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From California dreaming to California data: Challenging historic models for landfill CH_4 emissions

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Abstract

Improved quantification of diverse CH_4 sources at the urban scale is needed to guide local GHG mitigation strategies in the Anthropocene. Herein, we focus on landfill CH_4 emissions in California, challenging the current IPCC methodology which focuses on a climate dependency for landfill CH_4 generation (methanogenesis), but does not explicitly consider climate or soil dependencies for emissions. Relying on a comprehensive California landfill database, a field-validated process-based model for landfill CH_4 emissions (CALMIM), and select field measurements at 10 California sites with a variety of methods, we support the contrary position: Limited climate dependency for methanogenesis, but strong climate dependency for landfill CH_4 emissions. Contrary to the historic IPCC empirical model for methanogenesis with kinetic constants related to climate, we demonstrate a simpler and more robust linear empirical relationship ($r^2 = 0.85$; n=128) between waste mass and landfill biogas recovery [126×10^{-6} Nm^3 CH_4 hr^{-1} Mg_{waste}^{-1}]. More interestingly, there are no statistically significant relationships with climate, site age, or status (open/closed) for landfill biogas recovery. The current IPCC methodology does not consider soil or climate drivers for gaseous transport or seasonal methanotrophy in different cover soils. On the other hand, we illustrate strong climate and soil dependencies for landfill emissions—e.g., average intermediate cover emissions below 20 g CH_4 m^{-2} d^{-1} when the site's mean annual precipitation is >500 mm y^{-1}. Thereby, for the California landfill CH_4 inventory, the highest-emitting sites shift from landfills containing the largest mass of waste to sites dominated by intermediate cover types having a reduced rate of soil CH_4 oxidation during the annual cycle. These differences have profound implications for developing more realistic, science-based urban and regional scale GHG inventories for landfill CH_4 while reducing uncertainties for this important anthropogenic source.

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Introduction

Methane (CH_4) is the 2nd most important greenhouse gas (GHG), accounting for about 20% of positive radiative forcing (Myhre et al., 2013). However, considering indirect effects associated with increased atmospheric ozone and water vapor, CH_4 is responsible for about 40% of positive forcing. Even though annual emissions of 500-600 Tg are well-constrained by atmospheric measurements, their allocation to various natural (e.g., wetlands, termites, caribou) and anthropogenic sources (e.g., oil/gas production & transport, domesticated ruminants, rice production, coalbed leakages, wastewater, landfills) remains highly uncertain. Landfills are currently considered to be the 3rd largest source of atmospheric CH_4 in California (Deshpande, 2014) as well as the US, estimated at 18% of the total US methane emissions by source (USEPA, 2014, USEPA, 2015). However, recent field measurements for the city of Indianapolis, for example, have demonstrated that landfills may account for >30% [33 ± 10%] of the total urban CH_4 source (Cambaliza et al., 2015). Regional- and

urban-scale CH_4 inventories guide local mitigation strategies; thereby, we need the best estimates for individual sources including landfill CH_4.

In the U.S., the first "sanitary" landfills during the 1950's and 1960's were operated under minimal regulatory guidance (some states and municipalities) with minimal engineering (e.g. soil cover on top of buried waste to reduce animal vectors, blowing waste and nuisance odors). Beginning in the 1970's and accelerating in the 1980's -1990's under the U.S. EPA Subtitle D landfill regulations and Clean Air Act amendments, municipal solid waste landfills are now highly engineering and monitored facilities. Currently, routine practices include control of inputs, compaction of waste, "cell" construction with engineered synthetic liner systems and collection/management of landfill leachate [liquids], onsite or offsite leachate treatment, engineered structures for collection and management of runoff to minimize infiltration and leachate generation, internal and external monitoring of leachate and biogas, engineered cover systems, and engineered systems for collection and utilization of biogas. Some typical landfill cover types and thicknesses are shown in the Supplemental Information (Table S1- Cover Types). Individual landfill sites can have multiple daily, intermediate, and final covers at a particular site. This greatly complicates both the monitoring and modeling of emissions because of varying source strengths, wind directions, topography, and CH_4 concentration gradients affecting diffusive flux through each individual cover type. The cover soils provide a major barrier to gaseous emissions, while concurrently promoting internal anaerobic conditions in the buried waste for methanogenesis. In addition, the interaction of seasonal climate with the different cover soils, resulting in soil moisture and temperature changes with depth through an annual cycle. These alterations can result in large temporal variations in both soil gas transport and microbial methane oxidation rates.

The biodegradable fractions of landfilled waste (paper, food, garden waste) decompose anaerobically via a complex collection of microbial reactions mediated by hydrolytic, fermentative, acidogenic, acetogenic, and methanogenic microorganisms. The first observations of methane production from organic matter decomposition were made by the Italian physicist Alessandro Volta in 1776, after reading of the presence of a "flammable gas" from the research of Benjamin Franklin in the US (Theresa, 2012). Ever since these initial observations, the major assumption has been that waste decomposition and biogas formation is related to the amount of degrading organic material. The early biogas generation models were empirical in nature and possessed a variety of mathematical forms (i.e., single component/multiple component kinetic models; lag time/no lag time). However, all of these models shared the common goal to predict future biogas generation and potential recovery rates from past landfill performance. The initial biogas model development in the US occurred in California about 4 decades ago, following the first project to commercially recover landfill gas during the U.S. "energy crisis" of the 1970's at the Palos Verdes Landfill. Model validation consisted of a comparison between predicted and actual annual biogas recovery over a period of a few years, to derive the empirical constants to optimize the model fit. For some examples of the first applications of these equaions see EMCON (1980) and Halvadakis et al. (1983), which correlated landfill biogas production to the total landfilled waste.

During the 1980's, the use of these predictive models for biogas projects diminished. It was recognized that a multiplicity of operational and engineering factors (e.g. waste type, compaction, moisture aviability) control both the quantity and quality of recoverable landfill biogas (Klink and Ham, 1982). One could not know, a priori, whether a particular model was accurate and predicted biogas recovery; moreover, utilization hardware purchased solely on the basis of empirical modeling had resulted in expensive mistakes. Installation of biogas control and collection systems is becoming routine as part of more optimized landfill design and management practices. For commercial biogas projects, a preferred strategy often consisted of installing gas collection infrastructure, evaluating gas quantity & quality, and committing to gas utilization hardware based on site-specific economics for a preferred utilization option. Although a few sites have historically upgraded the gas to pipeline quality during periods of high natural gas prices, the majority of the >600 current U.S. projects focus on electrical generation for sale to the local grid or direct gas use in industrial/commercial boilers [see http://www.epa.lmop.gov]. At individual sites, the gas recovery infrastructure is expanded in a timely manner concurrent with landfill expansions, often including both horizontal collectors and vertical wells.

In the late 1980's and early 1990's, there was a revival of interest in 1st order models to estimate biogas *generation* as the starting point for *emission* estimates for three major applications:

1. Clean Air Act Regulations [http://www.epa.gov/ttn/atw/landfill/landflpg.html] addressing emissions of total non-methane organic compounds (NMOCs) with inclusion determined on the basis of landfill size, modeled biogas generation, subtraction of any recovered gas, the assumption that the remainder is emitted, and the application of a default or measured mixing ratio for total NMOCs in the emitted gas. The U.S. EPA developed the LANDGEM Model [**LAND**fill **G**as **E**missions **M**odel] for this regulatory initiative from the Scholl Canyon model (EMCON, 1980), one of the original models formulated for an early Los Angeles area biogas recovery project, which is still active today. This site-specific model was thus expected to reasonably model biogas generation at all U.S. sites. In practice, when applied to individual sites, prescribed regulatory default values are applied for L_o (biogas yield per unit waste, m gas/m waste) and k (kinetic constant, 1/t). The L_o values are assumed to vary with regional waste

characterization and the *k* values with climate. Also, an assumed "recovery efficiency" factor (typically 75%) is added to account for the difference between measured gas recovery and modeled "theoretical" gas generation--this factor has rarely been determined in field settings addressing *all* CH_4 pathways (recovery, emissions, oxidation, lateral migration, and internal storage). [See discussion in Spokas et al. (2006)].

2. National-scale greenhouse gas (GHG) inventory reporting based on the National GHG Inventory Program landfill methodology of the Intergovernmental Panel on Climate Change (IPCC, 1996, 2006). Originally, either empirical ("mass balance") or multi-component 1st order kinetic models (termed "first order decay" - FOD) models were allowed (IPCC, 1996) when national-scale GHG inventories under the United Nations Framework Convention on Climate Change were first completed for the 1990 base year. For these first inventories, these models were typically applied to the entire landfilled waste mass for a country. In time, they were increasingly applied to specific sites with emissions summed for a national estimate. In the latest guidelines (IPCC, 2006), a multi-component FOD model based on the biodegradable organic carbon in various waste fractions is recommended for all countries with default values for kinetic constants (*k*) based on climate. The IPCC FOD methodologies are also used for regional GHG accounting including the current California GHG inventory under the jurisdiction of the California Air Resources Board (CARB) [See http://www.arb.ca.gov/cc/inventory/inventory.html].
3. Approved methodologies for Kyoto Protocol compliance [including the Clean Development Mechanism (CDM)] and voluntary carbon markets. The project design document for landfill gas recovery CDM projects typically references IPCC (2006) for baselines estimates using the approved consolidated methodology ACM-0001. There are also "avoided waste to landfill" methodologies where credits are given to projects which avoid landfilling of waste via alternative strategies such as composting (i.e. AM-0025). These allow the monetization of certified emission reductions (CERs) solely on the basis of modeled landfill emissions [See http://cdm.unfccc.int].

Historically, the first field studies to quantity landfill CH_4 emissions [for example Boeckx et al. (1997)] were being conducted at the same time that the first IPCC (1996) national GHG inventory guidelines (see Smith and Bogner, 1997). Similar to the early landfill biogas projects, "field validation" for the IPCC emissions consisted of comparing modeled biogas generation to limited measured biogas recovery data, primarily for 9 full-scale Dutch landfills (Oonk, 2010; Van Zanten and Scheepers, 1995).

Are these empirical models accurate? Realistically, one might argue that landfills fall somewhere between engineered digesters and anaerobic ecosystems in more open environmental settings (e.g., wetlands) (Bogner et al., 2000). In general, when applied to specific sites, these models can yield very large underestimates or overestimates for predicted vs. actual gas recovery, their original application (Thompson et al., 2009). For example, we note that landfill biogas CDM projects have consistently underperformed relative to baseline predictions (Couth et al., 2011), while well-operated California sites can recover 2-3 times the "predicted" biogas generation (Spokas et al., 2011). Thus, even for gas recovery predictions, the models have had difficulties in accurately predicting rates (Thompson et al., 2009). With regard to emissions, both the LANDGEM and IPCC FOD models were developed prior to a critical mass of field data on actual emission rates and mechanisms, and neither model was field-validated for emissions (Scheutz et al., 2009). During the last decade, field measurements have consistently indicated that unlike gas generation, landfill CH_4 emissions are not related to the biogas generation rate, but on: (1) the physical properties of site-specific cover materials to retard gaseous emissions; (2) presence of a biogas recovery system; and (3) methanotrophic CH_4 oxidation in site-specific cover soils related to seasonal soil microclimate conditions (Spokas and Bogner, 2011).

Seasonality and soil microclimate differences impact the CH_4 budget for wetlands (Morin et al., 2014) and other ecosystems (Cicerone et al., 1983; Sass et al., 1990). Not surprisingly, the same dependency exists for landfills; seasonal oxidation can vary from negligible to more than 100% (uptake of atmospheric CH_4) (Bogner et al., 1997; Sadasivam and Reddy, 2014; Scheutz et al., 2003; Yang et al., 2014). However, in addition to the current IPCC (2006) methodology not being independently field-validated for emissions (as discussed above), this methodology only allows a constant 10% annual oxidation at well managed sites, based on the first study to model annual oxidation at a small landfill in New Hampshire, USA (Czepiel et al., 1996a,1996b). Published literature has confirmed that CH_4 emissions from landfill cover soils, similar to other soil sources of atmospheric CH_4, have high spatial and temporal variability due to soil texture and microclimate-dependencies for gaseous transport and methanotrophic oxidation (Albanna et al., 2007; Bogner et al., 1997; Chanton and Liptay, 2000; Chiemchaisri et al., 2011; Czepiel et al., 1996a, 1996b; Goldsmith Jr. et al., 2012; Harborth et al., 2013; Lee et al., 2009; Pawłowska et al., 2003; Pratt et al., 2013; Rachor et al., 2013). Moreover, unique to landfill soils, emissions are also dependent on site-specific engineering and management factors, including the cover thickness & texture; areal extent of daily, intermediate, and final cover soils; and the direct effect of biogas extraction systems on soil gas CH_4 concentration gradients which control diffusive flux (Abichou et al., 2006a; Bogner et al., 2011; Perdikea et al., 2008). With small-scale rates (static chambers) ranging over 6-7 orders of magnitude for individual cover materials (<0.001 to >1000 g

CH_4 m^{-2} d^{-1}) (Li et al., 2004; Park and Shin, 2001) and large-scale "whole landfill" rates (e.g., aircraft-based mass balance techniques) significantly higher, but still ranging over 2-3 orders of magnitude (<160 to >1600 g CH_4 s^{-1}) (Peischl et al., 2013), it is clear that a significant challenge remains to quantify and model site-specific CH_4 emissions. Moreover, one must also consider the uncertainties associated with diverse field techniques (e.g., diffusion accumulation chambers, tracer techniques, micrometeorological techniques, aircraft mass balance) (Lai et al., 2012; Levy et al., 2011; Mann and Lenschow, 1994). Finally, since each field campaign represents a snapshot in time, a robust modeling framework is needed to integrate diel and seasonal rates over a typical annual cycle for each cover design at a specific site.

Herein, we challenge the adequacy of current inventory empirical models for landfill CH_4 emissions. Unlike the theoretical models which address the seasonality of GHG fluxes in other managed and natural ecosystems (Bond-Lamberty et al., 2007; Davi et al., 2006; Li et al., 2004; Parton, 1996), the current landfill methodology does not consider major climate and soil-microclimate drivers for CH_4 emissions from landfill cover soils with variable thickness, soil textures, and seasonal- and climate-dependent oxidation rates. All of these factors critically influence CH_4 emission rates through landfill cover soils (Park and Shin, 2001; Scheutz et al., 2009). California Landfill Methane Emissions Model (CALMIM) is an evolving site-specific, field-validated, process-based model originally developed for California in 2007–2010 (Bogner et al., 2011; Spokas et al., 2011; Spokas and Bogner, 2011) (CALMIM available at http://www.ars.usda.gov/services/software/download.htm?softwareid=300). Through a finite-difference solution to soil gas diffusion transport, CALMIM theoretically predicts a typical annual cycle for landfill CH_4 emissions based on the average site-specific climate and user inputted cover soils (Spokas et al., 2011). Using this predicted soil microclimate, soil CH_4 oxidation is estimated by empirical models correlated to soil moisture and temperature characteristics (Spokas and Bogner, 2011).

It is also important to examine the current status of "top down" emissions estimates inclusive of landfill CH_4 and other waste sector emissions in addition to the "bottom up" models. The most recent global estimates are included in the EDGAR-HTAP dataset, which is a harmonized 0.1° x 0.1° gridded air pollution database (Janssens-Maenhout et al., 2012). For landfill CH_4, EDGAR-HTAP uses country-level inventory data using IPCC (2006) for the developed countries. For developing and middle income countries not required to report annually, in addition to issues associated with IPCC (2006) as discussed above, there can be large disparities between the quality and quantity of temporally-varying national waste data, the basis for inventory calculations using IPCC (2006). For EDGAR-HTAP, the country-level data are dispersed on a 0.1° X 0.1° global grid according to population density. Thus, these data have the added convenience of 0.1° X 0.1° gridding but, as these estimates are based on IPCC (2006), they do not consider any of the major drivers for landfill CH_4 emissions now known from literature and field measurements as discussed above. Moreover, for both developed and developing countries, landfill sites are becoming increasingly dissociated from dense urban population centers as older landfills are filled and closed with new remote sites developed outside of urban corridors (El Baba et al., 2014).

Making use of a new large California landfill dataset (Walker, 2012), the field-validated process-based model (CALMIM), and existing data for measured California landfill emissions from existing studies (Bogner et al., 2011; Goldsmith et al., 2012; Jeong et al., 2013; Peischl et al., 2013; Spokas et al., 2011; see Table S4), we focus on:

1. The first data-based analysis refuting the current hypothetical linkage of biogas generation (k_o) to climate,
2. Application of the theoretical diffusion based CALMIM model to a new statewide inventory estimate for California. Results are compared to the current 2010 inventory, including the distribution and characterization of the highest-emitting Californian landfill sites, and
3. Systematic examination of the climatic dependencies of the new and old GHG inventory values.

2.0 Materials and methods

2.1 California datasets

In late 2012, a comprehensive dataset for permitted California landfills was developed by the California Dept. of Resources Recovery and Recycling [CalRecycle] (Figure S1). The complete electronic database (Walker et al., 2012) is provided in the supplemental files (Spreadsheet S2: CA-LANDFILLDATABASE.xlsx). From this collection, we initially used the 2010 data for 129 California sites with LFG recovery data to examine relationships between normalized landfill recovery [Nm^3 LFG h^{-1} Mg_{WIP}^{-1}], landfill age, size, operating status (open or closed), and local climate. These 129 sites with engineered biogas recovery represent 89.3% of the total 2010 WIP in permitted California landfills (Table S3). Both the *waste in place* (WIP) and biogas recovery data were independently reported by individual site operators to CalRecycle. General climate data [mean annual temperature (MAT) and precipitation] were derived from existing interpolated resources (Lawrimore et al., 2011; Legates and Willmott, 1990; Peterson and Vose, 1997).

We used the 2010 California GHG Inventory (Deshpande et al., 2014) as a reference point for current estimated landfill CH_4 emissions using the IPCC (2006) methodology. Previous publications provided

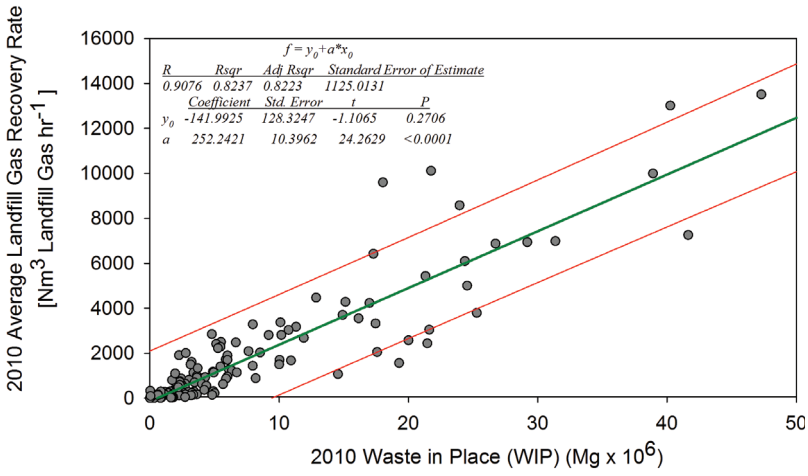

Figure 1

Relationship between the 2010 California waste in place (WIP) (x 10^6 Mg) and the annual landfill biogas recovery (252 × 10^{-6} Nm^3 LFG $hr^{-1}Mg^{-1}$ or 126 Nm^3 CH_4 $hr^{-1}Mg^{-1}$) from the California database (Walker et al., 2012).

This figure illustrates the comparison between the total waste in-place at 128 California landfills to annual average landfill gas recovery rates which has been normalized to 50% CH_4. Despite the variety of individual landfill sites, there is a statistically significant correlation between all the sites ($P < 0.001$), which allows an estimate of the average biogas recovery rate for all landfill sites in California.

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measured field data for 10 California landfill sites (Bogner et al., 2011; Goldsmith et al., 2012; Jeong et al., 2013; Peischl et al., 2013; Shan et al., 2013; Spokas et al., 2011).

2.2 CALMIM model

We utilized the data given in Walker et al. (2012) for data on WIP, waste footprint, cover materials, biogas recovery, and CH_4 content. Therefore, consistent with recent literature emphasizing strong seasonal dependencies for CH_4 transport, oxidation, and emissions in other managed and pristine soil ecosystems (Cao et al., 1995; Wille et al., 2008), CALMIM modeling was utilized to generate an estimate of site emissions and these results were compared to the existing 2010 California inventory (Deshpande et al., 2014) relying on IPCC methodology (IPCC, 2006; Supplemental Information). The major research questions were:

- Which sites and cover materials were responsible for the highest emissions and largest % of landfill CH_4 emissions?
- Statewide, how does monthly CH_4 oxidation vary over an annual cycle?
- How do "net" emissions with oxidation relate to the important climatic variables affecting oxidation rates (i.e., temperature, precipitation)?

3.0 Results

3.1 Database correlations

There was a relatively robust linear relationship [Fig. 1; $R^2 = 0.82$] observed between waste WIP (tons) and average biogas recovery rate for the landfills in the California dataset [n=128 (dropped Puente Hills)]. From this, we can estimate a normalized LFG recovery rate of 126 x 10^{-6} Nm^3 CH_4 hr^{-1} Mg_{waste}^{-1}. It is interesting to note that almost 90% of the waste in permitted California landfills has engineered gas extraction (Table S2). Figure 1 suggests that a relatively constant rate of gas generation and recovery can be maintained over long time periods for a wide variety of small to large, open and closed sites across diverse climatic regions of California. In addition, this simple relationship is further supported when examined against values from other US and international landfills (Figure S2), with improved predictability of closed landfills in the USEPA landfill methane outreach program database, with only 2% of sites falling outside of the 95% confidence intervals of this relationship (Figure S2c).

To address whether biogas recovery rates are related to climate and landfill operational factors (e.g., landfill age, open or closed status), we initially screened the California data for correlations (Figure S4) and step-wise regressions (Table S3). The only statistically significant correlation for the entire dataset was between *biogas flow* and *WIP* (Figure 1). The step-wise regression analysis indicated that *WIP* was a dominant factor controlling biogas recovery rate ($P = 2 \times 10^{-16}$); *disposal starting year* was also statistically significant ($P = 0.01$), but with a much lower coefficient (17.1 ± 6.7; Table S3). Notably, none of the climate variables (air temperature or precipitation) were statistically significant in this regression analysis, which suggests the lack of climate dependency on the biogas production rate.

3.2 New 2010 CALMIM inventory compared to 2010 CARB inventory estimates

Using CALMIM, the 2010 CH_4 emissions were estimated at 337,430 Mg CH_4 yr^{-1} compared to the CARB inventory value of 301,748 Mg CH_4 yr^{-1} (Figure S3). Despite this numerical similarity, the spatial distribution for these predictions is drastically different (Figure 2). The similarity of the totals suggests that, for selected

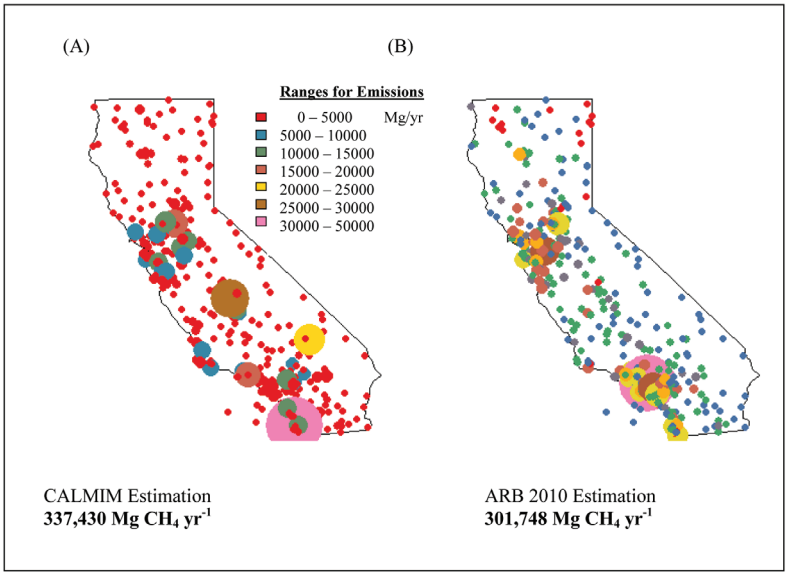

Figure 2

Comparison of the spatial distribution of the (A) CALMIM estimations and the (B) CARB 2010 FOD estimations. Values are in Mg CH_4/yr per site.

This figure illustrates differences in the spatial distribution of the two different inventories, with the new CALMIM modeling predictions shown in the left panel and the existing 2010 CARB estimates (IPCC, 2006) shown in the right panel. The existing CARB inventory is based on the mass of waste in-place, whereas the CALMIM inventory predicts the average emissions based on local climate, cover materials, and cover areas (Walker, 2012).

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sites, there may also be a serendipitous similarity for some sites between the measured emissions and current CARB inventory values. The top ten emitting landfill sites differ between the new CALMIM (Fig. 2A) and the 2010 CARB inventory (Fig. 2B). Using CALMIM, the highest-emitting sites are in the desert areas, Central Valley, and higher elevation mountain sites with low annual oxidation due to lack of favorable conditions for CH_4 oxidation. Focusing on the intermediate cover, which is 47% of the total reported landfill area but accounts for 96% of the estimated landfill emissions, there is a very strong relationship with precipitation (Figure 3A). Notably, for sites receiving >500 mm of precipitation, the predicted intermediate cover emissions were less than 15 g CH_4 m^{-2} d^{-1}. Moreover, for sites receiving <500 mm of precipitation, there is an exponential increase in the emission rate with decreasing precipitation, which is attributed to the lack of adequate soil moisture at these locations to support soil CH_4 oxidation activity (Figure S3) (Boeckx et al., 1997; Spokas and Bogner, 2011). For mean annual air temperature (MAT; Figure 3B), the relationship is less robust, likely confounded by corresponding precipitation differences. However, there is the suggestion of an optimum MAT of 11°C associated with the lowest emissions and highest rates of soil CH_4 oxidation. This temperature is, of course, below optimum temperatures for methanotrophic oxidation in controlled laboratory studies (typically 30–40 °C) (Börjesson and Svensson, 1997; Spokas and Bogner, 2011), since it

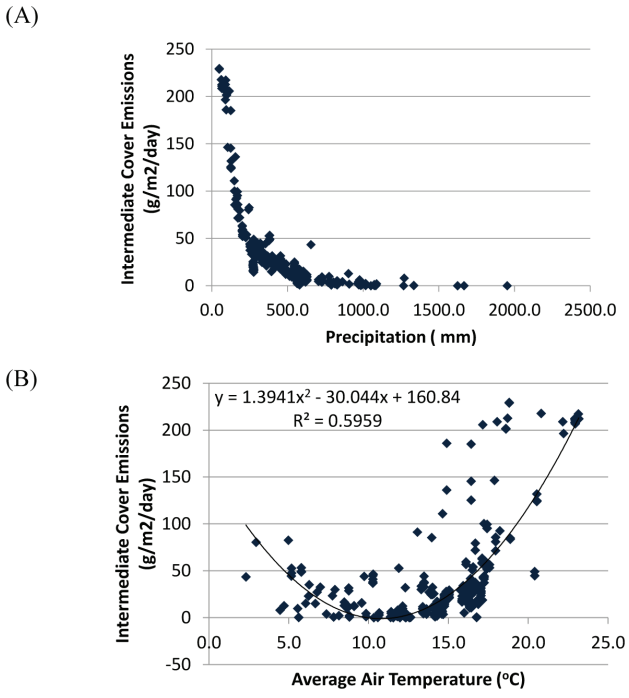

Figure 3

Illustration of the relationship between the predicted intermediate cover emission rate (g/m^2/day) and the site's (A) average annual precipitation and (B) the average air temperature (all California landfill sites; n = 371).

This figure compares the relationship between predicted landfill emissions at a site and the corresponding average annual precipitation and temperatures for California. The predicted emissions are controlled by the average precipitation, and to a lesser degree by the annual temperature.

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Table 1. Monthly totals (Mg CH_4/month) for the new California statewide inventory summarizing the amount of methane oxidized, percent oxidation, and the estimated surface emissions with and without soil oxidation

	Total estimated emissions with soil oxidation (Mg/month)	Total emissions without oxidation (Mg/month)	CH_4 oxidized (Mg/month)	% oxidation prediction
Jan	6,403	164,045	157,641	96
Feb	5,972	157,461	151,489	96
Mar	5,500	162,088	156,589	97
Apr	5,183	185,449	180,267	97
May	5,849	201,810	195,962	97
Jun	7,874	216,885	209,011	96
Jul	12,005	229,276	217,271	95
Aug	28,114	239,786	211,672	88
Sep	73,885	251,906	178,022	71
Oct	89,611	269,914	180,303	67
Nov	66,465	283,247	216,782	77
Dec	30,569	249,318	218,749	88
Annual Totals (Mg/yr)	**337,430**	**2,611,187**	**2,273,758**	**87%**

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integrates annual temperature and precipitation cycles. In particular, both desert areas of California [high MAT, low precipitation] and high elevation areas [lower MAT] are associated with higher emissions and lower soil oxidation capacities.

From CALMIM modeling, Table 1 shows that 2010 monthly CH_4 emissions for California vary about 17-fold with minimum rates in April [5,183 Mg] and maximum rates in October [89,611 Mg], which agrees with the seasonal pattern observed in prior California field assessments (e.g., Goldsmith et al., 2012; Park and Shin, 2001; Yazdani and Imhoff, 2010). Lower emissions are typically observed during periods of higher precipitation events (wet season: Aug–Mar) and then elevated surface emissions during the summer (June–Sept). This large differential in rates is attributed to variable CH_4 oxidation rates in cover soils coupled to fluctuating soil moisture and temperature conditions. Without soil oxidation, the seasonal difference is only predicted to be 2-fold by the model due to the lower impact of temperature changes on soil gas diffusion rates (Table 1). For the entire state, monthly totals of CH_4 oxidation range from 151,000 to 217,000 Mg, or an annual total of 2,273,758 Mg CH_4 oxidized for the entire state in one year. This amounts to an average statewide landfill CH_4 oxidation flux density of 62 g CH_4 m^{-2} d^{-1}, accounting for the total area of Californian landfills.

3.3 CALMIM results compared to field data

CALMIM modeled results for landfill CH_4 emissions at 10 California landfill sites were compared to published field measurements, including seasonal data where possible (Bogner et al., 2011; Goldsmith Jr et al., 2012; Peischl et al., 2013; Shan et al., 2013). Figure 4 compares site-specific CALMIM inventory estimates for the 10 sites (Figure S8) to field measurements using multiple methods taken at various times and various dates during 2005–2014. All of the total site emissions, where available, were normalized on an area basis (g CH_4 m^{-2} d^{-1}) for this comparison using the Walker (2012) database for 2010 footprint areas. For all of the sites, the field measurements and CALMIM inventory estimates are within the same order of magnitude (Table 2).

One must also keep in mind that a field measurement campaign only represents a "snapshot" in time without any information regarding the temporal variability in emissions or oxidation over the annual cycle. To a large extent, this figure also illustrates the difficulty of site-specific emissions comparisons to CALMIM modeling in the absence of site-specific data for the major drivers for oxidation and emissions (soil moisture, soil temperature). The site-specific differences between measured and modeled values may be due to the variability in the physical characteristics of site-specific cover soils (e.g., texture, thickness) and annual soil microclimate (i.e., soil moisture, temperature). Figure 4 illustrates the range of field measurements (shown in the colored points for the month the measurements were conducted) compared to CALMIM-modeled CH_4 emission ranges for each site (upper blue line represents no soil oxidation, black line oxidized flux prediction, and shaded region for the range between the oxidized and non-oxidized emission estimate). As each field campaign represents only a snapshot in time, it is important to put the measurements into the context of

Table 2. Comparison of the CALMIM and CARB inventory for the sites with field measurements

California Solid Waste Information System Identifier	CARB (g CH_4/m^2/day)	CARB 2010 emissions (MT CH_4/yr)	2010 Waste-In-Place (tons)	Int (gCH_4/m^2/day)	Daily (gCH_4/m^2/day)	Final Cover (gCH_4/m^2/day)	Site Calculated CALMIM Emissions (g CH_4/m^2/day)	CALMIM Total (Mt CH_4/yr)	Range of field measurements (g CH_4/m^2/day)	Percentage of Intermediate Cover at Landfill
01-AA-0008	16.03	2723	10103797	14.11	7.65	0.00	13.55	2,301	5.6 – 7.1	91%
01-AA-0009	36.38	12627	44281078	29.41	7.96	0.00	23.99	8,328	0.7 – 12.8	80%
21-AA-0001	13.18	4333	14143215	15.98	7.51	0.00	15.60	5,126	5.3-12.0	96%
19-AA-0012	10.26	6670	29409357	40.10	8.60	0.00	27.90	18,133	1.5	69%
19-AA-0050	4.32	606	6225912	66.83	9.21	0.00	60.77	8,527	0.08-2.43	89%
19-AA-0053	33.33	29537	124963317	40.10	8.60	0.00	7.09	6,287	0.88 (final cover) 38.4 (plane whole site) 8.5 (chambers)	17%
19-AA-0056	8.57	5265	23441895	45.14	8.65	0.00	44.27	27,202	0.05 (final cover)	98%
27-AA-0010	4.35	2025	8388784	24.47	8.12	0.00	23.95	11,143	56.4 (intermediate cover)	97%
30-AB-0035	21.12	13105	52017040	36.78	8.17	0.00	36.10	22,397	4.3 – 20 (intermediate cover)	98%
43-AN-0008	10.17	2403	7312751	22.19	7.72	0.00	21.28	5,030	0.07 – 20.9 (intermediate cover)	94%

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expected emissions variability over a typical annual cycle (Figure 5; Figure S9). The main observation from the new CALMIM inventory and the field measurements is the lack of any significant relationship between these two estimates and the WIP (Figure S10).

4.0 Discussion

Based on the correlation between WIP and average biogas recovery rates in the 2010 California dataset, we can estimate a normalized LFG recovery rate of 126 x 10^{-6} Nm^3 CH_4 hr^{-1} Mg^{-1}_{waste} (Figure 1), which appears very robust with the existing data from other studies (Figure S2). Unlike previous estimates based on small datasets or laboratory studies (Gioannis et al., 2009), this is the first time that a large internally-consistent database of full-scale sites has been available for this analysis. It is important to note that these data include older landfill sites (>50 years old), the first U.S. engineered landfills [1960s], and the first biogas recovery projects [1970s]. The average recovered CH_4 concentration was 36.5 ± 11% CH_4 (v/v), which is lower than the typical range for produced biogas [50% CH_4]. This could be due to mixing with air, since many California recovery systems tolerate lower CH_4 concentrations to comply with strict air quality regulations [including quarterly surface scans for elevated CH_4 concentrations at ground level] and to minimize nuisance odors. We normalized the biogas recovery data to 50% CH_4 to remove this variable effect.

Coupled with local climate, there is a strong seasonal imprint on CALMIM's prediction of the site's emission profile (Table 1). In a California study, Park and Shin (2001) documented temporally variable CH_4 emissions, including maximum fluxes temporally corresponding with maximum surface temperatures above optimum for CH_4 oxidation. For California studies, Yazdani and Imhoff (2010) observed lower CH_4 oxidation rates in the Fall (Oct) than the Spring (March), and Bogner et al. (2011) measured lower wet season (March) and higher dry season (August) CH_4 fluxes. Park and Shin (2001) documented temporally variable CH_4 emissions, including minimum fluxes corresponding with minimum surface temperatures (cooler, wet season; March) and maximum fluxes corresponding with maximum temperatures (above optimum for CH_4 oxidation). This dependency has been observed ever since the first field and laboratory study for annual oxidation in landfill soils, which led to the current 10% default in IPCC (2006) for annual soil methane oxidation, based on one site in New Hampshire, USA (Czepiel et al., 1996a). However, this temporal variability, which takes into account local soils and climate, has not been previously embedded in an inventory methodology.

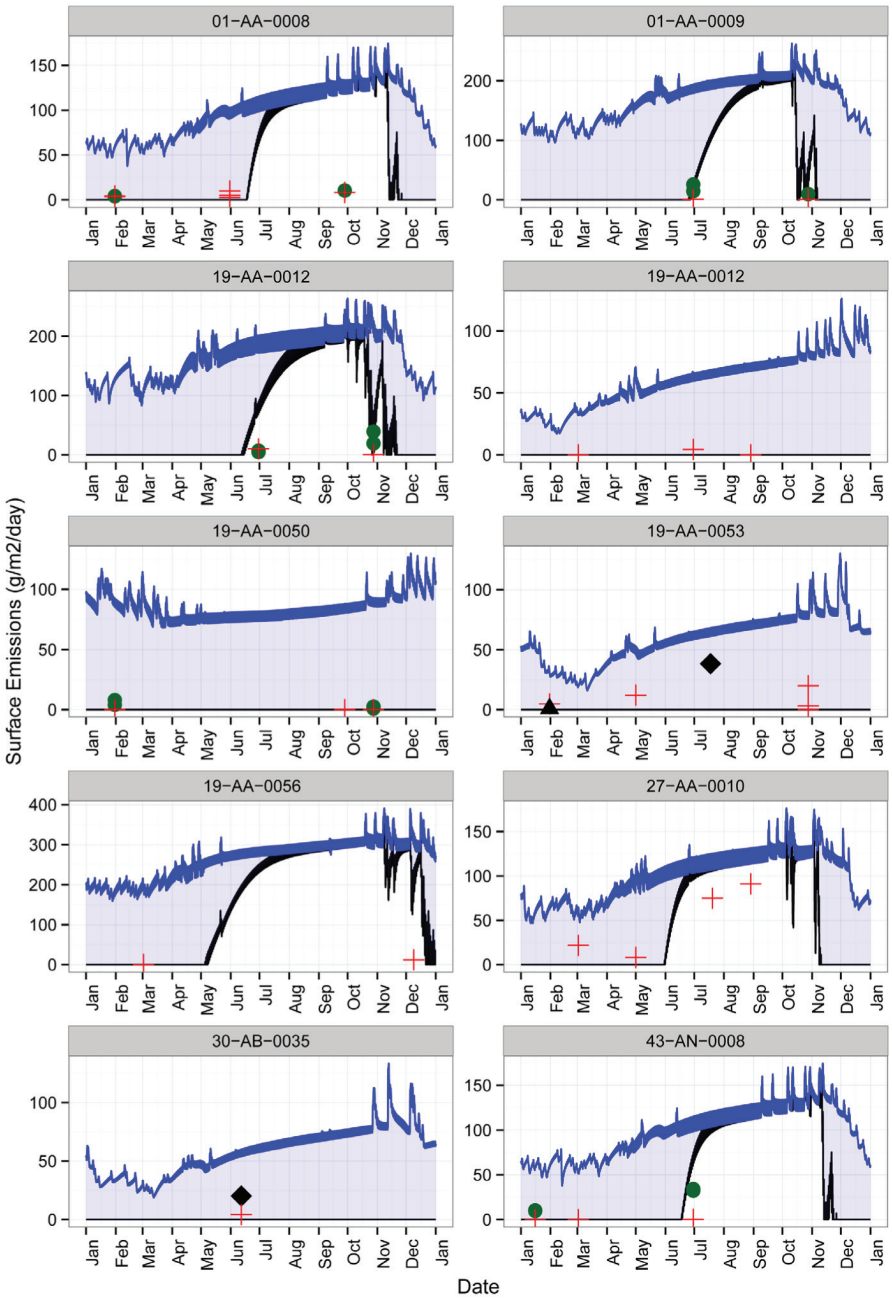

Figure 4

Illustration of the predicted emission rates for the CALMIM model for 10 California landfill sites compared to the corresponding field measurements.

All units are in g m^{-2} d^{-1}. Field results are plotted for the month of the measurement with different symbols representing the different techniques: Red plus sign indicates surface chambers (Spokas et al, 2011; Shan et al, 2012), black diamond/triangles indicates aircraft plume measurements (Peischl et al, 2013; Tratt et al, 2014; Turner et al, 2015), and the green circle stand for vertical radial plume mapping methodology (Goldsmith et al, 2012). For CALMIM results, the blue line represents surface emissions without soil methane oxidation and the black line is the predicted emissions with soil methane oxidation, the region between these predictions is shaded in light blue.

The CALMIM estimates were compared to 10 sites from the literature to compare the predicted site emissions to assessments of site emissions by different field methods. Overall, the CALMIM estimate is within the same order of magnitude as the field assessments with individual differences related to field variability in cover thickness and annual weather.

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In a recent review of field studies using stable carbon isotopic methods, average oxidation has generally been 30–40% across a variety of sites (Chanton et al., 2009).

To improve inventory estimates for landfill CH_4 emissions, it is clear that the seasonality of soil oxidation, consistent with site-specific cover soils and climate, need to be considered. Previous literature has described 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 (Müller et al., 2006)], but similar seasonal models have not been developed for landfills. There have also been a number of recent studies attempting to improve the mathematical prediction of landfill CH_4 emissions inclusive of spatial and temporal variability (Chiemchaisri et al., 2011; Goldsmith Jr et al., 2012; Harborth et al., 2013; Rachor et al., 2013) and consideration of major controls on soil methanotrophy (Albanna et al., 2007; Bogner et al., 1997; Chanton and Liptay, 2000; Czepiel et al., 1996a; Lee et al., 2009; Pawłowska et al., 2003; Pratt et al., 2013). However, to date, the universal default method for estimating landfill CH_4 emissions has retained reliance on empirical models for biogas generation; indeed, recent proposals have suggested additional modifications including further revisions for k values assumed to be related to

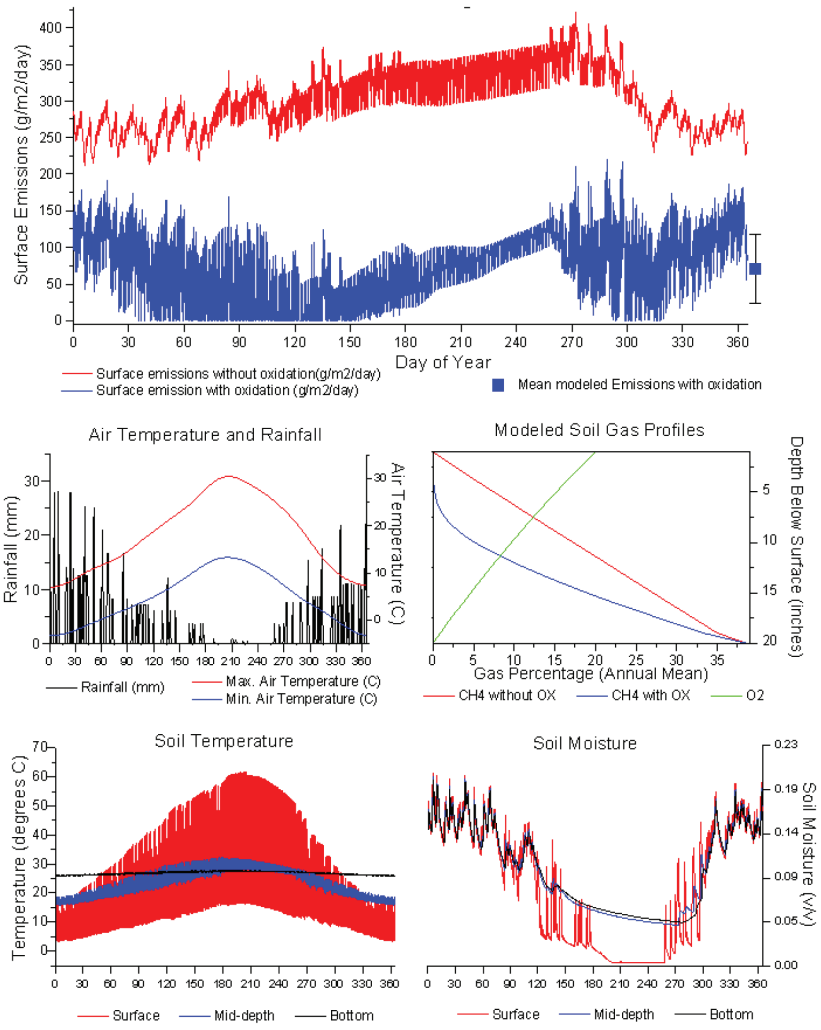

Figure 5

Typical annual cycle for landfill emission predictions from a 50 cm intermediate cover, precipitation, air temperature, soil gas profile, soil moisture, and soil temperature for landfill located in Redding, California (40.59 °N; –122.39 °W).

The CALMIM model output highlights the important characteristics of the annual cycle to landfill emissions. These differences are controlled by the annual climate and soil type at the site. As seen in this figure, the soil temperature is highly variable and the temperature differential increases as soil dries (day 160–250). This decrease in moisture also limits the activity of the soil methanotrophs leading to an increase in the predicted emissions during this time period (blue line in top graph; day 160–250).

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climate (Amini et al., 2012; Garg et al., 2006; Karanjekar, 2012; Sormunen et al., 2013). Concurrently, there have also been more mechanistic models developed to simulate gas diffusion and/or advection processes in landfill cover soils (Abichou et al., 2006a, 2011, 2006b; De Visscher and Van Cleemput, 2003); however, these detailed modeling efforts have complex requirements for site-specific input parameters with uncertain variability which cannot be readily translated to a known precision for regional inventory purposes. Finally, some recent studies have also proposed the use of artificial neural networks (ANN) to account for overall soil complexity in the absence of robust mechanistic models addressing interrelated factors (Young et al., 2001). As an example, Abushammala et al.(2013a) utilized an ANN to predict the percentage of oxidation for a particular landfill, which they assumed could account for a variety of climatic and soil properties at a particular site, then proposed inserting this improved percentage in the IPCC guidelines in place of the current 10% default value (Abushammala et al., 2013b). However, ANN models would require separate training (calibration) for different soil textures, climates, and cover geometries, greatly complicating their application.

CALMIM, like all models, is an abstraction from reality and represents a simplification of complex soil processes. By simplifying the emissions process to 1-D diffusion inclusive of seasonal oxidation at a particular site, this model represents a first step toward accounting for the site-specific seasonality of landfill CH_4 emissions neglected by current inventory methods. As whole site measurements of landfill emissions become more common, there are implications that the homogenous source assumption has on the ultimate validity of the estimation methods (Tratt et al., 2014).

Using California as a test case, with homage to the California origins of the 1st order kinetic framework for the IPCC (1996, 2006) inventory methodology for landfill CH_4 emissions, we used field data from 128 currently-permitted landfill sites to develop a simple empirical relationship for biogas generation & recovery from the waste mass. Importantly, this direct relationship circumvents issues with selection of kinetic constants and "recovery efficiency" assumptions made with no field data support, which has been much discussed in previous literature (Di Bella et al., 2011; Oonk, 2012; Xue and Liu, 2013). The strong correlation (Fig. 1) indicates a universal biogas production-recovery rate per unit mass waste that is statistically robust across

California landfills of different sizes, geometries, ages of waste, and climatic regions. This relationship also holds at other non-California sites (Figure S2). Since landfill covers are designed to limit precipitation/infiltration entry with designated regulatory cover designs (Coccia et al., 2013; Hanson et al., 2010), this also provides thermal insulation to preserve the self-heating effect of the anaerobic microbial decomposition reactions.

Previously, only a limited number of sites or test cells were typically used for the development of kinetic models for biogas generation requiring individual site "calibration" (Amini et al., 2013; Emcon, 1980; Faour et al., 2007; McBean, 2011), including the Dutch studies underpinning the current IPCC model based on degradable organic carbon (Oonk, 2010; Oonk and Boom, 1995; Van Zanten and Scheepers, 1995).

Using CALMIM, which was previously developed and field-validated for California, we developed a new 2010 statewide inventory for landfill CH_4 emissions and compared the results to field measurements. The highest-emitting sites shifted significantly from the CARB inventory, from the sites with the largest mass of waste (CARB) to the sites with low annual oxidation and large areas of thinner intermediate cover soils (CALMIM). For the entire state, based on cover types, CH_4 emissions averaged 10.6 (daily), 325.3 (intermediate), and 1.5 (final) g CH_4 m^{-2} yr^{-1}, respectively, resulting in >95% of the total emissions originating from intermediate cover areas. This shift from sites with the largest waste mass (CARB) has profound implications for developing improved local and regional inventories consistent with a growing database of whole landfill measurements (e.g. Peischl et al., 2013; Cambaliza et al., 2015) and will result in greatly-improved CH_4 inventories inclusive of landfill sources.

In comparisons with data from field campaigns at 10 sites, CALMIM model results show good agreement with field data and are consistent with literature indicating elevated emissions from thinner intermediate cover soils (Abichou et al., 2006a). From the CALMIM results, the ten highest-emitting landfill sites are characterized by >70% of the waste footprint being covered by intermediate cover soils. Conversely, the CARB results indicate the highest emissions consistently occurring at sites with the largest amount of waste, despite the fact that some of these sites also have large areas of final cover (Table 2). This association of high CH_4 emissions with large areas of final cover is inconsistent with literature indicating lower emissions from thicker final cover soils (Abichou et al., 2006a; Goldsmith Jr et al., 2012; Park et al., 2001).

We recognize that we are proposing a new methodology for GHG inventory calculations for landfill CH_4 emissions that differs significantly from historic methods based on estimated generation with climate dependencies and subsequent allocation of a fraction of the estimated generation to surface emissions. However, as field and laboratory studies over the last two decades have emphasized the soil- and climate-related dependencies for emissions, and as herein demonstrated for California, it is time to reconsider the historic methodology which is misleading with respect to average annual emissions at specific sites, the regional [spatial] distribution of emissions, and the seasonal [temporal] variability of emissions. For ultimately reducing landfill CH_4 emissions in California, thicker intermediate covers could be installed, as is already practiced at some sites (see Figure S8). Some remaining uncertainties, requiring further study, include:

(1) The magnitude of daytime CH_4 emissions from the daily filling area (where daily cover is placed at the end of working day), especially where this area overlies older cells with fully methanogenic waste (discussed in SI, Cambaliza et al., 2015). This is the norm at large sites with multiple layers of cells where the intermediate cover is stripped before new overlying cells are developed. As a result, daytime emissions for "daily cover" areas may be substantially higher than nighttime emissions (after placement of daily cover).
(2) Determining optimum cover soil thickness. CALMIM modeling indicates that for a particular soil at a particular location (latitude/longitude), an "optimum" thickness can be determined for minimum emissions due to maximum CH_4 oxidation. At thicknesses greater than the optimum there are seasonal diffusional limitations for O_2 transport to the lower portion of the soil profile and, hence, reduced oxidation.
(3) International field validation of CALMIM. Because CALMIM includes embedded globally-validated climate and soil microclimate models (Spokas et al., 2011), it should be applicable to other U.S. sites and international sites—this is currently being tested using available emissions measurements.

To conclude, in order to achieve a better science-based quantification of landfill CH_4 emissions there is the need to replace the current GHG inventory methodology with a more robust approach based on the correct drivers, including site-specific cover soils and climate-based estimation of seasonal oxidation in landfill cover soils.

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Contributions

- Contributed to conception and design: JB, KS, MF, SW
- Contributed to acquisition of data: JB, KS, SW, MC
- Contributed to analysis and interpretation of data: KS, JB, MF, SW
- Drafted and/or revised the article: KS, JB, MF, SW
- Approved the submitted version for publication: JB, KS, MF, SW

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Competing interests

The authors have declared that no competing interests exist.

Supplemental material

- **Figure S1. Location of all California landfill sites in the database used here, colors indicate whether the site has a landfill gas recovery system.**
  A) Blue dots indicate no LFG recovery system, B) Red dots indicate sites with a LFG recovery system and corresponding flow data in the database (n = 128), and C) Green dots are sites which have a landfill gas recovery system, but did not report any flow data in the database (see supplemental file Spreadsheet S2 *CA_LANDFILLDATABASE.xlsx*). doi: 10.12952/journal.elementa.000051.s001

- **Figure S2. Comparison of (A) other landfill waste in place (WIP) and landfill gas recovery data from peer reviewed literature sources, B) comparing to the data from the landfill methane outreach program voluntary database (link) for B) open and (C) closed landfills.**
  Note the improved capturing of the "closed" site data by this empirical relationship. doi: 10.12952/journal.elementa.000051.s002

- **Figure S3. Spatial distribution of the (A) California waste-in-place estimates from the Walker (2012) database and (B) the CARB 2010 landfill CH4 emission estimates (Mg CH_4/yr).**
  Notice the correlation between waste in place and the predicted CH_4 emission plots in (C). doi: 10.12952/journal.elementa.000051.s003

- **Figure S4. Correlations across the entire CA landfill database for those sites with gas recovery data (n=129).**
  Shown in the Figure is the Pearson correlation with the numbers showing only those correlations that were significant at the 95% confidence interval (P=0.05). Note the significant correlation (r=0.96) between WIP (waste in place) with flow and average annual flow of landfill gas. There were no other significant relationships observed between any other quantities. doi: 10.12952/journal.elementa.000051.s004

- **Figure S5. Comparing the geographic locations of the top emitting landfill sites between the (A) CALMIM and (B) CARB values following first order decay model (current IPCC methodology).** doi: 10.12952/journal.elementa.000051.s005

- **Figure S6. Estimated annual area normalized intermediate cover emissions for all California landfills (g m^{-2} d^{-1}).**
  Note the geographical clustering of the results, which is similar to the trends in the annual climate variability in California (see **Figure S7**). doi: 10.12952/journal.elementa.000051.s006

- **Figure S7. Geospatial distribution of annual mean (A) precipitation (mm of water) and (B) air temperature (°C) for California landfill sites.** doi: 10.12952/journal.elementa.000051.s007

- **Figure S8. Distribution of the 10 landfill sites in California where field data was compared to the CALMIM model predictions (Figure 4 in manuscript).** doi: 10.12952/journal.elementa.000051.s008

- **Figure S9. Impact of cover thickness (cm) for a sandy loam textured cover material on the predicted annual surface emission and soil oxidation rates at a simulated California landfill (36.9 °N; 121.8 °W) summed over the annual cycle.** doi: 10.12952/journal.elementa.000051.s009

- **Figure S10. Comparison of the relationship between WIP and the total CARB emissions (shown in blue), the new CALMIM inventory estimates (shown in red), and the average of the field measurements (shown in green).**
  Note the lack of any significant relationship between the CALMIM and the field measurements with the waste in place (Table 2). doi: 10.12952/journal.elementa.000051.s010

- **Table S1. Typical landfill cover types**
  This table details the typical cover configurations for daily, intermediate and final cover areas of a landfill. doi: 10.12952/journal.elementa.000051.s011

- **Table S2. Waste in place as a function of LFG recovery**
  This table details the division of the waste in place (WIP) with and without landfill gas recovery in 2010. As seen in the table, 89.3% of the WIP is under landfill gas recovery in California. doi: 10.12952/journal.elementa.000051.s012
- **Table S3. Stepwise regression output for the 2010 Database**
  This table presents the results of the multiple linear regression modeling examining other potential relationships in the 2010 dataset. The WIP is significant at the $P<0.001$ level ($P = 2 \times 10^{-16}$) and the Disposal Start Year is also significant ($P=0.0125$). This analysis was conducted in R. doi: 10.12952/journal.elementa.000051.s013
- **Table S4. Comparison of the top ten CALMIM emitting California sites**
  This table presents the results of the highest 10 emitting California landfill sites when calculated using the CALMIM model. doi: 10.12952/journal.elementa.000051.s014
- **Table S5. Comparison of the top ten CARB emitting California sites**
  This table presents the results of the top 10 emitting California landfill sites when calculated using the CARB (IPCC) methodology. doi: 10.12952/journal.elementa.000051.s015
- **Spreadsheet S1: CALMIM_CARB_InventoryComparison.xlsx**
  This Microsoft Excel file contains the site emission estimate following the IPCC methodology, the 2010 waste in place (tons) data, and results of the CALMIM (Mg/yr) modeling for all the sites in the SWIS California database. doi: 10.12952/journal.elementa.000051.s016
- **Spreadsheet S2: CA-LANDFILLDATABASE.xlsx**
  This Microsoft Excel file contains the data from the Walker et al. (2012) compilation. doi: 10.12952/journal.elementa.000051.s017

Data accessibility statement

All data is included in the supplemental information along with the CALMIM model at http://www.ars.usda.gov/services/software/download.htm?softwareid=300.