

A process-based inventory model for landfill CH₄ emissions inclusive of seasonal soil microclimate and CH₄ oxidation

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[1] We have developed and field-validated an annual inventory model for California landfill CH₄ emissions that incorporates both site-specific soil properties and soil microclimate modeling coupled to 0.5° scale global climatic models. Based on 1-D diffusion, CALMIM (California Landfill Methane Inventory Model) is a freely available JAVA tool which models a typical annual cycle for CH₄ emissions from site-specific daily, intermediate, and final landfill cover designs. Literature over the last decade has emphasized that the major factors controlling emissions in these highly managed soil systems are the presence or absence of engineered gas extraction, gaseous transport rates as affected by the thickness and physical properties of cover soils, and methanotrophic CH₄ oxidation in cover materials as a function of seasonal soil microclimate. Moreover, current IPCC national inventory models for landfill CH₄ emissions based on theoretical gas generation have high uncertainties and lack comprehensive field validation. This new approach, which is compliant with IPCC “Tier III” criteria, has been field-validated at two California sites (Monterey County; Los Angeles County), with limited field validation at three additional California sites. CALMIM accurately predicts soil temperature and moisture trends with emission predictions within the same order of magnitude as field measurements, indicating an acceptable initial model comparison in the context of published literature on measured CH₄ emissions spanning 7 orders of magnitude. In addition to regional defaults for inventory purposes, CALMIM permits user-selectable parameters and boundary conditions for more rigorous site-specific applications where detailed CH₄ emissions, meteorological, and soil microclimate data exist.

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1. Introduction and Background

[2] In addition to natural wetlands, atmospheric methane (CH₄) has multiple anthropogenic sources with high uncertainties [Bousquet *et al.*, 2006], including rice production, ruminant animals, natural gas leakages, biomass burning, and landfills. With a 100-year global warming potential (GWP) that is 25 times higher than CO₂ and a short atmospheric lifetime of about 12 years [Forster *et al.*, 2007], reductions in CH₄ emissions from specific sources such as landfills can positively impact atmospheric concentrations within decadal timeframes. According to global estimates summarized in the IPCC 4th Assessment Report, annual landfill CH₄ emissions

of approximately 600–700 Mt CO₂ equivalent yr⁻¹ constitute half the total emissions from the waste sector, or between 1 and 2% of total anthropogenic GHG emissions of about 49 Gt CO₂ eq. [Bogner *et al.*, 2007; Rogner *et al.*, 2007]. Landfill gas, as generated, contains 50–60% CH₄ (v/v). In the absence of controls (such as engineered gas recovery and well-maintained cover materials), landfills can be potent local sources of atmospheric CH₄. Moreover, in both developed and developing countries with a history of landfilling, inventory estimates indicate that landfills can be nationally significant sources of atmospheric CH₄—for example, in the U.S., landfills are currently the third largest anthropogenic source of CH₄, after natural gas systems and ruminant animals [U.S. Environmental Protection Agency, 2011].

[3] Compared to other CH₄ sources, current global estimates for annual landfill CH₄ emissions are especially problematical with high estimated uncertainties up to >200% [Intergovernmental Panel on Climate Change (IPCC), 2006]. For national inventory reporting to the UNFCCC (United Nations Framework Convention on Climate Change), emissions are estimated using IPCC Tier I and II methodologies [IPCC, 2006] based on a first order kinetic equation for landfill CH₄ generation, termed a first order decay (FOD) model. The estimated mass of CH₄ generated in a particular

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year is based on the waste landfilled in that year summed with the predicted CH₄ generated from waste landfilled in previous years. Thus CH₄ generation relies on the annual mass of landfilled waste, assumed or reported waste composition, a CH₄ generation potential ($m m^{-1}$), and a kinetic constant (t^{-1}) for each biodegradable waste component which is assumed to differ with climate (e.g., wet/dry; tropical/temperate). As appropriate for specific countries, two subtractions can also be applied to yield the CH₄ emitted—these are the annual CH₄ recovery from engineered landfill gas recovery projects and a further 10% reduction for methanotrophic CH₄ oxidation in cover materials, based on one older study, *Czepiel et al.* [1996]. Some of the questionable assumptions of the current methodology include the application of a kinetic equation suitable for homogeneous waste decomposition, omission of the physical effect of cover soils on emissions, the use of a single [10%] oxidation factor, the assumption that modeled generation is related to residual emissions at sites with high rates of gas recovery, and the assumption that reliable annual waste data exist for model input [*IPCC*, 2006; *Bogner et al.*, 2007; *Scheutz et al.*, 2009].

[4] Addressing the waste data first, current approaches include: (a) use of data with variable quality and quantity from national waste statistics, surveys, or IPCC guidance documents [*IPCC*, 1996, 2006]; (b) estimates based on population alone [e.g., *Nakicenovic et al.*, 2006]; and (c) because waste generation is related to affluence as well as population, the use of surrogate variables linked to demographic or economic indicators for which national data are annually collected, including per capita gross domestic product (GDP) per capita, energy consumption, or private final consumption [e.g., *Richards*, 1989; *Bogner and Matthews*, 2003; *Mertins et al.*, 1999]. More realistically, annual waste mass and composition data are lacking for many countries and regions, data quality is variable, national definitions are not uniform, and inter-annual as well as site-by-site variability is often not well quantified [*Bogner et al.*, 2007].

[5] Importantly, neither the existing IPCC multicomponent FOD methodology for landfill CH₄ emissions [*IPCC*, 2006] nor the single component LANDGEM methodology used in the U.S. [*U.S. Environmental Protection Agency*, 2005] were ever field-validated for surface CH₄ emissions. Rather, historic model validation consisted of comparing modeled generation to measured gas recovery [e.g., *Peer et al.*, 1993; *van Zanten and Scheepers*, 1995; *Scharff and Jacobs*, 2006; *Thompson et al.*, 2009]. This approach was consistent with the original intended purpose of these models for predicting gas recovery for commercial landfill gas utilization projects. Moreover, when these models began to be applied to emissions more than a decade ago [*IPCC*, 1996, 2006; *Czepiel et al.*, 1996], comprehensive field measurement programs for landfill CH₄ emissions were just beginning. At the site-specific level, use of the first order models as the starting point for emissions estimates becomes especially problematic as there can be large discrepancies between modeled and measured CH₄ pathways. Indeed, the application of the current IPCC model to the two main field validation sites for this project indicated that modeled CH₄ generation [*IPCC*, 2006], using site-specific disposal data and regional California waste composition data, was only a fraction of the currently measured CH₄ recovery.

[6] In general, field and laboratory data over the last decade have demonstrated that both landfill CH₄ emission and oxidation rates can vary by several orders of magnitude in field settings with measured emissions related to the implementation of engineered gas extraction as well as the seasonal properties of site-specific cover materials to retard gaseous emissions and promote methanotrophic oxidation [e.g., *Scheutz et al.*, 2009]. In particular, detailed CH₄ mass balance studies at field scale (7 cells at 3 landfill sites) showed that, while CH₄ recovery could be generally correlated to FOD-modeled generation at sites where waste inputs were well-quantified, there was no correlation between modeled generation and measured emissions, which varied over about 6 orders of magnitude [*Spokas et al.*, 2003; *Bogner and Spokas*, 2010]. Moreover, the 10% default value for CH₄ oxidation value is derived solely from the first study in the literature to quantify annual CH₄ oxidation [*Czepiel et al.*, 1996]. This assessment relied on field measurement of emissions, supporting laboratory oxidation studies, and the application of a seasonal climatic model for a single small U.S. landfill (Nashua, New Hampshire) which did not have engineered gas recovery. A recent review summarizing a variety of lab and field investigations for landfill CH₄ oxidation indicated an average of $35 \pm 6\%$ for landfill cover soils with differing characteristics and seasonal variability [*Chanton et al.*, 2009]. Recent literature has emphasized the dependency of emissions of cover soil thickness and texture, as well as microbial oxidation rates which vary spatially and temporally with seasonal climatic trends [*Jones and Nedwell*, 1990; *Kightley et al.*, 1995; *Bogner et al.*, 1997; *Klusman and Dick*, 2000; *Scheutz et al.*, 2009]. For modeling purposes, the major controls are: (1) engineered gas recovery which lowers CH₄ concentrations at the base of the cover, in turn reducing the driving force for diffusive flux of CH₄ to the atmosphere [*Bogner et al.*, 1997; *Park and Shin*, 2001; *Zhang et al.*, 2008] and (2) major surface processes, which rely on the site-specific properties of the cover materials as well as seasonally variable CH₄ transport and methanotrophic oxidation [*Maurice and Lagerkvist*, 2003; *Zhang et al.*, 2008; *Scheutz et al.*, 2009]. The three major types of cover materials include thin daily covers over recently placed refuse; thicker intermediate covers overlying older refuse with high rates of methanogenesis; and final covers which are placed when a site reaches final grade. Oxidation rates are strongly coupled to engineered controls (cover design; landfill gas recovery); for example, engineered gas extraction can facilitate oxidation due to reduced rates of gross CH₄ flux to the base of cover soils. Observed CH₄ transport and oxidation rates are strongly linked to infiltration events and temperature changes at various temporal scales, both in natural ecosystems [*Morrissey and Livingston*, 1992; *Hargreaves and Fowler*, 1998] and landfill cover soils [*Maurice and Lagerkvist*, 2003; *Scheutz et al.*, 2009].

[7] The purpose of this project was to develop an improved site-specific landfill CH₄ inventory methodology for California by focusing on the fundamental processes which control emissions. The model addressed herein (CALMIM, California Landfill Methane Inventory Model) is an annual landfill CH₄ emissions inventory model developed for California landfill sites and field-validated for daily, intermediate, and final cover soils during 2007–2008. CALMIM models typical annual emissions based on 1-D diffusional flux and

seasonal oxidation in site-specific cover soils, focusing specifically on inputs and outputs which can be validated at field scale. A major driver for this study was a research review for California [Farrell *et al.*, 2005] which indicated that an improved landfill CH₄ inventory methodology was a high priority due to uncertainties associated with current methods. An important consideration for California was, according to data compiled by the California Department of Resources Recycling and Recovery, >90% of the waste in place in permitted California landfills is currently under active gas extraction, which constitutes a major control on emissions. Moreover, CALMIM is also compliant with current IPCC National Inventory Guidelines for CH₄ emissions from solid waste disposal sites [IPCC, 2006] as a “Tier III” model using “validated higher quality” methods [IPCC, 2006]. It is important to note that California has greenhouse gas reporting requirements which are separate and distinct from U.S. national greenhouse gas inventory reporting to the UNFCCC and other evolving U.S. requirements.

[8] CALMIM is designed for site-specific applications and is the first landfill inventory model which decouples emissions from gas generation modeling. Although the literature contains several complex, process-based models which rigorously address the seasonality of gaseous carbon and nitrogen fluxes in other managed and natural ecosystems (e.g., CENTURY [Parton, 1996]; CASTANEA [Davi *et al.*, 2006]; and LPJmL [Bondeau *et al.*, 2007]), similar seasonal models have not been developed for landfill settings [de Visscher and van Cleemput, 2003; Molins *et al.*, 2008; Scheutz *et al.*, 2009]. Therefore, consistent with recent literature emphasizing strong seasonal dependencies for CH₄ transport, oxidation, and emissions in other managed and pristine soil ecosystems [Cao *et al.*, 1995; Wille *et al.*, 2008], a major goal of this study was to develop a functional, field-validated annual CH₄ emissions model for California landfill sites. As California landfills must currently comply with a variety of existing Federal, state, and local regulations pertaining to operational practices and monitoring, a secondary consideration was to realistically limit default input data requirements to readily available information. CALMIM also contains “advanced” features which can be implemented when additional site-specific data are available.

2. Methods

2.1. Model Structure and Components

[9] Table 1 provides an overview of the model structure, components and default boundary conditions. CALMIM (<https://www.ars.usda.gov/services/software/download.htm?softwareid=300>) is a freely available JAVA program which integrates site-specific data (location and cover design) with climatic simulation and one-dimensional soil microclimate and gas diffusion models for daily, intermediate, and final cover areas inclusive of CH₄ oxidation over a typical annual cycle. Figure 1 gives an overview of model components and linked structure. CALMIM includes: (1) the effect of engineered gas extraction; (2) the variable physical effects of daily, intermediate, and final cover materials to retard emissions; and (3) seasonal moisture and temperature effects on both gaseous transport and methanotrophic CH₄ oxidation in cover soils. The major driving force for emissions is the CH₄ concentration gradient through user-selectable cover mate-

rials, which is, in turn, related to the presence of engineered gas extraction systems and the efficiency of CH₄ oxidation in any particular cover soil. Both transport and oxidation are rigorously linked to seasonal climatic and soil microclimate variability through modified versions of existing, globally validated models: Global TEMPSIM, Global RAINSIM, SOLARCALC, STM² [Spokas and Forcella, 2006, 2009]. Thus, CALMIM estimates annual CH₄ emissions while accounting for climate-induced variability on transport and microbial oxidation. Although more complex models exist for predicting the flow of landfill gas as a function of diffusion and advection [Findikakis and Leckie, 1979; Findikakis *et al.*, 1988; Lang *et al.*, 1989; Kindlein and Ahrens, 2006; Donovan *et al.*, 2010; Yu *et al.*, 2010], a number of the assumptions in these models are often violated in field settings (e.g., homogeneity of waste mass; uniform characteristics; static CH₄ generation rates). (Please consult the auxiliary material for a detailed discussion of diffusive versus advective processes in landfill cover soils.)¹ Moreover, required model input parameters are often unknown, highly variable or cannot be directly measured in field settings (e.g., gas flux to the base of soil cover). Thus, the theoretical complexity of existing models linked to various uncertainties relative to field settings hinders our ability to arrive at a robust tool that can be field-validated for prediction of surface CH₄ emissions. Therefore, we relied on a 1-D gaseous diffusion model, since this approach focuses directly on the factors that control surface emissions (e.g., cover soil characteristics, microbial CH₄ oxidation, climate, and CH₄ concentration gradient through the cover materials). Each of the model components shown in Figure 1 will be described in separate sections below. Many components have both default settings as well as settings which can be customized by the user based on field measurements or site management practices. Such site-specific practices including various cover materials and engineered gas recovery are extremely important for landfill settings which, compared to other CH₄-emitting settings such as wetlands or rice production systems, represent a highly managed endpoint [Bogner *et al.*, 2000].

2.1.1. Overview of Model Structure and Site-Specific Inputs

[10] Required CALMIM inputs include the site location (latitude and longitude), cover description (material type and layer thickness), and the corresponding CH₄ concentration gradient. The site information is collected from the user through data input screens (Figure S1). Each daily, intermediate, and final cover material, up to a total of 10 different covers, is modeled separately with the results summed for an estimate of annual total site emissions. The user can choose between typical California cover designs (see Table 1) or a customized sequence using the “cover designer” where any layered soil sequence can be entered. For a particular cover, the minimum thickness for any layer is 2.5 cm with a maximum total thickness of about 2.5 m, which is related to limits for typical PC memory resources. USDA standard soil texture classes, alternative daily cover (ADC) and other non-soil materials (e.g., composts, biosolids, tire chips, geomembranes) are also available with their corresponding transport properties taken from published literature (Table S1 in the

¹Auxiliary materials are available in the HTML. doi:10.1029/2011JG001741.

Table 1. Overview of CALMIM Input Parameters, Bundled Models, and Outputs

| Model Inputs | Description | Value/Units/Reference | |
|----------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Site | latitude | decimal degrees (+N, -S) | |
| | longitude | decimal degrees (-W, +E) | |
| | waste footprint | acres | |
| Cover characteristics | coverage | 0–100% of waste footprint | |
| | organic matter | low-high (0–5%) | |
| | vegetation presence | 0–100% cover (slider bar); modifies incoming solar radiation [Si = (1 – Veg%) * Si] | |
| | gas recovery system | 0–100% coverage (slider bar); reduces the lower methane concentration in default cover scenarios | |
| Cover type selection | | | |
| | Daily | temperature: upper temperature: lower CH ₄ : upper CH ₄ : lower oxygen: upper oxygen: lower CH ₄ oxidation rate (optimum) | air temperature simulation 25°C 2 ppmv 0.3% (v/v) 20% (v/v) 5% (v/v) 1 μg CH ₄ g _{soil} ⁻¹ d ⁻¹ |
| Intermediate | temperature: upper temperature: lower CH ₄ : upper CH ₄ : lower oxygen: upper oxygen: lower CH ₄ oxidation rate (optimum) | air temperature simulation 35°C 2 ppmv 45% (v/v) 20% (v/v) 1% (v/v) 200 μg CH ₄ g _{soil} ⁻¹ d ⁻¹ | |
| | Final | temperature: upper temperature: lower CH ₄ : upper CH ₄ : lower oxygen: upper oxygen: lower CH ₄ oxidation rate (optimum) | air temperature simulation 40°C 2 ppmv 55% (v/v) 20% (v/v) 0% (v/v) 400 μg CH ₄ g _{soil} ⁻¹ d ⁻¹ |
| Custom | user selectable boundary conditions | | |
| Layer characteristics | material | various materials (Table 2) | |
| | thickness | variable: 2.5 cm to 2.5 m (1 to 100") | |
| Bundled models | GlobalTempSIM | air temperature simulation <i>Spokas and Forcella</i> [2009] | |
| | GlobalRainSIM | precipitation simulation <i>Spokas and Forcella</i> [2009] | |
| | SolarCalc | solar radiation simulation <i>Spokas and Forcella</i> [2006] | |
| | STM ² | soil temperature and moisture model <i>Spokas and Forcella</i> [2009] | |
| | Gas diffusion | oxygen and methane diffusion <i>Campbell</i> [1985] | |
| Model outputs ^a | Daily surface CH ₄ emissions | with oxidation without oxidation | g CH ₄ m ⁻² d ⁻¹ g CH ₄ m ⁻² d ⁻¹ |
| | Soil nodes (2.5 cm layer in cover) | soil temperature | °C |
| | | soil moisture | volumetric (cm ³ cm ⁻³) |
| | | air-filled porosity | cm ³ cm ⁻³ |
| | | oxygen concentration | % O ₂ |
| | | CH ₄ concentration: with oxidation | % CH ₄ |
| | | CH ₄ concentration: without oxidation | % CH ₄ |
| | | CH ₄ oxidation rate | g CH ₄ m ⁻² d ⁻¹ |
| | | CH ₄ oxidation percentage | % |
| | | bulk density | g cm ⁻³ |
| | fraction of time oxidizing | 0 to 100% (0–1) | |
| | Simulated weather data | maximum air temperature | °C |
| | | minimum air temperature | °C |
| | | precipitation | mm |

^aModel outputs are written directly to Excel compatible files for each cover type.

auxiliary material). If the concentration gradient is not known, the model utilizes default settings based on the cover type selected (daily, intermediate, or final) (Table 1). The default settings are based on values taken from the literature; in general, higher base CH₄ concentrations reflecting mature methanogenesis characterize the intermediate and final cover soils.

[11] Engineered gas recovery systems consisting of either vertical wells or horizontal collectors are an important influence on emissions. CALMIM requires input on whether

engineered gas recovery underlies each particular cover type and the corresponding spatial extent of coverage, expressed as % of total area with engineered gas extraction. Using the default gas concentrations, the model scales the base CH₄ concentration using the following formula:

$$\text{CH}_4 \text{ Base} = (\text{CH}_{4_Default})(1 - 0.3 * \text{Coverage } \%), \quad (1)$$

where CH₄_Default is the default cover concentration (Table 1), and Coverage % is the aerial extent of the gas recovery system

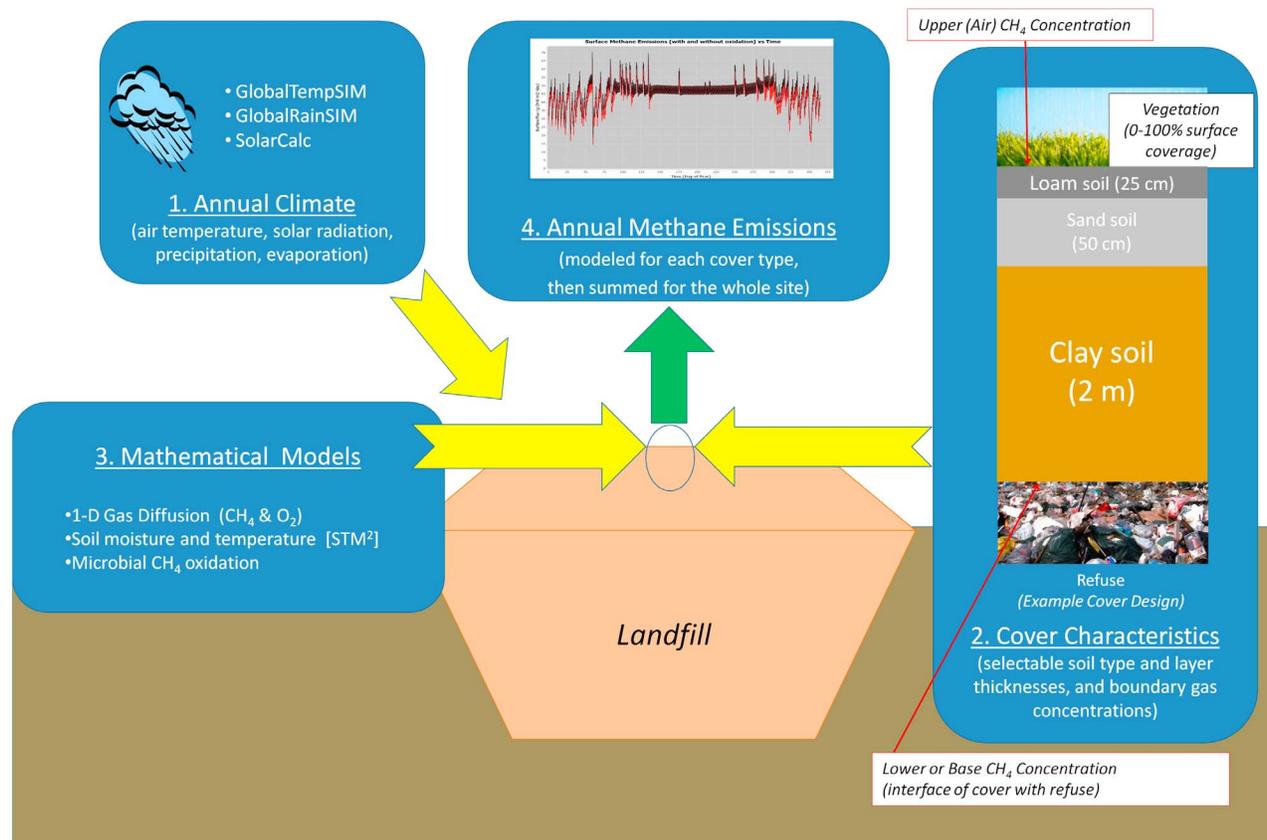


Figure 1. Graphical overview of the CALMIM model.

under the particular cover type (range of 0–1 representing 0–100%). If the user enters a custom gas concentration at the base of the cover, this linear correction is not performed, since the measurement would already include the correct concentration reduction attributed to the gas recovery system [Bogner *et al.*, 1997; Zhang *et al.*, 2008]. The estimation of a 30% reduction due to a gas recovery system covering 100% of the cover type is a conservative estimate, based on the field observations ranging from <1% to 35% v/v CH₄ at the base of final covers with a gas recovery system in place [Bogner *et al.*, 1997; Zhang *et al.*, 2008; Bogner *et al.*, 2011]. We strongly advocate the field measurement of this gradient as the driving force for emissions, using a statistically significant number of soil gas probes monitoring CH₄ concentration at the waste-soil interface for the various cover types; field values are entered in the custom boundary dialog of the model (Figure S1c in the auxiliary material).

2.1.2. Climate Simulation Models

[12] The existing models SolarCALC, GlobalTempSIM and GlobalRainSim [Spokas and Forcella, 2006, 2009] were incorporated into CALMIM to simulate a typical annual cycle of air temperature, precipitation, and incoming solar radiation referenced to site latitude and longitude. These models were previously validated for a number of global locations [Spokas and Forcella, 2006, 2009; Kahimba *et al.*, 2009] and rely on 30-yr (1961–1990) interpolated databases of Legates and Willmott [1990a, 1990b], Willmott and Matsuura [1995], and New *et al.* [1999]. Although the CALMIM model

was developed for application in California, these bundled simulation models confer global applicability at the 0.5×0.5 degree [latitude-longitude] scale. Average diurnal air temperature patterns are simulated in CALMIM using methods described by Cesaraccio *et al.* [2001] yielding air temperature values interpolated down to 10 min intervals for an annual cycle.

2.1.3. Soil Microclimate Model

[13] The soil microclimate simulation is linked to both site-specific soils (discussed in the next section) and a modified version of the existing soil temperature/moisture model, STM² [Spokas and Forcella, 2009]. The original STM² boundary conditions, developed for agricultural settings, were altered for CALMIM (Table 1) because landfills have a heat source (decomposing refuse) and saturated gas boundary conditions at the cover/refuse interface. CALMIM also permits the user to override these defaults through user-selectable boundary conditions (Table 1 and Figure S2c). In general, CALMIM incorporates default soil physical properties based on the soil texture and selected alternative cover materials permitted in California (Table S2 in the auxiliary material). It should be noted that the soil properties were derived from literature and databases for a variety of ecosystems [e.g., Clapp and Hornberger, 1978; Wösten and van Genuchten, 1988; Bouma, 1989] and not specifically for landfill soils. Compared to agricultural and other non-landfill soils, landfill covers are compacted to higher bulk densities [Spokas and Bogner, 2011], adding conservatism to the

transport modeling because the more highly compacted landfill soils would be expected to have lower effective diffusion coefficients and lower gaseous fluxes. The accuracy of these assumptions requires additional evaluation.

2.1.4. Diffusion/Oxidation Modeling and CALMIM Output

[14] Gas diffusion was assumed to obey Fick's law, which is widely used and observed to provide satisfactorily comparisons for gas transport in soils [Grable and Siemer, 1968; Šimůnek and Suarez, 1993; Moldrup et al., 1998, 2000, 2003]. From Fick's law,

$$J = D_s \frac{dC}{dz} \approx D_s \frac{\Delta C}{\Delta z}, \quad (2)$$

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 concentration, and z is depth. Moldrup et al. [1998] suggested a soil-type dependent gas diffusivity model (referred to as the Buckingham-Burdine-Campbell equation) for gas diffusivity:

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

where $D_{a,T}$ is the free-air diffusion coefficient at temperature T , ϕ is the total soil porosity ($\text{cm}^3 \text{cm}^{-3}$), θ_{air} is the air filled porosity ($\text{cm}^3 \text{cm}^{-3}$), and B is the Campbell B or the slope of the soil moisture retention curve in a $\log(\theta)$ - $\log(-\Psi)$ coordinate system [Campbell, 1985]. This model of the soil diffusivity was found to provide better prediction than other models across multiple soil types [Rolston and Moldrup, 2002; Moldrup et al., 2004]. Temperature also influences diffusion and can be accounted for by the relationship:

$$D_{a,T} = D_{a,20C} \left(\frac{T}{293K} \right)^{1.75}, \quad (4)$$

where $D_{a,T}$ is the free air diffusion coefficient at temperature T , $D_{a,20C}$ 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. For the flux calculation, Fick's law was solved at each time step using the Thomas algorithm [Campbell, 1985]. The mass balance at any node N is given by:

$$J_N - J_{N-1} - U_N = 0, \quad (5)$$

Where J_N is the gas flux at node N , J_{N-1} is the flux at node $N-1$, and U_N is the sink at node N (of oxygen or methane). For oxygen consumption, the assumptions by Campbell [1985] were used (surface consumption rate of $5 \times 10^{-4} \text{ g O}_2 \text{ m}^{-3} \text{ sec}^{-1}$ with an exponential decrease with depth). Therefore, oxygen diffuses in from the atmosphere and is attenuated by the average heterotrophic bacterial O_2 consumption in soils, prior to being available for CH_4 oxidation.

[15] For CH_4 oxidation, extensive supporting laboratory studies using daily, intermediate, and final cover soils from the two major field validation sites permitted the development of empirical relationships for node- and time-specific oxida-

tion rates [Spokas and Bogner, 2011]. These relationships scale the rate of CH_4 oxidation as a function of soil temperature and soil moisture potential for each node and time step. Optimal oxidation rates from the California soils ranged from 112 to 644 $\mu\text{g CH}_4 \text{ g}^{-1} \text{ d}^{-1}$, with an optimal temperature of 27.6°C and soil moisture potential of -33 kPa [Spokas and Bogner, 2011]. The impact of temperature on microbial oxidation is estimated as a Gaussian function and the impact of soil moisture as a sigmoid function [Spokas and Bogner, 2011]. The default optimum rates for CH_4 oxidation capacity as a function of cover type are given in Table 1. However, these values can be altered (Figure S2c) if site-specific data are available. These empirical models are a simplification of the complex microbial dynamics of the various populations of methanotrophic bacteria present in landfill cover soils [Scheutz et al., 2009]. However, similar empirical models are used to explain other biological responses to soil moisture and temperature by both microbial species [e.g., Stark and Firestone, 1995] and plant processes [e.g., Watt et al., 2010]. Due to the fact that CH_4 oxidation alters the concentration gradient and thereby the flux of CH_4 through the entire cover, the non-oxidized and oxidized scenarios are modeled independently to adequately account for the net difference in the surface emissions as a result of methanotrophic activity. This also allows quantification of the overall impact of CH_4 oxidation, as well as visualization of the temporal effects (daily or seasonal) in the standard output plots.

[16] Standard model output generated by CALMIM includes surface CH_4 emissions with and without CH_4 oxidation, site percent oxidation estimate, graphs of the profile (surface, middle and bottom nodes) of the nodal soil temperature, soil moisture, air-filled porosity, oxygen concentration, methane concentration (with and without oxidation), and corresponding CH_4 oxidation rate for the annual cycle, as well as the annual average depth profile of CH_4 oxidation. CALMIM also automatically generates EXCEL-compatible output files which archive the results of each simulation, including the calculated soil properties as a function of depth and time during model simulation (e.g., profiles for soil temperature/moisture, air-filled porosity, O_2 concentration, CH_4 oxidation rate, and CH_4 surface flux and soil gas concentrations with and without CH_4 oxidation (Figure S2d)).

2.2. Sensitivity Analysis

[17] In order to isolate the response behavior of individual input variables, model sensitivity analysis was conducted by incrementally varying single input parameters (cover properties, thickness, extent of gas recovery) and examining impact on resulting emission and oxidation rates. Of course, this analysis does not validate the model, but confirms the reasonable operation of the model over a wide range of inputs, as well as the overall sensitivity of outputs to variable input parameters.

2.3. Field Validation

[18] Field validation was conducted over two years at two California sites, including the coastal Marina Landfill (36.71°N, 121.762°W, Monterey County) and the Scholl Canyon Landfill (34.158°N, 118.196°W, Los Angeles County). The field validation and model development were independent efforts and collected data were not utilized for

parameterization of the numeric model. Because both sites had full gas recovery systems and engineered cover soils as primary controls on emissions, we conducted four field campaigns at each site focusing on the historically wettest and driest months in order to capture the seasonal wet (March 2007, 2008) and dry (August 2007, 2008) extremes. Methane emissions were quantified using multiple randomized deployments of 9 stainless steel static chambers across the three major cover types (daily, intermediate, and final). Because static chambers can quantify the spatial variability of both positive fluxes and negative fluxes (uptake of atmospheric CH₄) across a given cover type, this is the method of choice for small-scale process-related studies. Moreover, because we were developing an annual inventory model, non-soil fluxes associated with cracks, fissures, and piping leakages were not considered, since California and U.S. regulations require quarterly monitoring of surface CH₄ concentrations followed by remediation and re-monitoring as part of normal operations and maintenance (i.e., South Coast Air Quality Management District Rule 1150.1; see <http://www.aqmd.gov/rules/reg/reg11/r1150-1.pdf>). The properties of the cover materials and soil methods are described in detail by *Spokas and Bogner* [2011] and *Bogner et al.* [2011]. Weather stations (Onset Computing) and depth arrays of soil temperature/moisture sensors were installed at each site to continuously monitor wind speed, air temperature, relative humidity, and soil temperature and moisture profiles (Onset Computing). (Names are necessary to report factually on available data; however, the USDA neither guarantees nor warrants the standard of the product, and the use of the name by USDA implies no approval of the product to the exclusion of others that may also be suitable.) Sampling and analysis techniques for chamber samples, soil gas probes, and source gas (composite landfill gas) are discussed in detail by *Bogner et al.* [2011]. Gas samples were analyzed at the USDA-ARS laboratories in St. Paul and Morris, MN. Soil moisture (TDR) and temperature (RTD) were also measured at each of the >800 chamber locations. The minimum detectable CH₄ flux was $\pm 12 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$.

[19] In addition, field measurements of CH₄ emissions from intermediate cover materials at three additional California Landfills (Kirby Canyon; 37.185°N 121.671°W, Lancaster; 34.747°N 118.116°W; and Tri-Cities; 37.51°N 121.99°W) [*Green et al.*, 2009] were compared to CALMIM results. All of these sites are large, active municipal solid waste landfills [$>1200 \text{ t d}^{-1}$] with operational landfill gas collection systems underlying these cover soils. The Lancaster site is located in an arid, high desert region (Mojave Desert), while the Tri-Cities and Kirby Canyon sites are characterized by a Mediterranean climate. Field measurements included both static chambers and an aboveground technique using a TDL (tunable diode laser) instrument for vertical and horizontal radial plume mapping [see *Green et al.*, 2009].

[20] For all five field validation sites, stable carbon isotopes for CH₄ for selected chamber and probe samples were analyzed at Florida State University. Fractional CH₄ oxidation (as % oxidation) was calculated using published methods [*Liptay et al.*, 1998; *Chanton and Liptay*, 2000; *Chanton et al.*, 2008] based on a comparison of the $\delta^{13}\text{C}$ for anoxic zone CH₄ compared to the emitted CH₄ (chambers or probes).

2.4. Statistical Model Validation

[21] Although Pearson correlation coefficients (R^2) were calculated as a routine measure of correspondence for climatic and soil microclimate outputs, significant R^2 values do not automatically correlate to model accuracy [*Willmott*, 1982]. Therefore, for air and soil temperature comparisons an “index of agreement” or modeling index (d) was calculated with the following expression:

$$d = 1 - \left[\frac{\sum_{i=1}^n (x_i - y_i)^2}{\sum_{i=1}^n (|x_i - \bar{x}_i| + |y_i - \bar{y}_i|)^2} \right], \quad (6)$$

where x_i are the field measured values with a mean of \bar{x}_i and y_i are the modeled values and corresponding \bar{y}_i [*Willmott* 1981; *Mayer and Butler*, 1993]. The value of d will range between 0 and 1, with a value of 1 indicating perfect model agreement [*Willmott*, 1981].

[22] Two other statistical measures [root mean square error (RMSE) and mean absolute error (MAE)] were also calculated, since the units are the same for the parameter as the observed quantity and therefore allow a more meaningful comparison. These statistical measures have been used in other modeling comparisons [e.g., *Wegehenkel*, 2000; *Winslow et al.*, 2001; *Spokas and Forcella*, 2006] and are recommended measures in assessing model performance [*Willmott*, 1982].

[23] Surface CH₄ flux and oxidation results were analyzed by comparing the mean and associated standard deviation of the measurement compared to the modeled annual surface CH₄ emission and associated estimated CH₄ oxidation.

3. Results and Discussion

3.1. Sensitivity Analysis

3.1.1. Effect of Variable Soil Texture

[24] A 30 cm soil cover with a base CH₄ concentration of 10% (v/v) was assumed to have different soil textures and was analyzed under the same climatic conditions (Marina Landfill, Monterey County). Figure 2a indicates the variability in the CH₄ emission rate with and without oxidation along with the total estimated annual CH₄ oxidized. Diffusive flux is reduced by finer soil texture (Figure 2a). For this scenario, the variability in the prediction ranged from 46 to 163 g CH₄ m⁻² d⁻¹ without oxidation and 18 to 122 g CH₄ m⁻² d⁻¹ with oxidation, as a function of soil texture. Typically, coarser soil textures resulted in higher predicted surface emissions both with and without oxidation. On the other hand, the estimated annual amount of CH₄ oxidized as a function of soil texture ranged from 21 to 41 g CH₄ m⁻² d⁻¹. Coarser textured soils resulted in higher predicted oxidation capacities, while finer-textured soils have a lower total CH₄ oxidation capacity, which is in agreement with the literature [*Scheutz et al.*, 2009].

[25] The percent CH₄ oxidation (Figure 2b) is a function of the non-oxidized diffusive flux and is the parameter commonly quantified by current isotopic methods for positive CH₄ fluxes [*Liptay et al.*, 1998; *Chanton and Liptay*, 2000; *Chanton et al.*, 2008]. Unfortunately, these methods cannot be applied to negative fluxes (uptake of atmospheric

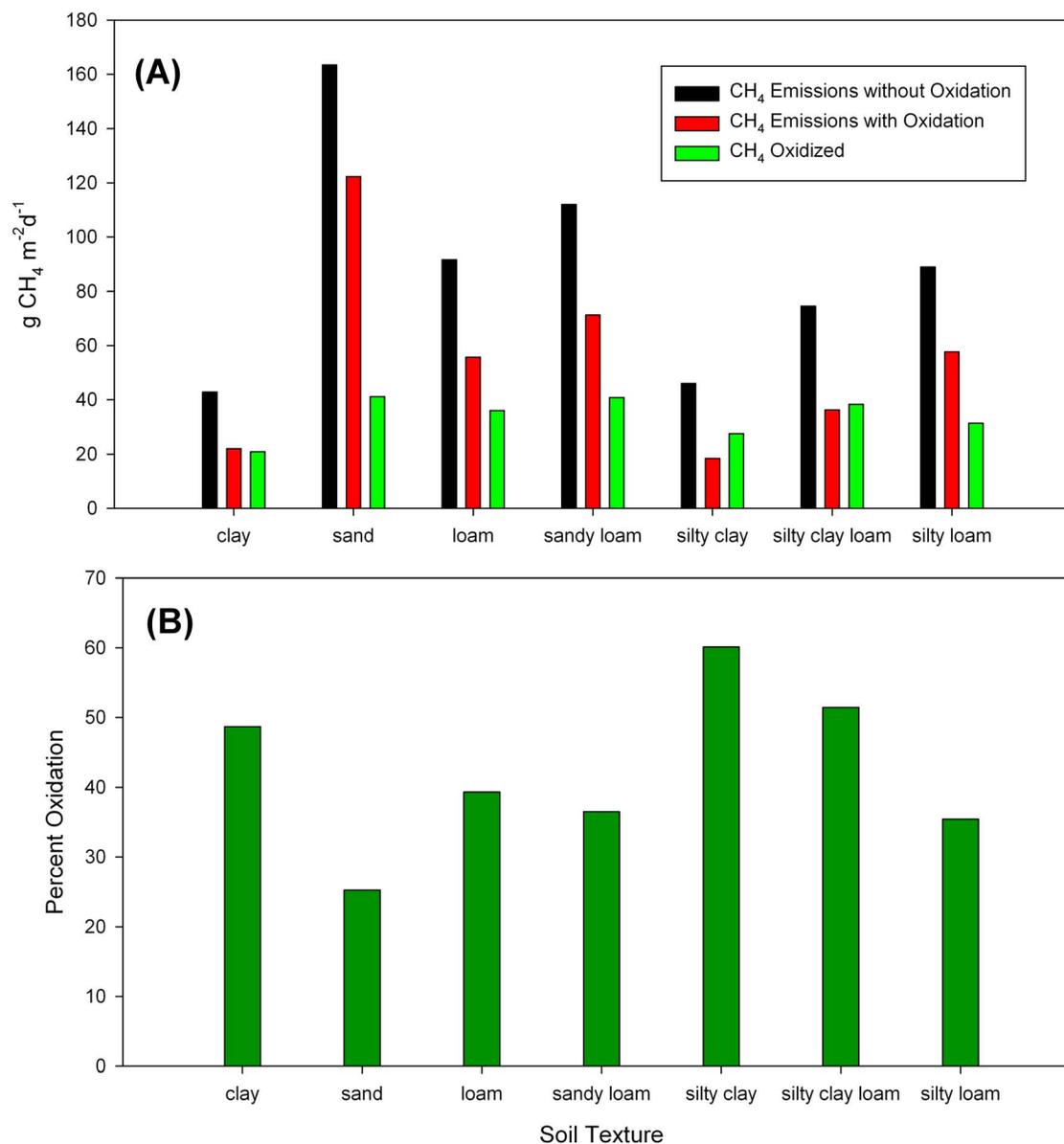


Figure 2. Impacts of soil texture on (a) net CH₄ surface emissions with oxidation, net surface emissions without oxidation, and mass of CH₄ oxidized as well as (b) the comparison of the percent oxidation of various soil textures under identical boundary conditions for a 30 cm thick soil layer.

CH₄) and, because of the observed variability in field results, may be difficult to apply where positive CH₄ fluxes are low. Importantly, percent oxidation is only a relative measure of the CH₄ that is oxidized in a particular landfill cover soil and is not a direct quantitative assessment of the CH₄ oxidation rate. Because percent CH₄ oxidation is a function of the non-oxidized diffusive flux, it is therefore highly variable across soil textures and climates. This oxidation percentage varied from 25 to 60% across soil textures in the soil texture analysis (Figure 2b) within the same climatic region, with coarser-textured soils having higher predicted oxidation capacities. However, finer-textured soils typically have higher percent oxidation due to the reduced magnitude of CH₄ flux as a function of the soil texture (Figure 2a). Because of these relationships, the depth- and climate-dependent oxidation

rate (g CH₄ m⁻² d⁻¹) would be the preferred measure of oxidation capacity in a particular cover soil cover for a particular climate rather than the percent oxidation. Moreover, both published field data [Bogner *et al.*, 2007; Borjesson and Svensson, 1997; Scheutz *et al.*, 2009; Zhang *et al.*, 2008] and CALMIM model output indicate that the percent CH₄ oxidation at a particular site can range from 0 to 100%, with high temporal variability. In CALMIM, this variability is directly attributable to the coupling of soil cover properties and climatic driving forces to estimate soil microclimate as a function of depth. The microclimate data are then utilized to estimate the rate of microbial CH₄ oxidation based on the empirical relationship with temperature and soil moisture [Spokas and Bogner, 2011]. However, even though the in situ oxidation rate (g CH₄ m⁻² d⁻¹) would be the preferred

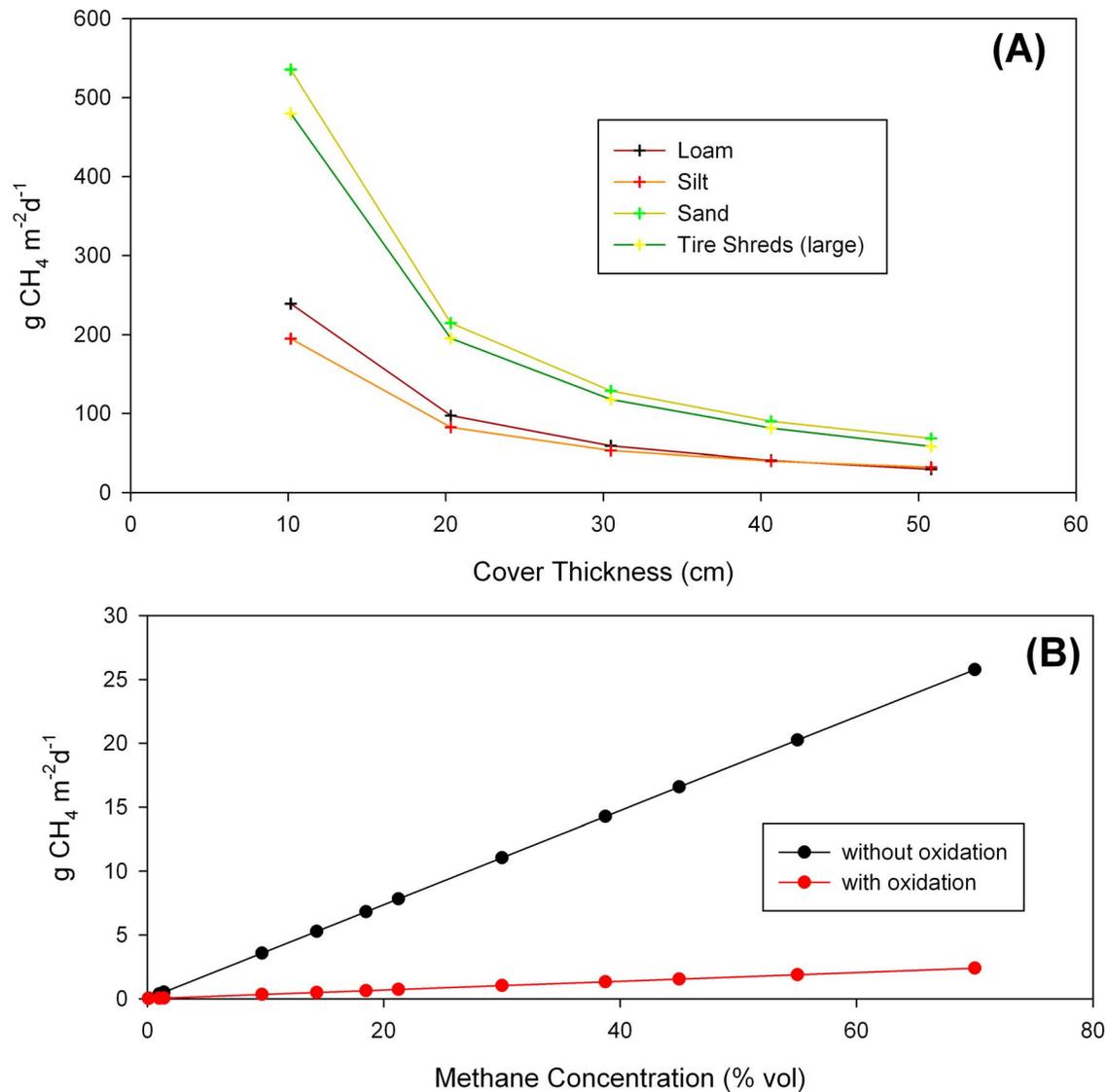


Figure 3. Impacts of (a) various soil texture on the resulting emissions (with oxidation) of various soil cover materials as a function of layer thickness and (b) the relationship between base CH_4 concentration and the corresponding surface emissions with and without oxidation for a 100 cm clay soil cover.

measure for oxidation within a particular soil cover in a particular climate, it is not currently possible to quantify this oxidation rate in the field; instead, one must rely on numerical modeling coupled to laboratory studies [Bogner *et al.*, 2000].

3.1.2. Effect of Cover Soil Thickness and CH_4 Concentration Gradient

[26] The thickness of a variety of cover materials (Figure 3a) and the concentration gradient (Figure 3b) across a uniform 100 cm clay cover were independently varied. There is a nonlinear response to the changing thickness of the cover soil (Figure 3a). On the other hand, alterations in the concentration gradient result in a linear relationship with surface flux (Figure 3b), which is consistent with the assumption of diffusive flux (equation (2)). As discussed above, the CH_4 oxidation percentage is determined relative to the net flux of CH_4 into the base of the cover material. This

can be seen in Figure 3b, where the 100 cm clay cover was capable of oxidizing virtually all of the gross diffusive CH_4 flux to the base of the cover material. As discussed above and in the auxiliary material, the model does not account for advection in its current form.

3.1.3. Effect of Gas Recovery System

[27] The sensitivity of the model to the presence of an engineered gas recovery system was examined for a 30-cm clay cover. This dependency was scaled by altering the base concentration according to the relationship given in equation (1) (Figure 4a) and the assumption for diffusive transport results in a direct linear relationship between surface flux and the concentration gradient (Figure 4b). However, the estimated CH_4 oxidation potential in the cover is equivalent for each scenario, because this is dependent on the soil texture (e.g., O_2 diffusion profile), soil moisture, and temperature (which were held equal for all scenarios) (Figure 4b).

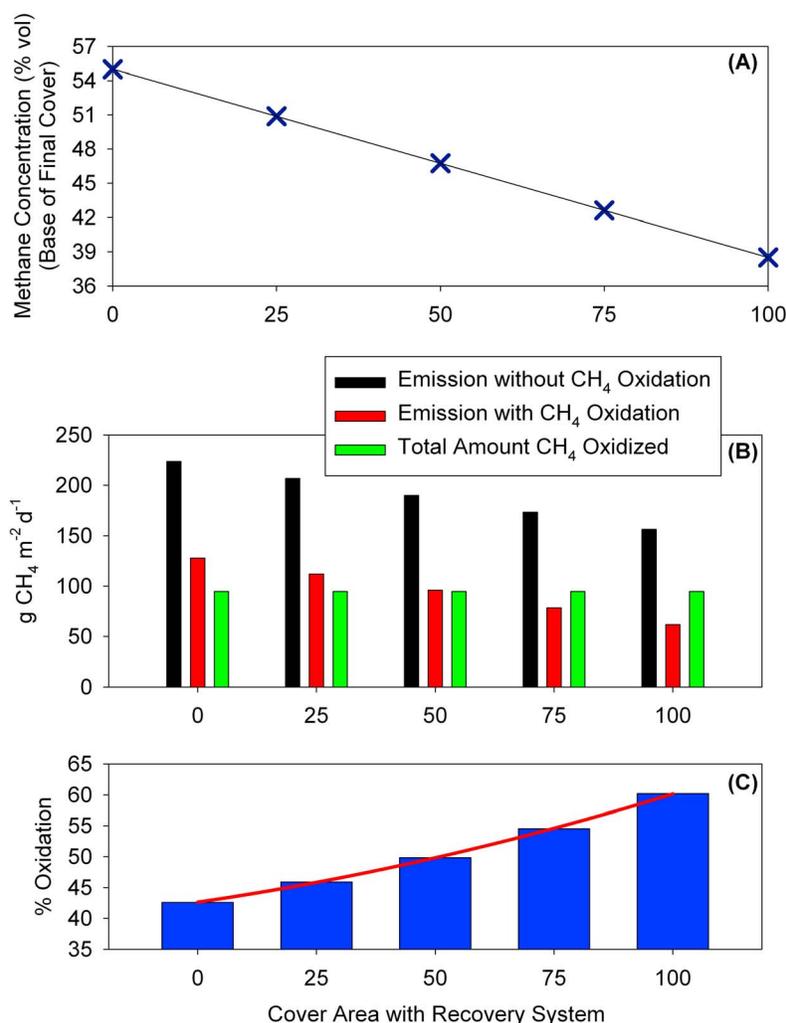


Figure 4. Impacts of an engineered gas recovery system for a 30 cm clay cover, using the default concentration profile for a final cover on (a) base CH₄ concentration as a function of aerial coverage of the recovery system, (b) comparisons of predicted emissions with and without oxidation and the total CH₄ oxidation predicted in the cover material, and (c) comparison of the estimated % CH₄ oxidation as a function of the recovery system configuration.

Therefore, the percent oxidation is not related to the amount of CH₄ oxidized (same in all scenarios), but is a function of the non-oxidized flux (Figure 4c). Importantly, this same pattern has been observed in other measurement campaigns [Chanton *et al.*, 2011a, 2011b]. The presence of a recovery system (with 100% coverage) for this particular scenario reduced emissions by over 50% (128 to 62 g CH₄ m⁻² d⁻¹), even though the base concentration was only reduced by 30%, due to the increased impact of oxidation on the reduced (net) CH₄ flux to the atmosphere at the top of the cover soil. These modeling results agree with other studies indicating that the optimal mechanism to reduce surface CH₄ emissions is to reduce the CH₄ loading into the base of the cover soil [Park and Shin, 2001; Zhang *et al.*, 2008; Chanton *et al.*, 2011a, 2011b].

3.1.4. Effect of Climate

[28] Table 2 presents the data from the comparisons of three different landfill cover scenarios:

- [29] 1. Daily cover (30 cm daily cover of sand),
 - [30] 2. Intermediate cover (30 cm sandy loam), and a
 - [31] 3. Final cover (0.8 m final cover: [30 cm sandy clay loam (bottom), 25 cm clay, and 25 cm loam (surface)]).
- [32] These comparisons assumed the default CH₄ boundary conditions for the cover type (Table 1) and were analyzed at various global locations. As can be seen in Table 2, there is considerable variability in the prediction of surface CH₄ emissions as a function of the global climate and cover type. Typically, higher emissions were predicted in colder climates, where soil microclimate conditions for CH₄ oxidation are not optimal year-round (Figure S2). For the daily cover, the variability ranged from 4.3 to 5.8 g CH₄ m⁻² d⁻¹ across the various climates. However, larger differences were observed for the intermediate and final cover types across these climates. In particular, one can see the range in the percent oxidation from 3.5 to 12% for the intermediate covers and 32 to 100% for the final cover as a function of climate (Table 2).

Table 2. Model Results for Various Global Locations for the Three Simulated Default Cover Designs

| Lat/Lon | Average Annual Temp (°C) | Average Annual Precipitation (mm) | Daily | | Intermediate | | Final | | % CH ₄ Oxidation | | |
|------------------------|--------------------------|-----------------------------------|------------------------------------------------------------------------|---------------------------------------------------------------------|------------------------------------------------------------------------|---------------------------------------------------------------------|------------------------------------------------------------------------|---------------------------------------------------------------------|-----------------------------|--------------------|---------------------|
| | | | Without Oxidation (g CH ₄ m ⁻² d ⁻¹) | With Oxidation (g CH ₄ m ⁻² d ⁻¹) | Without Oxidation (g CH ₄ m ⁻² d ⁻¹) | With Oxidation (g CH ₄ m ⁻² d ⁻¹) | Without Oxidation (g CH ₄ m ⁻² d ⁻¹) | With Oxidation (g CH ₄ m ⁻² d ⁻¹) | Daily (% Oxidation) | Int. (% Oxidation) | Final (% Oxidation) |
| London, UK | 9.6 | 607.3 | 5.0 | 4.8 | 512.5 | 452.8 | 202.1 | 18.1 | 4.5 | 11.7 | 90.8 |
| Rio De Janeiro, Brazil | 21.5 | 123.3 | 4.6 | 4.1 | 470.9 | 386.0 | 161.3 | – | 9.4 | 18.1 | 100 |
| Vancouver, Canada | 10.3 | 986.6 | 4.7 | 4.7 | 507.5 | 446.2 | 187.3 | 10.1 | 4.7 | 12.1 | 94.0 |
| Stockholm, Sweden | 5.0 | 406.2 | 5.2 | 5.0 | 527.3 | 479.3 | 215.7 | 59.3 | 3.4 | 9.3 | 73.4 |
| Cairo, Egypt | 21.1 | 24.3 | 4.7 | 4.3 | 490.1 | 409.0 | 204.9 | 1.7 | 7.5 | 16.6 | 99.2 |
| Lima, Peru | 21.5 | 123.3 | 4.7 | 4.3 | 490.4 | 404.6 | 213.7 | 0.01 | 8.0 | 17.5 | 99.9 |
| Sydney, Australia | 17.6 | 869.8 | 4.7 | 4.4 | 493.4 | 415.1 | 201.1 | 0.1 | 7.3 | 15.9 | 99.7 |
| Mexico City, Mexico | 13.7 | 737.7 | 4.8 | 4.6 | 506.5 | 434.9 | 210.8 | 7.1 | 5.9 | 14.2 | 97.0 |
| Beijing, China | 9.9 | 457.0 | 5.0 | 4.8 | 518.4 | 459.7 | 223.0 | 55.6 | 5.2 | 11.7 | 77.1 |
| Juneau, Alaska U.S. | 6.4 | 1766.2 | 4.9 | 4.7 | 507.1 | 456.8 | 155.8 | 27.1 | 3.7 | 10.0 | 83.6 |
| Moscow, Russia | 3.65 | 425.8 | 5.2 | 5.1 | 532.0 | 415.9 | 219.1 | 72.9 | 3.4 | 8.9 | 68.7 |
| Barrow, Alaska | -12.3 | 70.05 | 5.8 | 5.8 | 585.4 | 565.9 | 254.8 | 174.7 | 1.2 | 3.5 | 32.2 |

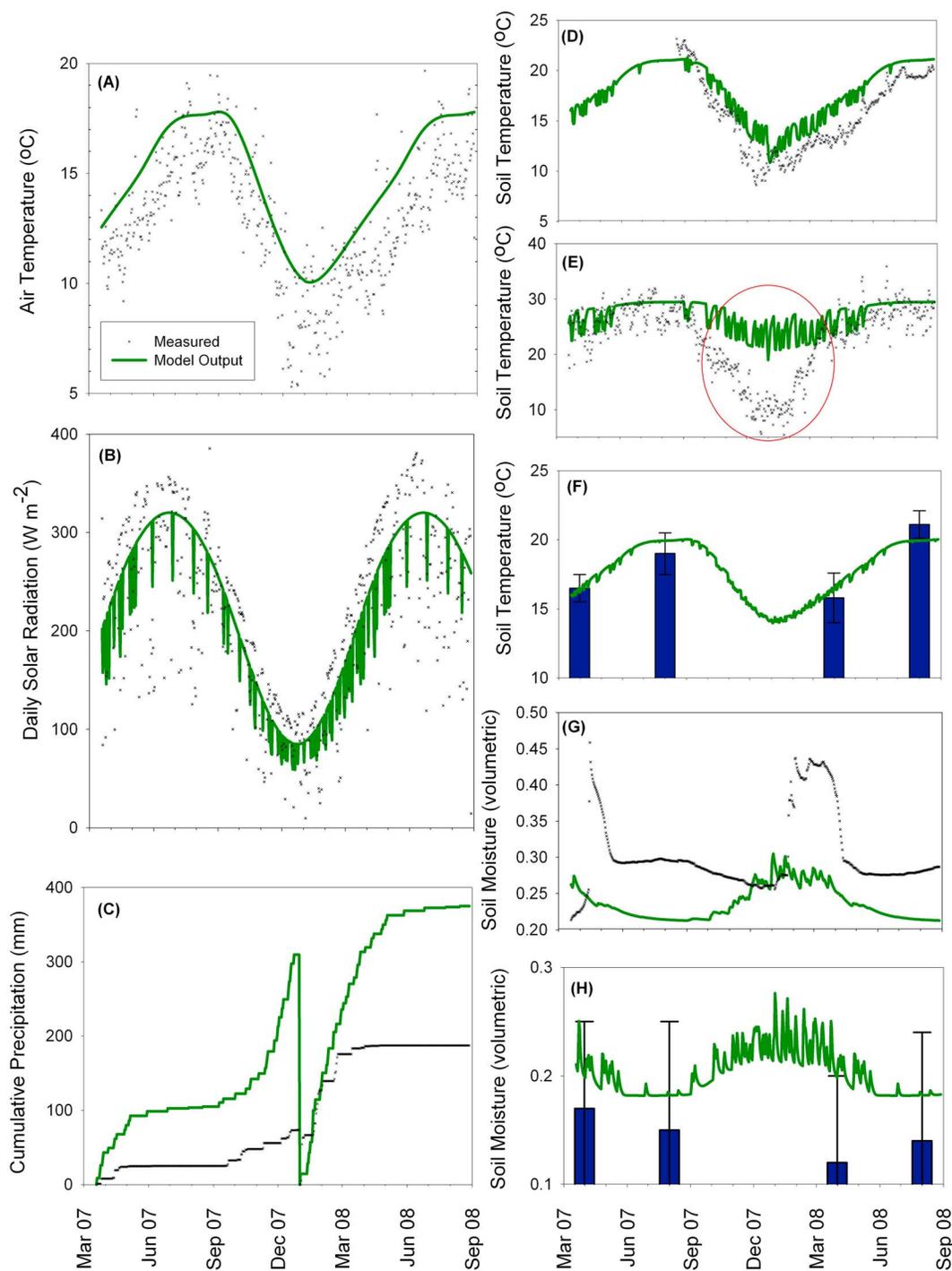


Figure 5. Model validation at the Marina landfill site, including comparisons of (a) air temperature, (b) solar radiation, (c) precipitation, (d) 10 cm final cover soil temperature, (e) 15 cm intermediate cover soil temperature, (f) 5 cm daily cover soil temperature, (g) 10 cm volumetric moisture in the final cover, and (h) 15 cm volumetric moisture in the intermediate cover.

These higher estimates for the percent oxidation have been supported by recent field measurements [e.g., *Chanton et al.*, 2009], but depend on the cover soil type and particular climate. As seen in these simulations, the attenuating role of CH_4 oxidation increases with greater cover thicknesses

and warmer climates. There was strong seasonal variability observed for the global sites (Figure S2), with equatorial sites possessing reduced annual variability compared to the northern colder locations.

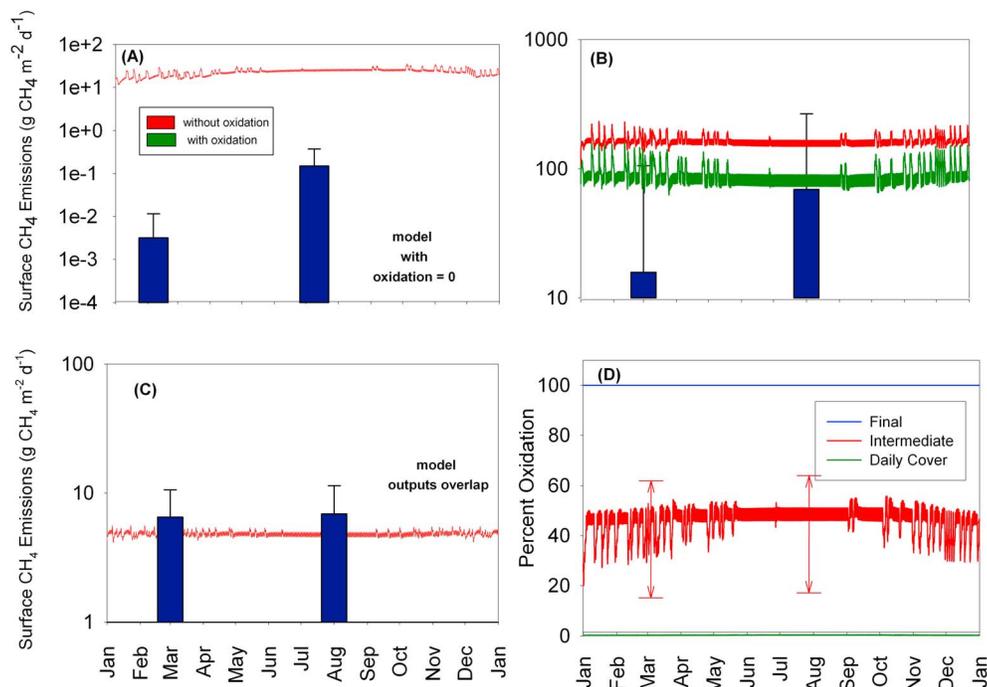


Figure 6. Model validation at the Marina landfill site for the predicted (a) surface methane emissions of the final cover, (b) surface methane emissions in the intermediate cover, (c) daily cover surface methane emissions, and (d) estimated percent oxidation of the three cover types. The average and standard deviation of the associated field data are overlaid on these plots and the range of percent oxidation from the field assessments are shown by the arrow.

3.2. Field Validations

3.2.1. Marina Landfill

[33] Figure 5 compares model results and field data for the northern California coastal site (Marina) using model parameters in Table S2 in the auxiliary material. Average air temperature predictions (Figure 5a) matched the overall trend ($R^2 = 0.694$; d-index = 0.831), with a slight positive bias (RMSE = 2.45°C; MAE = +2.10°C). The solar radiation predictions (Figure 5b) were correlated ($R^2 = 0.572$; d-index = 0.869) and had small relative errors (<10%) in the magnitude of the daily incoming radiation estimate (RMSE = 60.4 W m⁻²; MAE = +46.1 W m⁻²). Precipitation predictions were somewhat overestimated due to the ongoing drought in California during 2007–2008 (Figure 5c). However, the Mediterranean pattern, where a majority of the annual precipitation falls in the cooler part of the year (November–March), was accurately simulated, despite relative differences in predicted quantities. Not surprisingly, results for the climate simulations were comparable to other published validations for these models [Spokas and Forcella, 2006, 2009; Kahimba et al., 2009].

[34] The predicted and measured soil temperature at 10 cm in the final cover and 15 cm depth in the intermediate cover area are shown in Figures 5d and 5e, respectively. These shallow depths were chosen based on the observations that maximum rates of soil CH₄ oxidation are typically found in the upper portion of the soil profile (e.g., 5–25 cm) where optimum microclimate conditions exist for methanotrophic activity as a function of O₂ availability, soil temperature,

moisture, and CH₄ supply [e.g., Scheutz et al., 2009]. For the final cover, the model demonstrated good prediction of the overall cover soil temperature trend ($R^2 = 0.919$; d-index = 0.814) and a RMSE of 2.4°C and a MAE of 2.1°C. These errors are virtually identical to the air temperature prediction errors and similar in magnitude to errors observed in other modeling studies [Granberg et al., 1999; Cannavo et al., 2006; Bittelli et al., 2008]. This is vital, due to the importance of soil temperature on microbial reactions [Riveros-Iregui et al., 2007; Or et al., 2007]. It should be noted that this correspondence to field data was achieved using modeled meteorological data and not site-specific weather data which could, of course, improve model comparisons. Due to a localized decrease in the lower boundary temperature (soil-refuse interface) which was not reproduced in the modeling (steady state condition), the measured intermediate cover soil temperature did not match the modeled temperature as well during the winter (Figure 5e). Overall, the intermediate cover comparisons at Marina were relatively poor ($R^2 = 0.462$; d-index = 0.595; RMSE = 6.7°C and a MAE of 4.9°C). Figure 5f illustrates the modeled temperature profile for the daily cover at 5 cm. Due to operational constraints, it was not possible to monitor the daily cover on a continuous basis. Point measurements from the field monitoring (average and standard deviation; Figure 5f) were compared to model results. Overall, the model did follow the same trend as the individual measurements, and generally the predicted daily temperature was within the standard deviation of the field measurements.

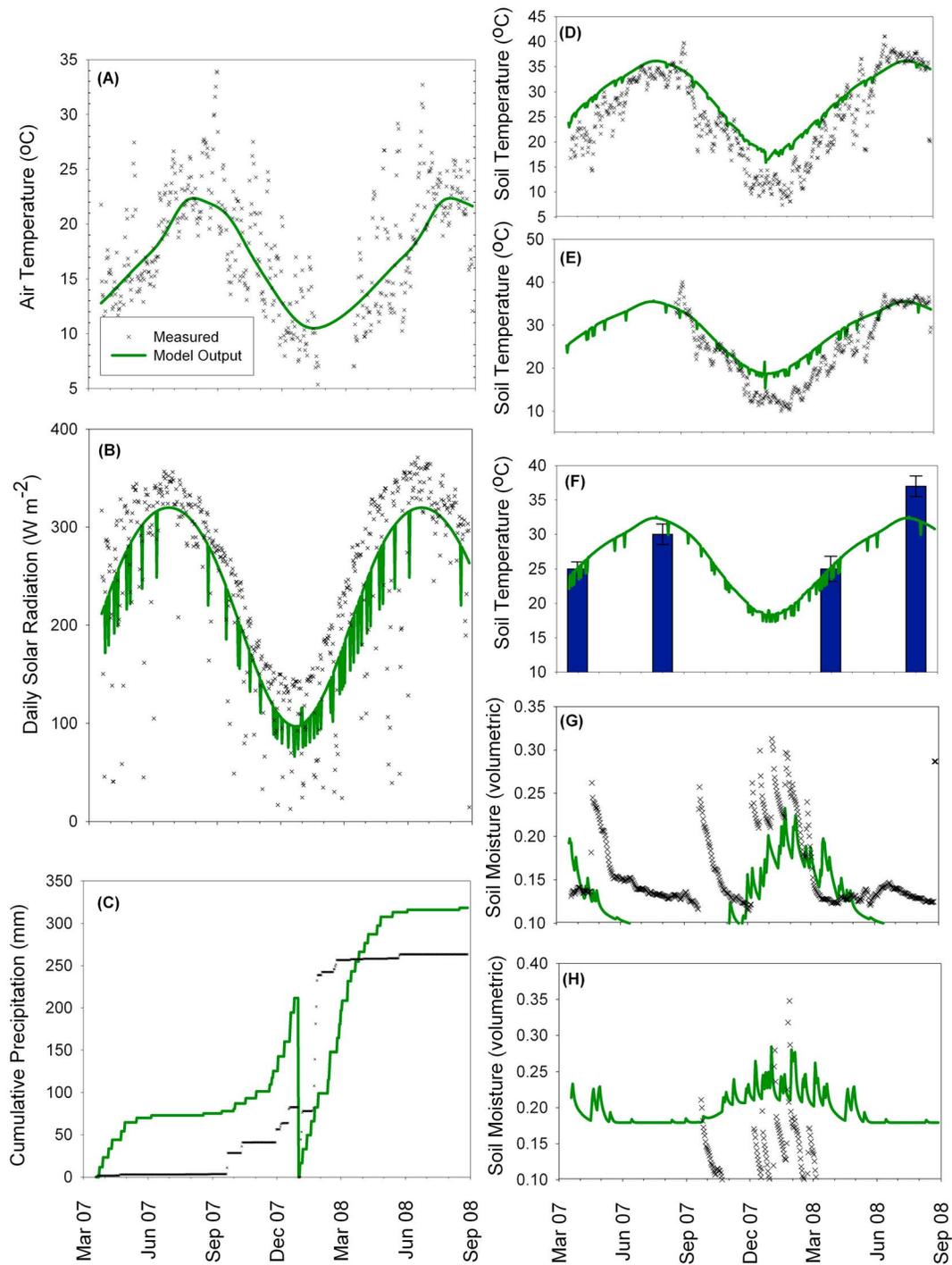


Figure 7. Model validation at the Scholl Canyon landfill site, including comparisons of predicted (a) air temperature, (b) solar radiation, (c) precipitation, (d) 10 cm final cover soil temperature, (e) 15 cm intermediate cover soil temperature, (f) 5 cm daily cover soil temperature, (g) 10 cm volumetric moisture in the final cover, and (h) 15 cm volumetric moisture in the intermediate cover.

[35] Volumetric soil moisture predictions for the final (10 cm) and intermediate (15 cm) are shown in Figures 5g and 5h, respectively. Only the final cover at Marina was instrumented with soil moisture sensors. However, soil moisture was not further statistically compared, since the model used simulated annual weather data rather than site-specific data.

Nevertheless, predicted soil moisture profiles for the final cover matched the seasonal trends observed in the field data (Figure 5g). Also, the dry season range of volumetric moisture contents measured in the field overlapped the modeled output (Figure 5h), suggesting a good match for measured-to-modeled soil physical parameters (Table S1) at this site.

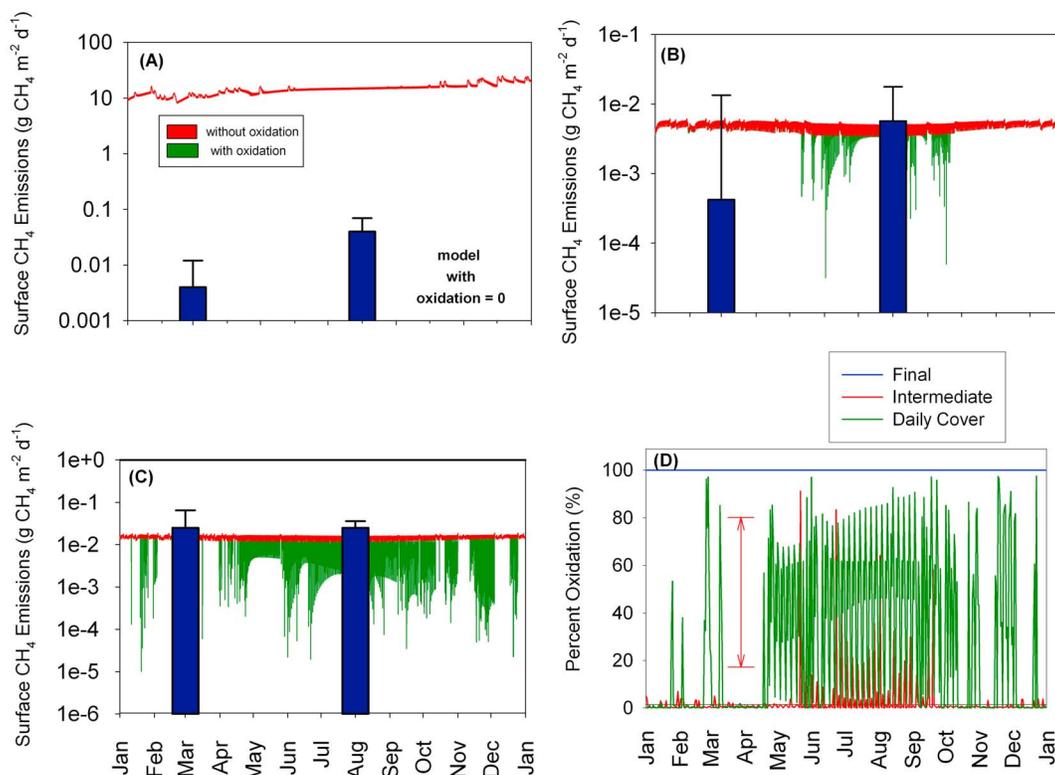


Figure 8. Model validation at the Scholl Canyon landfill site for the predicted (a) surface methane emissions of the final cover, (b) surface methane emissions in the intermediate cover, (c) daily cover surface methane emissions, and (d) estimated percent oxidation of the three cover types. The average and standard deviation of the associated field data are overlaid on these plots and the range of percent oxidation from the field assessments is shown by the arrow.

[36] Model outputs for predicted surface CH_4 emissions at Marina during an annual cycle with and without oxidation are shown for the final (Figure 6a), intermediate (Figure 6b) and the daily cover areas (Figure 6c), with the corresponding field measurement averages and standard deviations. As seen in Figure 6, the model results were typically within the same order of magnitude as the field measurements but slightly higher, indicating that the model results were conservative for annual inventory purposes. Daily cover area comprises a small fraction of the typical landfill footprint (<4 ha). The final cover had very low measured fluxes ($<0.1 \text{ g CH}_4 \text{ m}^{-2} \text{ d}^{-1}$). The corresponding modeled percent oxidation is also shown for the final, intermediate, and daily cover types in Figure 6d. The estimated range of percent CH_4 oxidation from the isotopic measurements was 1 to 84% with an average of 30 to 40% for all cover types depending on whether the estimation was made from chamber flux measurements or soil gas probes, which is similar to range reported by *Chanton et al.* [2009]. In general, the model predicted 100% CH_4 oxidation over the full annual cycle for the final cover soil, 50% for the intermediate cover soil, and less than 1% oxidation occurring in the daily cover. These oxidation percentages are solely estimates from the ratio of the modeled fluxes with and without oxidation. Therefore, the error associated with each prediction is difficult to ascertain.

[37] In large part, the numeric differences between percent oxidation between cover types (Figure 6d) are related to the

significantly lower oxidation potential for the daily cover compared to the intermediate and final covers, because the daily cover had not previously been exposed to elevated CH_4 concentrations [*Spokas and Bogner, 2011, and references therein*]. These differences are accounted for in the model by scaling the rate of CH_4 oxidation in the model as a function of cover type selected (Table 1).

3.2.2. Scholl Canyon Landfill

[38] Figure 7 (using model input parameters in Table 2) compares model results and field data for the Scholl Canyon site (Los Angeles County). The air temperature predictions (Figure 7a) matched the overall trend ($R^2 = 0.722$; d-index = 0.521), with a slightly higher positive bias (RMSE = 3.9; MAE = $+3.0^\circ\text{C}$) than at Marina. Solar radiation predictions (Figure 7b) were well-correlated to field data ($R^2 = 0.779$; d-index = 0.608) with small relative errors (<10%) relative to the magnitude of the daily average prediction (RMSE = 57.9 W m^{-2} ; MAE = $+42.3 \text{ W m}^{-2}$). Mediterranean precipitation patterns (Figure 7c) were simulated but with an overestimation due to the extreme drought conditions for 2007–2008 in southern California.

[39] The predicted and measured soil temperatures at 20 cm in the final cover and at 15 cm in the intermediate cover are shown in Figures 7d and 7e, respectively. As discussed above, CH_4 oxidation activity would be optimized at these shallow depths. CALMIM predicted the final soil cover temperature trend ($R^2 = 0.920$; d-index = 0.846) with a

Table 3. CALMIM Model Comparisons for Intermediate Cover Areas at Other California Landfill Sites

| Site Name | Location | Field Measurements [Green <i>et al.</i> , 2009] (g CH ₄ m ⁻² d ⁻¹) | | Model Annual Prediction (With Oxidation) (g CH ₄ m ⁻² d ⁻¹) |
|--------------|---------------------|------------------------------------------------------------------------------------------------------------|-------------------|-----------------------------------------------------------------------------------------------------|
| | | Flux Chamber | VRPM ^a | |
| Lancaster | 34.747°N, 118.116°W | -0.21 to 0.47 Mean: +0.02 Median: 0 | 1 to 5 | 0.47 |
| Kirby Canyon | 37.185°N, 121.671°W | -0.04 to 0.05 Mean: -3.36 Median: 0 | 8 to 11 | 0.14 |
| Tri-cities | 37.510°N, 121.99°W | -0.02 to 9.2 Mean: +6.82 Median: +0.03 | 23 to 42 | 3.9 |

^aVRPM, vertical radial plume mapping [see Green *et al.*, 2009].

RMSE of 5.4°C and a MAE of 4.4°C. The intermediate cover was modeled adequately with a $R^2 = 0.944$; d-index = 0.892; RMSE = 4.7°C and a MAE of 3.8°C over the field monitoring period. Figure 7f compares modeled to actual soil temperatures at 5 cm for the daily cover; the superimposed bars indicate the point measurements (average and standard deviation). Intermediate and final cover soils at Scholl Canyon were highly compacted (estimated 2 g cm⁻³ for intermediate and final covers [Spokas and Bogner, 2011]). Therefore, we suggest that the differences between modeled and measured emissions at the Scholl Canyon site are an artifact of the assumption of the lower bulk density values in the CALMIM modeling (Table S1).

[40] Volumetric soil moisture predictions for the final (10 cm) and intermediate (15 cm) are shown in Figures 7g and 7h, respectively. As seen in the data, soil moisture responds directly to precipitation events. Moreover, the predicted soil moisture profiles for the final and intermediate covers closely match seasonal trends seen in the field data (Figures 7g and 7h). As mentioned above for the soil temperature comparisons, field deviations from the assumed soil physical properties for the highly compacted Scholl soils could lead to observed errors in the soil moisture predictions for the dry soil conditions. This difference is more dramatic for the Scholl Canyon site compared to Marina because the Marina cover soils had lower soil bulk densities closer to the assumed model parameters (Table S1). In addition, the extreme drought conditions also could explain the overestimation observed in the modeled soil moisture results (Figures 7g and 7h).

[41] Modeled surface CH₄ emissions with and without oxidation were compared to field measurements for the final (Figure 8a), intermediate (Figure 8b) and the daily cover (Figure 8c) with modeled % oxidation (Figure 8d) for all three covers. From the isotopic field measurements, the estimated range of CH₄ oxidation was 10–100% with an average of 48–52% depending on whether the estimation was made from chamber flux measurements or soil gas probes. The model predicted 100% oxidation for the final cover but <1% oxidation for the daily cover, with rapid responses to infiltration events (Figure 8c), thus capturing the response of oxidation to moisture and indicating that very low soil moisture resulted in reduced oxidation rates. The overall response of emissions and oxidation to soil moisture events is very significant for Scholl Canyon due to the lower CH₄ fluxes with CH₄ oxidation an important contributing mechanism to mitigating

emissions. Furthermore, as seen in Figure 8d, the percent oxidation of the daily cover can exceed the intermediate cover, since this percentage is both a function of the gas diffusion rates, soil microclimate conditions, and the soil texture differences (Figure 2b).

3.2.3. Other California Landfill Sites

[42] Comparisons at other California sites were limited to the intermediate cover areas at three sites (Lancaster, Kirby Canyon, Tri-Cities) and are summarized in Table 3 using model input parameters given in Table S2. In general, intermediate cover areas are the most important cover type at active landfill sites with respect to emissions because these generally comprise the largest percentage of the total surface area during the active filling phase. Intermediate covers are thinner than final soil covers, are placed when a cell is completed, and buried when new cells overlie older phases. Intermediate cover areas can remain exposed for extended periods of time (>3 years) but are characterized by well-established methanogenesis in the underlying waste which can result in higher surface emissions. Overall, there was good agreement between the flux measurements and the modeling results, with CALMIM outputs for the three sites exhibiting relatively low surface emission estimates. In general, the vertical radial plume mapping (VRPM) [Thoma *et al.*, 2010; Green *et al.*, 2009] results were consistently higher than the chamber and corresponding CALMIM results. These differences are attributable to uncertainties regarding the area contributing to flux using VRPM methods along with other complicating issues (e.g., model assumptions versus actual climatic stability, terrain, and interfering CH₄ sources from adjacent cells) [Babilotte *et al.*, 2010]. Furthermore, the VRPM method (as do all aboveground methods) captures secondary emissions from cracks, fissures, and piping system leakages. As discussed above, by regulatory mandate, these are detected and remediated on a quarterly basis as part of normal operations and maintenance and thus are not modeled for annual inventory purposes by CALMIM.

4. Conclusions

[43] CALMIM is an IPCC Tier III methodology for landfill CH₄ emissions relying on “validated higher quality” methods. Importantly, this project has developed a field-validated modeling methodology based directly on the physical and biochemical processes that control emissions during typical

annual climatic and soil microclimate variability for site-specific daily, intermediate, and final cover soils. As published literature has demonstrated, the “net” landfill CH₄ emissions to the atmosphere are dependent on the presence of engineered gas recovery, the site-specific cover materials, their seasonal moisture and temperature profiles, and the variability of seasonal methanotrophic CH₄ oxidation in various cover materials. A major focus of CALMIM as an annual inventory model is on the effect of larger-scale climatic processes and their influence on soil microclimate [Entin et al., 2000; Muttiah and Wurbs, 2002] as an important control on landfill CH₄ emissions in California. The accuracy of the global climate models embedded in CALMIM is adequate to establish typical or average annual conditions [Spokas and Forcella, 2009]. In general, as discussed above, CALMIM predicts field CH₄ emissions within the same order of magnitude and provides a framework for an improved methodology for predicting annual landfill CH₄ emissions. Comparisons of CALMIM modeling output to field measurements of emissions and oxidation at additional landfill sites outside of California has been initiated, including both U.S. and international sites.

[44] The current model represents an initial step with respect to the decoupling of landfill surface emission predictions from gas generation modeling. Some anticipated future improvements include facilitating the routine use of site-specific climate and soil microclimate data, potential inclusion of advective gas transport, as well as developing a default soils database specifically for gaseous transport in landfill cover soils with high compaction. However, for inventory purposes, the use of the current soils database within CALMIM, based on agricultural soils, adds conservatism to the modeling output, since estimated transport rates would typically be higher for agricultural soils with lower compaction. Importantly, the CALMIM results also illustrate the limitations of a historical dependence on the percent CH₄ oxidation as a measure of the total potential oxidation capacity of various landfill soil cover systems [e.g., Czepiel et al., 1996]. Rather, a more comprehensive accounting for the actual CH₄ oxidation rate is preferred, which is dependent on the magnitude of the non-oxidized flux and is a function of soil texture, climate, CH₄ and O₂ concentration gradients, and diffusive flux rates.

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