vs time is made for each flux and the slope of the best fit linear regression taken as dC/dt in the following equation:

(6) $J=(dC/dt)(V/A)$

where J= flux (mg CH_4 m^{-2} d^{-1})
dC/dt = change in concentration over time
V = volume of chamber (cm^3)
A = surface area under chamber (cm^2)

Isotope samples are collected from the chambers at the initiation and the end of the experiment. We use 50mL syringes and inject samples into evacuated vials for transport to

Florida State University. Sample vials are pressurized by multiple injection. Soil gas samples are obtained with a gas tight probe which is hammered into the soil to discrete depths. Samples are withdrawn from the probe using syringes through a septum port and gas is injected into evacuated vials as described above.

Gas Analysis

For methane concentrations below 1%, including plume samples, chamber samples and some probe samples, gas concentrations are determined on a gas chromatograph with a flame ionization detector (FID), a 1 mL sampling loop, and a 2-m 1/8 inch diameter stainless steel column packed with Carbosphere. N_2 and [O_2 + Ar] and higher methane concentrations are determined on a gas chromatograph with a thermal conductivity detector (TCD). Scott Specialty gases are used as standards.

Methane stable isotope ratios are determined using a Finnegan Mat Delta S-Gas Chromatograph Combustion Isotope Ratio Mass Spectrometer (GCC-IRMS) following methods adapted from Merrit et al. (1995). For methane concentrations below 700 ppmv, a cryogenic focusing device is used on the front end of the gas chromatograph. The standard deviation of replicate analyses is generally about 0.15‰.

When methane concentrations are above 700 ppmv, stable isotopic ratios are determined using direct injection on the GCC-IRMS. Very high concentration samples are diluted to 1% CH_4 by addition of nitrogen. Samples are then analyzed by injecting 0.1 to 0.5 ml of sample into the GCC-IRMS inlet system (Merrit et al., 1995).

RESULTS & DISCUSSION

A case study of a landfill, referred to as Landfill "A"

In this paper we will discuss emitted methane and % oxidation results from chamber samples to illustrate this technique. We will compare oxidation in a finished landfill cover (0.5 m thick topsoil over 1 m compacted clay separated with a geotextile membrane and with a gas extraction system) with a temporary covered area (30 cm sandy clay with a gas extraction system). First we must know the isotopic composition or $\delta^{13}C$ value of methane in the deep anoxic zone where there is no aerobic methane oxidation. Then we will look at isotopic $\delta^{13}C$ values of methane after it has passed across the gauntlet of methane oxidizing bacteria in the soil cover as captured in chambers. With a knowledge of the isotopic fractionation factor, α, we can calculate the % oxidation from this shift.

Anoxic zone methane (δA)

Anoxic zone methane was sampled at a gas well in the temporary covered area, in the finished cell area and at gas pipes leading to the flair. This anoxic zone methane represents the isotopic composition of methane before it is acted on by methanotrophic bacteria. Anoxic zone gas sampled in the finished cell was –59.20 ± 0.80, and –60.29 ± 0.32‰ in the temporarily covered area. Gas sampled from pipes directly before the flair was not different in $\delta^{13}C$ (-58.90‰ to –60.15‰, Table 1) indicating that little CH_4 was being lost to oxidation within the collection system. Values for the anoxic zone methane from each area, temporary and finished cell, were used in Equation 2 (and represent δA) to calculate the methane oxidation.

Emitted Methane Captured in Chambers, Flux Rates and % Oxidation

Emitted methane isotopic values include atmospheric and chamber samples, but for simplicity, only chamber samples will be considered here. Twenty chamber experiments were

conducted; sixteen focused on emissions from the top of the finished cell with four measurements conducted on the temporary covered area. CH_4 fluxes were determined by collecting samples sequentially over time.

The flux of CH_4 across the surface of the landfill within different zones was 123 ± 55 and 2364 ± 901 mmol CH_4/m^2d for the finished cell and the temporary covered area respectively. The $\delta^{13}C$ (isotopic composition) of excess methane (total methane corrected for ambient or background methane , equation 5) for each chamber was determined. Replicate measurements were performed to determine analytical uncertainty. Methane $\delta^{13}C$ varied from –57.2‰ to –61.9‰ in the temporary covered area, and –30.2‰ to –59.8‰ in the finished cell area. The more positive values in methane emanating from the finished cell indicate greater methane oxidation.

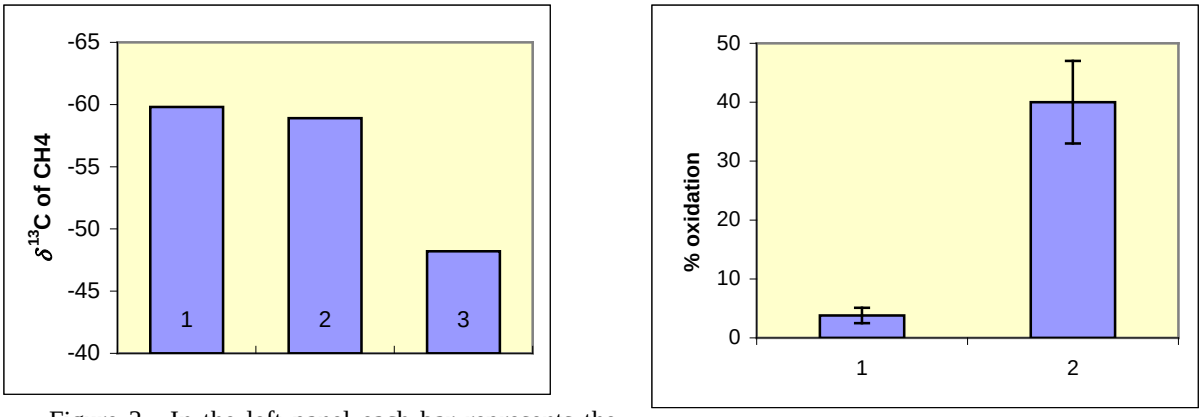

Figure 3. In the left panel each bar represents the isotopic value of methane. Bar 1 is the average of anoxic zone, or unaltered methane. Bar 2 is methane emitted from the surface of the temporary covered area. Bar 3 represents methane emitted from the finished soil cover. Obviously, the value of Bars 1 and 2 are similar, while Bar 3 has been shifted by the activity of methane oxidizing bacteria. The magnitude of this shift is proportional to the extent of methane oxidation, which is shown in the right panel for the temporary cover (Bar 1) and the finished cover (Bar 2).

From these values of excess methane and the anoxic methane $\delta^{13}C$ values given above, and a knowledge of the isotopic fractionation factor of the bacteria which was determined to be 1.03, we calculate % oxidation (with Equation 2) values that range from 0.2 to 6.1% in the temporary covered area and from 0 to 96% in the finished cell. Average values for % oxidation were 3.8 ± 1.3% and 40 ± 7% for the temporary cell and the finished cell respectively (Fig 3).

The greater oxidation in methane released at the surface from the finished cell was also consistent with gas samples drawn from within the landfill with probes. Methane sampled from within the cover soil atmosphere with a gas probe sampler was ^{13}C enriched in the finished cell relative to both methane within the temporary covered area and the anoxic zone methane (Figure 4).

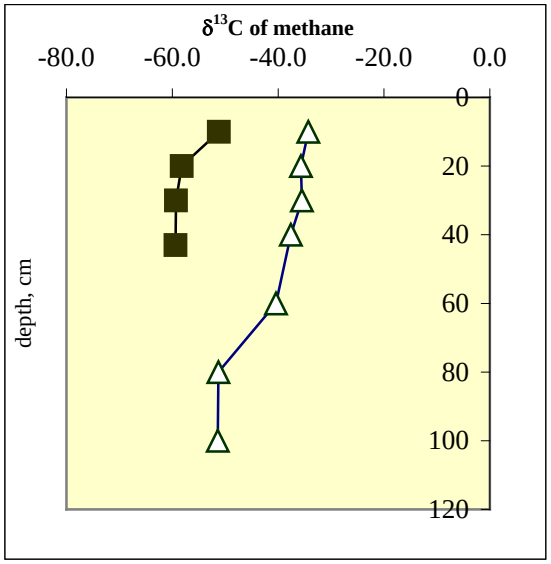

Figure 4. The isotopic composition of methane in probe samples as a function of depth in the landfill soil. The air in pore spaces in the finished cover contains ^{13}C enriched methane relative to the temporary cover indicating greater methane oxidation in the finished cover material. The open symbols represent the finished cell cover, while the filled symbols represent the temporary cell cover.

In conclusion, we have presented an isotopic method which may be used to determine differences in landfill cover soil oxidation. This method will be particularly useful to evaluate methane oxidation in landfill covers, and to contrast or evaluate differing covering materials. We suggest that this method may find broad application in the evaluation of methane oxidation in landfills and in the design of cover soils to attenuate methane emissions.

Acknowledgments. This work was supported by the National Science Foundation and Waste Management, Inc.

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