DEVELOPING A NEW FIELD-VALIDATED METHODOLOGY FOR LANDFILL METHANE EMISSIONS IN CALIFORNIA

K. SPOKAS*, J.BOGNER**, J.CHANTON***, AND G. FRANCO****

** U.S. Dept. of Agriculture (USDA) – Agricultural Research Service (ARS), St. Paul, Minnesota USA*
*** Landfills +, Inc., Wheaton, Illinois, and University of Illinois Chicago USA*
**** Florida State University, Tallahassee, Florida USA*
***** California Energy Commission, Sacramento, California USA*

SUMMARY: This project was initiated in the US by the California Energy Commission (CEC) in cooperation with the California Integrated Waste Management Board (CIWMB) and the Air Resources Board (ARB) to develop improved methods for landfill methane emissions for the California greenhouse gas inventory. This 3-year project (2007-2010) is developing a science-based, field-validated inventory method which links site-specific information on cover types and landfill gas recovery to climatic models, soil microclimate models, and one dimensional (1D) modeling of methane emissions inclusive of methanotrophic methane oxidation. Project tasks include (1) the development of a web-based JAVA template for collection of limited site-specific information; (2) incorporation (with modifications) of existing field-validated climate and soil temperature and moisture models; and (3) 1D modeling of seasonal methane oxidation and emissions for each cover type. The final product will incorporate (1) through (3) in a freely-available web-based JAVA tool. This type of methodology is a Tier IV methodology in the current IPCC National Inventory Methodology for Waste (IPCC, 2006) using advanced site-specific modeling tools with field-validation. The current project includes extensive field validation at two California sites (Scholl Canyon; Marina) over a 2-year cycle (>850 emission measurements and >2000 laboratory incubations); in addition, data from three additional California sites being monitored by Waste Management Inc. has been made available to the project. Project methods, which are based on expansion of previously-published field-validated modeling, emissions measurements, and supporting laboratory studies, represent a departure from a 20-year history of estimating emissions based on first order kinetic models focusing on methane generation. Whereas the first order models were the state-of-the-art when there were few field and laboratory studies of landfill methane emissions and oxidation, we can now develop an improved science-based methodology based on site-specific soils, the presence or absence of active gas recovery under various cover materials, and methane oxidation rates which vary seasonally with changes in soil moisture and temperature. Currently, the model is in the beta testing stage and is especially promising for significantly improving the annual quantification of methane oxidation in landfill cover soils.

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1. INTRODUCTION

In the US, estimation of annual national landfill methane (CH_4) emissions began more than a decade ago in the context of U.S. participation in the United Nations Framework Convention on Climate Change (UNFCCC). National greenhouse gas (GHG) inventories through the UNFCCC rely on the evolving Intergovernmental Panel on Climate Change (IPCC) national GHG inventory methodologies (the latest is IPCC, 2006). Overall, landfill methane contributes less than 2% of global anthropogenic GHG emissions (Rogner et al., 2007; Bogner et al., 2007) and U.S. greenhouse gas emissions (U.S. EPA, 2008). With respect to methane emissions alone, current U.S. inventories (U.S. EPA, 2008) indicate that landfills are the second largest source (after enteric fermentation from ruminant animals).

Historically and by necessity, annual GHG emission inventory methods require certain simplifications over monitoring methods for site-specific regulatory compliance. Inventories are typically done on a national or regional basis once a year for large numbers of diverse sites. It is neither feasible nor desirable to incorporate short-term, site-specific operational issues which are the subject of intensive regulation at solid waste disposal sites in many countries. Practically speaking, the most detailed inventory methodologies to date have incorporated only generalized regional differences for assumed landfill gas generation and emission rates.

From a scientific perspective, both types of methodologies can benefit from periodic improvements based on advances in scientific understanding through basic and applied research. With respect to landfill methane specifically, a common element for both types of methodologies has been the use of a first order kinetic equation for methane generation with accompanying assumptions related to the percentage recovered (by engineered landfill gas recovery), percentage oxidized (by methanotropic microorganisms in landfill cover materials), and the "net" percentage emitted to the atmosphere.

The use of first order equations dates back to the mid-1970s when the first commercial landfill gas (LFG) recovery projects began to estimate recoverable LFG at specific sites using first order decay (FOD) models, then fine-tuning the model parameters to match actual gas recovery history (e.g. EMCON, 1980). In the early days of LFG recovery, there were no field measurements of actual surface methane emissions and only a limited number of laboratory studies for LFG generation under various idealized conditions. Later, for regulatory guidance during the 1980s when the U.S. EPA was required to comply with legislative mandates under the Clean Air Act (CAA) Amendments, the same strategy of using a first order kinetic model was adopted. Subsequently, for GHG inventory calculations after the U.S. joined the UNFCCC, an evolution of methods has occurred within the U.S. EPA and their contractors. However, these methods still include the application of a first order kinetic model. The latest 2006 IPCC Inventory Guidelines for landfill methane emissions (IPCC, 2006) rely on "Tier I" default methods based on a multi-component first order kinetic model where the gas generation potential and rate are a function of waste composition and climate. The previous 1996 IPCC guidelines included a default Tier I "mass balance" method which did not take into consideration the time delay for landfill gas generation. However, most developed countries (which must report annual emissions) have typically used a FOD for the last decade or so. Developing countries, which are not currently required to report annual emissions, would have more typically used the mass balance method in the past but will use an FOD method in the future.

To a large extent, the use of the FOD models implies that all of the waste in a designated region is being landfilled in one large uniform & homogeneous landfill site under specific climatic conditions where the waste is decaying homogeneously with production of landfill gas. Monitored (actual) regional or national landfill gas recovery can be subtracted from the modeled generation and a credit for up to 10% methane oxidation in cover materials is allowed. However, there is no recognition of the numerous & complex physical and chemical processes in landfill cover soils, including the basic physical effect of the cover to limit emissions to the atmosphere. Furthermore, climatic factors also alter the activity of the soil methanotrophs. Historically, the 10% allowance for annual oxidation is referenced back to the first study which estimated seasonal oxidation for the Nashua, NH (US) landfill using a climatic model combined with field measurements of methane emissions and laboratory incubations for methane oxidation (Czepiel et al., 1996). Recent isotopic investigations indicate that this value is higher, typically around 35 ± 6% across a variety of latitudes (Chanton et al., 2009).

The current project to develop an improved landfill methane emissions inventory method for California was initiated in 2006, begun in 2007, and will be largely completed during 2009 (end date is February 2010). During the last decade, driven by a need for improvements in engineered LFG recovery systems (including quantification of emissions losses) and, more recently, by the need to more accurately quantify emissions for GHG inventory purposes, a wide range of field emissions measurements and supporting laboratory studies have been completed. We can make use of these recent research results linked to recently-available climatic models and databases to develop improved inventory models. The current (IPCC, 2006) national inventory guidelines for methane from waste also recognize the current state-of-the-science with respect to validated methods for field measurements, an expanded database of field measurements in the refereed literature, and evolving theoretical and empirical models appropriate for national or regional inventories. For the first time, these guidelines permit higher tier methods based on historic field measurements and more advanced models for improved national and regional inventories. This 3-year project for California would thus be consistent with Tier IV methods under the IPCC 2006 guidelines and would be the first such regional inventory methodology for landfill methane.

Project methods are based on expansion of previously-published field-validated modeling and measurement approaches (Bogner et al., 1997, 2000; Chanton and Liptay, 2000; Spokas et al., 2003, 2006), use of currently-collected site-specific data, integration of regional soils and climatic databases into improved models, and field validation at two California landfills over two annual cycles. The two field validation sites are the coastal Marina Landfill (Monterey, CA USA; 36.71^{o}N 121.762^{o}W) and the Scholl Canyon Landfill in the Los Angeles area (Glendale, CA; 34.158^{o}N 118.196^{o}W). Currently, the evolving regulation of landfill methane emissions for "early action" under California Assembly Bill 32 (AB 32) and credits trading through the California Climate Action Registry (CCAR) have also focused attention on improved landfill methane methodologies.

The goal of this project is to develop a JAVA-based web tool to allow prediction of site-specific methane emissions from landfills for various types of cover materials with or without active gas extraction. A data template inclusive of the major California cover types has been developed by the project team and reviewed by the CIWMB and the Air Resources Board (ARB). This template also permits the use of a customized layered sequence of cover materials and inclusion of a geomembrane. Gas emission modeling focuses on emissions as a mass flux per unit area and includes theoretical relationships, empirical factors, and scaling coefficients as appropriate. In particular, seasonal methane oxidation is modeled through the use of regional

soils and climatic databases, incorporation of results from laboratory batch incubations of a wide range of landfill soils under a variety of soil and moisture conditions, and use of supporting stable carbon isotopic methods to quantify field rates of methane oxidation.

2. MODEL STRUCTURE AND DEVELOPMENT

Figure 1 outlines the model structure and subcomponents. It must be emphasized that, for a regional greenhouse gas inventory, as opposed to regulatory compliance monitoring, certain simplifying assumptions must be made. Unlike previous landfill methane inventories, and based on field monitoring experiments conducted over the first year of the project (discussed below), this model is not based on a first order kinetic model for methane generation. Rather, emissions are being modeled directly based on a 1D diffusive flux calculation based on the concentration gradient for methane through the cover materials. The literature supports the assumption that diffusion, rather than advection, is the primary driving force for gas transport through the landfill cover soils (e.g. Molins et al., 2008). Both a review of the literature and the project-specific experiments have indicated that this approach is more straightforward and directly ties the measured emissions to the major driving force for transport (concentration gradient across the cover). By avoiding the modeling of theoretical landfill methane generation, the associated modeling errors and assumptions are thereby reduced. For a convenient comparison with older models, the current version of the model also incorporates a multi-component first order kinetic equation to allow a comparison of the results with the IPCC default methodology (IPCC, 2006).

Because the major controls on landfill methane emissions are the thickness and properties of the cover materials and the implementation of active landfill gas extraction, the model requires site-specific input data (left box in Figure 1). A data template has been developed that includes the major California cover types and ADCs (Alternative Daily Covers), the area of each type of cover materials, whether active landfill gas extraction is currently in place for each type of cover material, and other pertinent site characteristics. The template model also includes a customized user-friendly "cover designer" tool for multiple layers and geomembrane composite covers. As mentioned above (Figure 1), this template is programmed in JAVA for web-based access by site operators. A project goal is to minimize the site-specific information requirements to the most pertinent data needed for this inventory.

The second step is a regional weather simulator (GlobalTempSIM & GlobalRainSIM; Spokas and Forcella, 2009). The latitude and longitude of the site are used to extrapolate the daily climatic conditions from an interpolated database based upon 30-yr (1961 – 1990) air temperature and precipitation records. These two models are described in detail in Spokas and Forcella (2009). In addition, the solar radiation of the location is modeled using the validated SolarCalc model (Spokas and Forcella, 2006). The main purpose of these weather simulators is to incorporate average meteorological data as inputs for the soil moisture and temperature modeling.

As seen in Figure 2, the model does simulate the annual pattern of air temperature at both sites. For the Marina landfill site (Figure 2A), the overall Willmott d-index was 0.85, bias was +0.61 ^{o}C and a root mean square error (RMSE) of 2.2 ^{o}C for the period between January 2007 and January 2009. The value of the Willmott d-index will vary between 0 and 1, with a value of 1 indicating perfect model agreement (Willmott, 1981). For the Scholl Canyon landfill site (Figure 2B), the overall d-index was 0.88, with a bias of -0.02 ^{o}C and a RMSE of 2.9 ^{o}C for the period between January 2007 and November 2008.

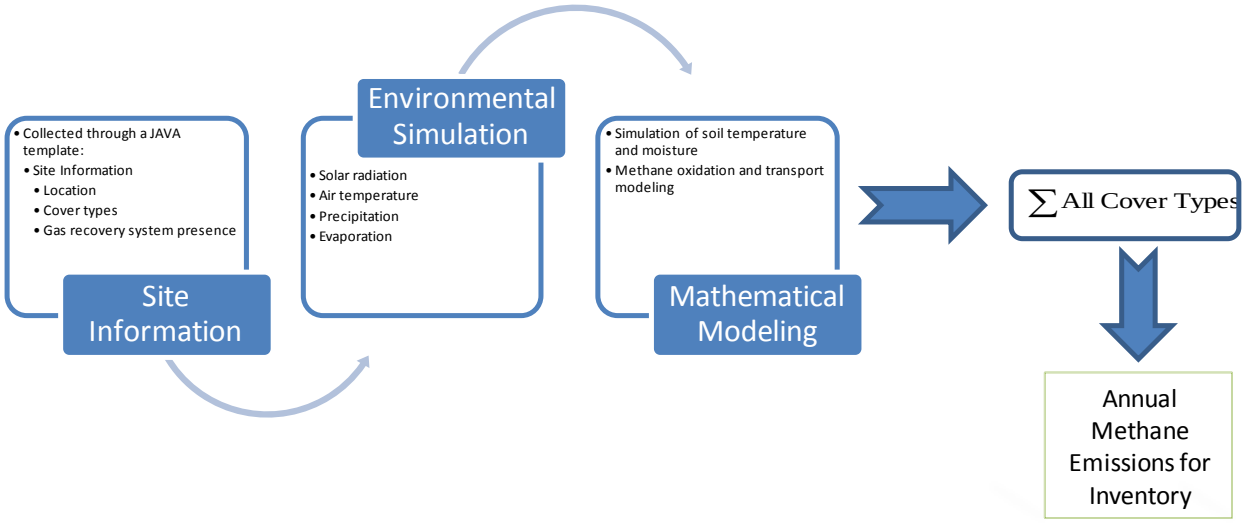

Figure 1. Structure for California landfill methane emissions inventory model

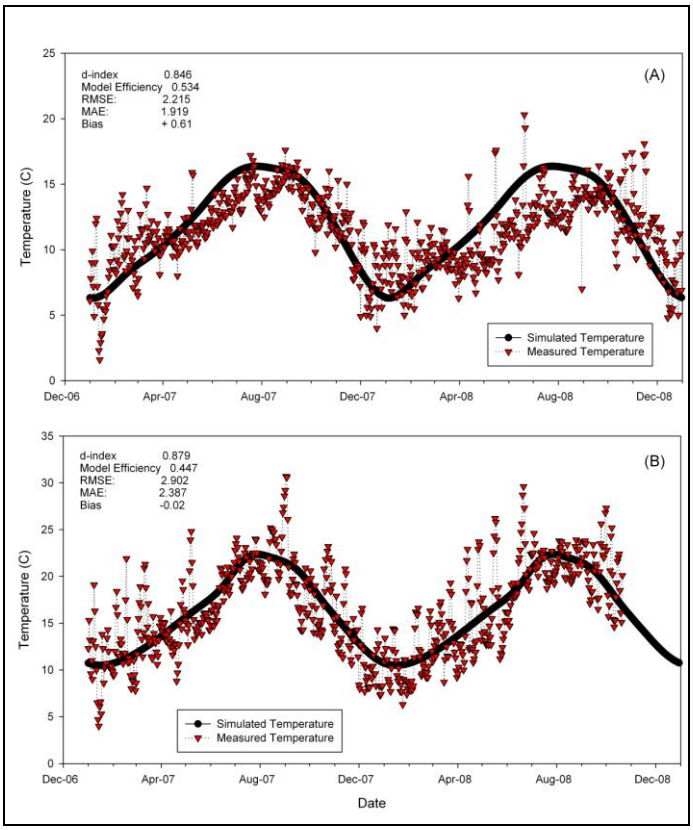

Figure 2. Comparison of simulated daily average air temperature versus measured average air temperature for 2006 to 2008 for A) Marina Landfill (Monterey, CA) and B) Scholl Canyon Landfill (Glendale, CA). Air temperature data at Glendale, CA was unavailable after November 21, 2008.

These statistical measures confirm the ability of the temperature model to provide an accurate estimation (± 2 to 3 ^{o}C) of the annual air temperature cycle at both sites.

The third step is the coupled modeling of soil moisture and temperature along with methane transport and oxidation in the cover soil. The USDA-ARS soil temperature and moisture model (STM^2) is being used to predict soil moisture and temperature as a function of depth in the cover soils. This model has been used successfully in predicting the soil microclimate for weed growth modeling (e.g. Spokas and Forcella, 2009). Continuous temperature and moisture profile data were collected at the field sites to validate the model for landfill cover soils. Figure 3 illustrates the comparison for the Marina final cover area. The 15 cm depth possessed a d-index of 0.93 and a RMSE error of 1.9^{o}C with an average bias of 1.5^{o}C. The 50 cm depth had an improved fit, with a d-index of 0.94, a RMSE of 1.4^{o}C and an average bias of 1.2^{o}C. Other cover types, depths and locations (Scholl Canyon) compared similarly (data not shown). As seen in the figures (2-3), the use of the climatic databases coupled to the soil temperature and moisture model do provide a means of estimating the variations in soil temperatures observed at various depths and times as a function of the climates at both sites.

The final step involves a 1-D diffusion model based on the methane concentration gradient (already demonstrated for CO_2 and O_2 flux estimation: Mitchell et al., 1999; Jones et al., 2000). From Fick's Law:

$$J = D_s \frac{dC}{dz} \approx D_s \frac{\Delta C}{\Delta z},$$

where J is the flux of gas species, $D_s = D_s(\theta,\phi)$ is the soil gas diffusion coefficient that varies with time as a function of soil porosity (ϕ) and volumetric water content (θ), C is the gas

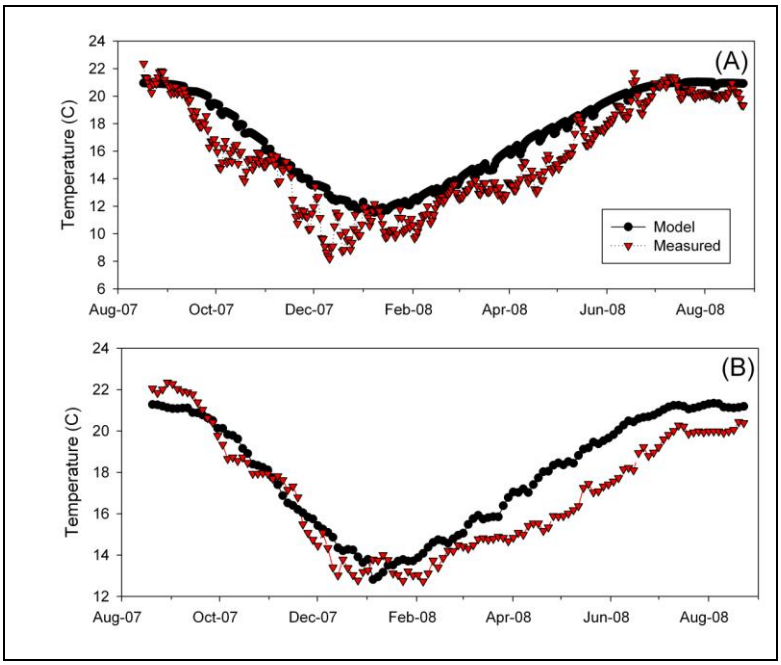

Figure 3. Comparison of modeled temperature predictions versus actual field measurements for locations (A) 15 cm depth in the final cover at the Marina site and (B) 50 cm depth in the final cover at Marina.

concentration, and z is depth. For the flux calculation, the concentration gradient is approximated by discrete differences ΔC and Δz or, in other words, the change in concentration with respect to depth. Moldrup et al. (1999) suggested a soil-type dependent gas diffusivity model (referred to as the Buckingham-Burdine-Campbell equation) for gas diffusivity:

$$D_s = D_{a,T}\left(\phi^2\right)\left(\frac{\theta_{air}}{\phi}\right)^{2+\frac{3}{B}},$$

where $D_{a,T}$ is the free-air diffusion coefficient at temperature T (0.16 cm^2/sec @ 20C), ϕ is the total soil porosity (cm^3 cm^{-3}), θ_{air} is the air filled porosity (cm^3 cm^{-3}), and B is the Campbell B or the slope of the soil moisture retention curve in a log(θ)–log(–Ψ) coordinate system. This model of the soil diffusivity was found to provide better prediction than other models across multiple soil types (e.g. Rolston and Moldrup, 2002; Moldrup et al., 2004). Temperature also influences diffusion and can be accounted for by the relationship:

$$D_{a_T} = D_{a20C}\left(\frac{T}{293K}\right)^{1.75},$$

where D_{a_T} is the free air diffusion coefficient at temperature T, D_{a20C} is the free-air diffusion coefficient at 20°C and T is the temperature (°K) (Jones, 1992). Since we know the soil texture, temperature and soil moisture content of each node at any given time step, the effective diffusivity can be calculated for each layer. If the layer becomes saturated, the diffusion coefficient is multiplied by 10^{-4} to account for the difference between air and water diffusion (Scheffer and Schachtschabel, 1984).

Methane oxidation is being modeled relative to a lower and upper soil moisture boundary which inhibits methane oxidation activity, as well as a lower and upper temperature limits. Laboratory incubations were performed to determine these limits and overall kinetics. Through the laboratory incubations no upper moisture limit was observed, as methane oxidation activity occurred in slurries (10:1 water:soil; maximum evaluated), except at a lower rate. The lower moisture limit, which suppressed 90% of the oxidation activity was approximately -600 kPa. This lower soil moisture limit has not been previously addressed in the literature as a soil matric potential, which is a better way to express moisture contents since it accounts for soil texture interactions. The temperature limits observed were similar to those previously reported in the literature. There also was a lag time (7-10 days) for recovery of methane oxidation rates in cyclic laboratory incubations if the soil was allowed to dry out to less than -600 kPa for 24 hours. These data will be used in conjunction with the modeled soil moisture and temperature profile data to assign a respective rate of methane oxidation for each depth interval through the landfill cover as a function of the soil microclimate. In this fashion, we believe that we will have a more accurate modeling of the methane oxidation potential of the cover soil as a function of both the regional climatic data as well as the specific cover soil type.

3. SAMPLE MODEL OUTPUT & FIELD VALIDATION

Field validation was carried out over two annual cycles at two California sites: the Scholl Canyon Landfill (north Los Angeles) and the Marina Landfill (near Monterey). The Marina site is a northern California coastal site while the Scholl Canyon Landfill is a southern California canyon fill. The validation has included wet and dry season field campaigns which include methane fluxes, soil gas profiles, and the stable carbon isotopes of methane in flux and profile samples to quantify methane oxidation (see Chanton et al., 2008). These are supplemented by

continuous monitoring of meteorological variables, soil moisture, and soil temperature at both sites. However, for the analysis presented here, only the preliminary analyses of the Marina intermediate (high flux area) and final cover areas (low flux) will be discussed (Table 1). These two areas provide a range of the emissions values seen at the two validation sites.

Table 1. Summary of collected field data for the two sites (flux units g m^{-2} d^{-1}). Standard deviation of the measurements is given in parentheses.

Site	March '07	August '07	March '08	September '08
Marina – Final Cover	0.003	0.003	0.029	0.105
	(0.008)	(0.013)	(0.061)	(0.260)
Marina – Intermediate Cover	0.029	41.5	33.1	105.8
	(0.015)	(59.5)	(89.2)	(200.1)

3.1 Marina Intermediate Cover Area

As seen in Table 1, there was a steady increase in the methane emissions from the Marina intermediate cover area (50 cm of sandy clay loam soil) starting with low values in March 2007 (0.03 g CH_4 m^{-2} d^{-1}) and then gradually increasing through September 2008 (106 g CH_4 m^{-2} d^{-1}). This increase was directly correlated to the increasing methane soil gas gradient (Figure 4) which is due to the elapsed time from initial placement of the intermediate cover. The intermediate cover was established in 2006, and it was in place approximately 8 months prior to the initial field sampling (March 2007). This increasing flux with time is expected when the intermediate cover remains in place and gas diffusion occurs. The methane concentration in the well gas at the intermediate cover area was between 50-60% methane for the entire time period. Diffusive transport will stop when the concentrations within the cover are equal (no concentration gradient). Data from the isotopic determination of methane oxidation suggested that approximately 24% of the emitted methane was oxidized (from flux chamber sampling) and the soil gas probe data suggested a range of 20 to 60% oxidation occurring. Note that these values represent either the average oxidation for the entire cover (chamber) or the percentage of oxidation up to the soil gas probe depth (10 cm), respectively.

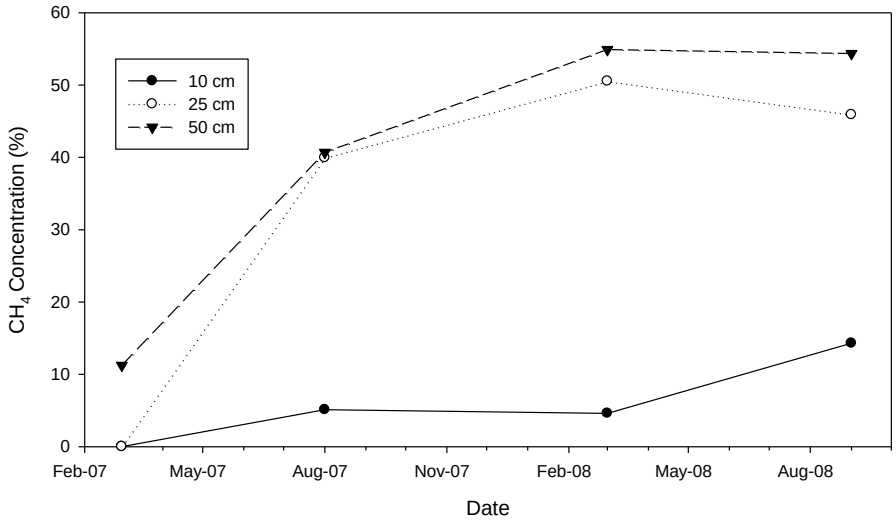

Figure 4. Changes in soil gas concentrations (at 10, 25 and 50 cm) in the intermediate cover area (50 cm total depth) at the Marina landfill during the two year study.

The model illustrates the dynamics of these changes in oxidation behaviour based on the annual cycle of air temperatures and precipitation pattern for the site. Figure 5A illustrates the predicted percent oxidation occurring for 1, 10, and 25 cm depths in the cover at the intermediate site. Note that these percent oxidation values represent only the methane being oxidized in each depth increment (node) and not the overall value that was determined from the isotopic analysis above. These rapid changes in percent oxidation are due to soil moisture limitations, water infiltration, temperature fluctuations and corresponding changes in the diffusive transport of both methane and oxygen through the cover soil. These corresponding alterations in the oxidation rate are thus a major determinant of the net surface emissions.

Figure 6A illustrates the predicted surface emissions with and without methane oxidation. The model prediction of 185 g CH_4 m^{-2} d^{-1} is slightly higher than the 101 g CH_4 m^{-2} d^{-1} observed from the flux chambers. However, the total amount of oxidation predicted from the model (Figure 7A) is also around 13-30% depending on time of year, where the isotopic methodology suggested a value closer to 24% for the seasonal average. This could be attributed to the differences in methane oxidation activity for the particular cover soil. The model utilizes the average oxidation rate for all the soils tested. Generally, the rate of oxidation increases when a soil has been exposed to high levels of methane for extended periods of time (such as >25% at 25 cm; Figure 4). This could explain the lower oxidation % and the higher flux prediction. Despite this difference, the preliminary model results reasonably replicated the field data.

3.2 Marina Final Cover Area

In contrast to the intermediate cover area, the final cover area at Marina (total thickness approx. 2.4 m consisting of sandy loam over clay), was characterized by very low fluxes (with 25-50% of all flux measurements indicating atmospheric uptake). The highest single flux value for the final cover area was approximately 1 g m^{-2} d^{-1}. Due to the low surface flux values and low soil gas concentrations, the isotopic oxidation method was not able to be applied at this area. The model predicted very low fluxes as well (Figure 6B). However, the model result is slightly higher at 2-4 g CH_4 m^{-2} d^{-1}. Overall our initial modeled result matches the field measurement within the same order of magnitude. Again, this could indicate that the rate of oxidation in the model should be increased. However, given the fact that the oxidation rate used comes from the average of all cover types across the two different sites, this does represent a conservative estimate for inventory purposes. The interesting point is that the 4.2 m cover would have only an emission rate of 4 to 18 g CH_4 m^{-2} d^{-1} even without methane oxidation activity. However, with methane oxidation the flux pattern changes drastically, with only trace emissions (2-4 g CH_4 m^{-2} d^{-1}) predicted from the modeling during the dry period (day of year 125-300; for this location) while during the rest of the year the cover is not emitting and is actually oxidizing atmospheric methane (methane uptake).

It is also interesting to note how the model captures the different behaviour in methane oxidation between the intermediate (Figure 5A) and final cover (Figure 5B) areas due to different soil textures (sandy clay loam versus a sandy loam). The sandy clay loam is better at retaining moisture (due to the higher number of smaller pores) than the sandy loam. Thus, the sandy clay loam increases the period of time when moisture conditions are favourable for oxidation. Despite this fact, the final cover is still predicted to be oxidizing over 65% of the diffusing methane.

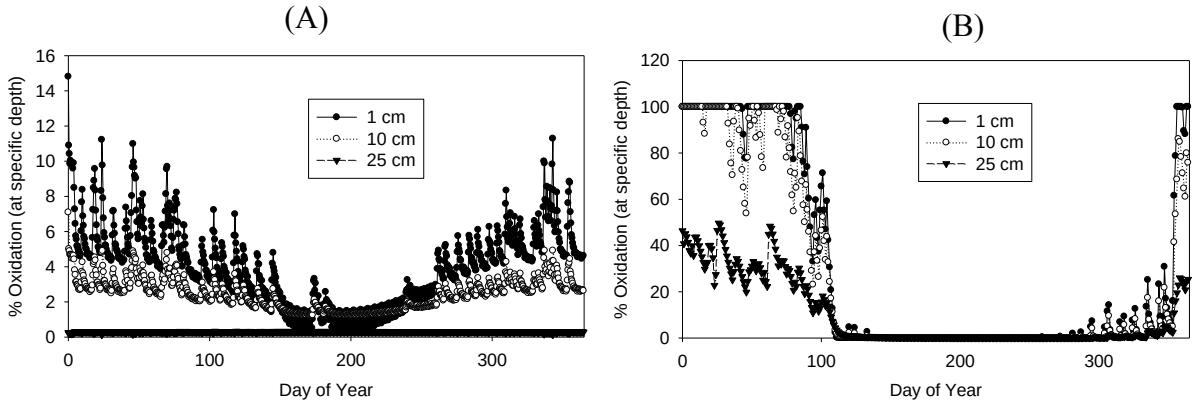

Figure 5. Results from the model prediction of percent oxidation occurring at 1cm, 10cm, and 25 cm throughout the year in the (A) intermediate cover area and (B) final cover at the Marina landfill. Note the higher % oxidation rates in the final cover are largely due to the smaller methane fluxes through the final cover (Figure 6).

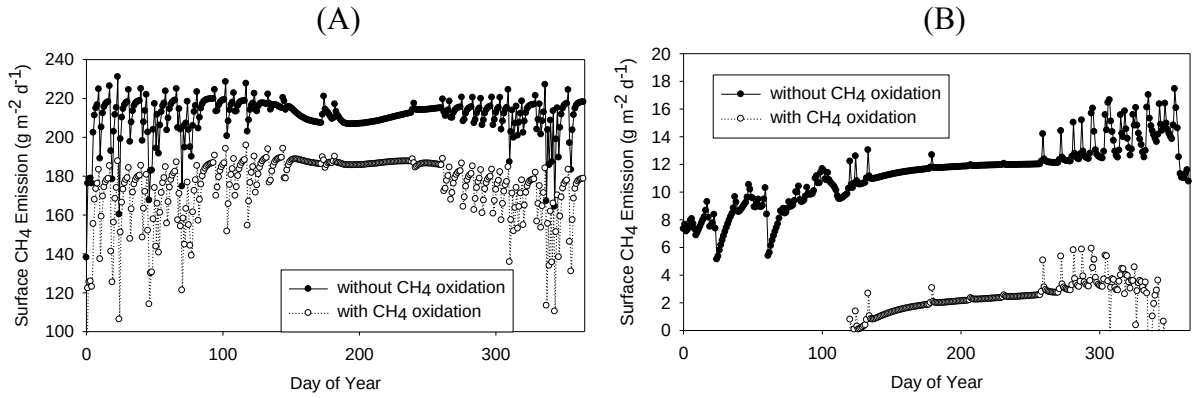

Figure 6. Predicted variations in surface methane emissions for the year at the (A) intermediate and (B) final cover areas (Marina landfill).

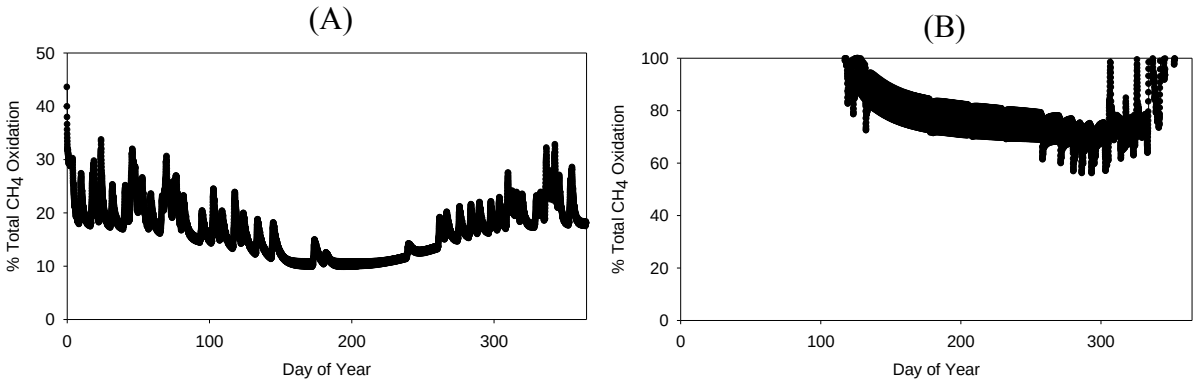

Figure 7. Predicted total percent oxidation for the (A) intermediate and (B) final cover areas at the Marina Landfill.

4. CONCLUSIONS

Based upon the extension of previously field-validated modeling and measurement methods integrated with regional soils and climatic databases, this project is developing an improved methodology for landfill methane emissions for the California greenhouse gas inventory. Previous inventory methodologies for landfill methane have typically relied on a first order kinetic model for theoretical methane generation based on the mass of waste in place and the filling history. This is the first regional inventory methodology for landfill methane emissions which relies on field-validated modeling of emissions as "net" emissions [inclusive of methane oxidation] rather than methane generation. Unlike the methane generation kinetic rate parameters, the inputs for the concentration gradient can easily be measured directly in the field and entered into the model for site specific calibration. If these values are not available, the model has default concentration profiles for the various types of covers based on published and data collected during this project. This more direct approach has fewer uncertainties than the previous indirect modeling approaches based on methane generation and is being directly field-validated through a series of project-specific field and laboratory experiments and simulations, as well as through reference to research results published during the last decade. The latest 2006 IPCC inventory guidelines permit the use of higher tier methods based on historic field measurements and more advanced models. Thus, this methodology for California would be the first such regional inventory methodology for landfill methane and would be considered a Tier IV method under the IPCC guidelines.

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