

Seasonal Greenhouse Gas Emissions (Methane, Carbon Dioxide, Nitrous Oxide) from Engineered Landfills: Daily, Intermediate, and Final California Cover Soils

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Compared with natural ecosystems and managed agricultural systems, engineered landfills represent a highly managed soil system for which there has been no systematic quantification of emissions from coexisting daily, intermediate, and final cover materials. We quantified the seasonal variability of CH₄, CO₂, and N₂O emissions from fresh refuse (no cover) and daily, intermediate, and final cover materials at northern and southern California landfill sites with engineered gas extraction systems. Fresh refuse fluxes (g m⁻² d⁻¹ [± SD]) averaged CH₄ 0.053 (± 0.03), CO₂ 135 (± 117), and N₂O 0.063 (± 0.059). Average CH₄ emissions across all cover types and wet/dry seasons ranged over more than four orders of magnitude (<0.01–100 g m⁻² d⁻¹) with most cover types, including both final covers, averaging <0.1 g m⁻² d⁻¹ with 10 to 40% of surface areas characterized by negative fluxes (uptake of atmospheric CH₄). The northern California intermediate cover (50 cm) had the highest CH₄ fluxes. For both the intermediate (50–100 cm) and final (>200 cm) cover materials, below which methanogenesis was well established, the variability in gaseous fluxes was attributable to cover thickness, texture, density, and seasonally variable soil moisture and temperature at suboptimal conditions for CH₄ oxidation. Thin daily covers (30 cm local soil) and fresh refuse generally had the highest CO₂ and N₂O fluxes, indicating rapid onset of aerobic and semi-aerobic processes in recently buried refuse, with rates similar to soil ecosystems and windrow composting of organic waste. This study has emphasized the need for more systematic field quantification of seasonal emissions from multiple types of engineered covers.

LANDFILLS ARE THE SECOND LARGEST anthropogenic source of CH₄ in the United States after ruminant animals; global estimates indicate that landfill CH₄ accounts for 1 to 2% of total global anthropogenic greenhouse gas emissions (USEPA, 2010; Bogner et al., 2007; Rogner et al., 2007). Field studies have documented landfill CH₄ emission rates spanning six to seven orders of magnitude with high spatial variability of three to four orders of magnitude within distances of <10 m (Bogner et al., 1997a; Scheutz et al., 2009). To date, however, the literature has focused on CH₄ emissions from final cover materials and not systematically addressed CH₄ emissions in parallel with other greenhouse gases (CO₂, N₂O) across the full range of daily, intermediate, and final cover materials at specific sites. In the current study, we also quantified emissions from active filling areas before daily cover placement, which is relevant to emissions of greenhouse gases from waste disposal in developing countries without engineered sites or use of daily cover.

Landfill CH₄ emissions are reduced by three major mechanisms, all of which are dependent on engineering and management practices (Bogner et al., 1997a; Scheutz et al., 2009; Bogner and Spokas, 2011). These mechanisms are (i) most important, the presence or absence of engineered gas recovery and control systems; (ii) the thickness and physical properties of engineered cover materials; and (iii) seasonal CH₄ oxidation by indigenous aerobic methanotrophs in daily, intermediate, and final cover soils. These mechanisms operate simultaneously to reduce emissions—for example, thicker cover materials retard transport and increase retention times to facilitate oxidation; engineered systems reduce CH₄ concentrations at the base of the cover, resulting in lower diffusive fluxes which can be more readily oxidized. Moreover, at well-controlled sites with optimized gas recovery systems (>90% of modeled generation), field-scale CH₄ mass balance data for individual cells with well-quantified waste inputs have demonstrated that, while measured CH₄ recovery can be linearly related to modeled CH₄ generation, no relationship exists between modeled CH₄ generation and small residual CH₄ emissions, which vary over several orders of magnitude (Spokas et al., 2006; Bogner and Spokas, 2011). Published literature over the last two decades

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Abbreviations: MDC, Marina daily cover; MFC, Marina final cover; MIC, Marina intermediate cover; NMOC, nonmethane organic compound; SDC, Scholl daily cover; SFC, Scholl final cover; SIC, Scholl intermediate cover.

has concluded that landfill cover soils can develop high capacities for CH₄ oxidation with rates dependent on cover soil properties as well as temporal changes in soil moisture, temperature, and other variables (Whalen et al., 1990; Kightley et al., 1995; Czepiel et al., 1996; Bogner et al., 1997b, 2010; Scheutz et al., 2003, 2009; Barlaz et al., 2004; Cabral et al., 2010; Spokas and Bogner, 2011). In a recent complementary laboratory study focusing on the same California landfill cover soils addressed in this paper, we demonstrated that a 60-d preincubation with CH₄ and adjustment of soil moisture potential to field capacity (-33 kPa) can optimize CH₄ oxidation rates to uniformly high values of 112 to 644 μg g soil⁻¹ d⁻¹ (Spokas and Bogner, 2011). These are the highest rates reported in the literature. When landfill cover soils develop high oxidation capacities, especially where gross fluxes are low, it is also possible to initiate oxidation of atmospheric CH₄ (Bogner et al., 1995, 1997b, 2010), as has been observed in natural ecosystems (Whalen and Reeburgh, 1990). Recent field measurements of landfill CH₄ emissions using a variety of aboveground techniques (micrometeorological, static and dynamic tracer, horizontal and vertical radial plume mapping) have shown wide variability in results (Babilotte et al., 2009), resulting from the complex topography and micrometeorology typical at many landfills, differing interpretation algorithms, and variable emission signals from daily, intermediate, and final cover areas in close proximity to each other. In addition to CH₄ emissions, there are only a few previous studies in the literature that have addressed landfill emissions of N₂O and/or CO₂ in parallel with CH₄ (Börjesson and Svensson, 1997; Bogner et al., 1999; Lee et al., 2002; McBrain et al., 2005; Rinne et al., 2005; Zhang et al., 2008).

In the United States, which is an Annex I country signatory to the United Nations Framework Convention on Climate Change, CH₄ emissions from landfills are annually reported using Intergovernmental Panel on Climate Change (IPCC) national greenhouse gas inventory guidelines (IPCC, 2006; USEPA, 2010). Under these guidelines, however, landfill emissions of CO₂ are considered biogenic, not separately reported for the waste sector, and quantified under annual carbon balances under Agriculture, Forestry, and Other Land Use. Current U.S. regulations implemented in state programs under the Clean Air Act amendments and successor legislation mandate only the regulation of estimated emissions of non-methane organic compounds (NMOCs) from landfills, using a hypothetical landfill gas generation model (LANDGEM) and the application of a default or measured mixing ratio for total NMOCs. Some previous studies have addressed field measurement of speciated NMOCs from landfills (Scheutz et al., 2003, 2009; Barlaz et al., 2004; Bogner et al., 2010). At the present time, the USEPA is considering the direct regulation of greenhouse gas emissions from many sources under the Clean Air Act, including the possible regulation of CH₄, N₂O, and biogenic CO₂ emissions (EPA-HQ-OAR-2010-0560; FRL-9175-9) from various waste management processes. Thus, it is important to better understand the rates and dynamics of landfill emissions based on field-scale measurements. The field study discussed herein was part of a broader study to develop and field-validate a new site-specific landfill inventory model for the state of California based on improved scientific understanding of CH₄ emissions inclusive of seasonal oxidation in

California landfill cover soils (Spokas et al., 2009; Bogner and Spokas, 2010; Spokas and Bogner, unpublished data, 2011). In general, the field results discussed in this paper will be linked to two complementary studies: a laboratory study, Spokas and Bogner (2011), as discussed above; and the CH₄ emissions model study (Spokas et al., 2009; Bogner and Spokas, 2010; Spokas and Bogner, unpublished data, 2011).

The purpose of this study was to quantify the spatial and seasonal variability of CH₄, CO₂, and N₂O emissions from fresh refuse and from daily, intermediate, and final cover materials at two contrasting California landfills: a northern coastal site (Monterey County) and a southern canyon fill (Los Angeles County). Both sites have a Mediterranean climate with precipitation predominately during the winter (December through March). The two sites exemplify a typical range for California climatic conditions: Los Angeles has higher mean annual temperatures (17.4°C) and lower mean annual precipitation (334 mm), while Monterey has lower mean annual temperatures (10.4°C) and higher mean annual precipitation (517 mm) (see <http://cdo.ncdc.noaa.gov/climatnormals/clim20/ca> [verified 26 Jan. 2011]).

Materials and Methods

To elucidate differences in gaseous emissions between the various types of cover materials at sites where engineered gas recovery underlies all cover types, this study applied simple static chamber techniques during wet- and dry-season field campaigns over 2 yr. Four intensive field campaigns were conducted at the two field sites (Scholl Canyon Landfill, 34°9.48' N, 118°11.76' W, Los Angeles County; Marina Landfill, 36°42.60' N, 121°45.72' W, Monterey County), two during optimum wet-season conditions (March 2007, 2008) and two during optimum dry-season conditions before the start of the rainy season (August 2007, 2008). See the Supplemental Information for monitoring locations within each site. The six soil cover materials (daily, intermediate, and final soil cover at two sites) were characterized using standard techniques as previously discussed in Spokas and Bogner (2011). Previous literature (Venterea et al., 2009) supports the use of static chambers to quantify the variability of fluxes across specific soil types. Because the engineered controls (gas extraction system; engineered cover materials) were major influences on emissions, we randomized fluxes across each cover type during four 1- to 2-wk seasonal campaigns in 2007 and 2008.

Each chamber had a fixed base, a removable top with gas-tight fittings for sampling using a syringe, a cross-sectional area of 0.10 m², and an average volume-to-area ratio of 0.24 m³ m⁻². Gaseous fluxes were measured using nine stainless steel chambers in parallel during multiple deployments over several days for each cover type for each season, including partial duplication from previous deployments. Chamber locations were recorded using a global positioning system (Garmin, Olathe, KS); in all, there were >850 fluxes completed. Because of site operational constraints, fresh refuse fluxes were typically limited to single deployments of the nine chambers before daily cover placement at the end of the working day. For each flux (30-min maximum), six gas samples were extracted using gas-tight syringes and retained in headspace vials. Fluxes were determined by the product of the change in concentration over

time (dC/dt) and the (chamber volume/chamber area) ratio (Rolston, 1986) with an acceptance criterion of $r^2 > 0.9$ for the linear regression of dC/dt . To sample the source gas, composite landfill gas samples were collected from the engineered gas extraction system, typically from wellfield headers located near the chamber deployment areas. Selected individual wells were also sampled to check whether there were significant deviations from the composite wellfield gas. Shallow subsurface soil temperature (RTD [resistance temperature device] probe), yielding a 0- to 5-cm composite temperature, and volumetric soil moisture (TDR [time domain reflectometry] probe) were also determined at each individual flux location.

All gas samples were analyzed for CH_4 , CO_2 , and N_2O by the USDA-ARS laboratories in St. Paul and Morris, MN, using gas chromatography and gas chromatography-mass spectrometry systems, as previously described in Spokas and Bogner (2011). The minimum detectable fluxes were $12 \text{ mg m}^{-2} \text{ d}^{-1}$ for CH_4 , $0.322 \text{ mg m}^{-2} \text{ d}^{-1}$ for CO_2 , and $0.7 \text{ mg m}^{-2} \text{ d}^{-1}$ for N_2O . In addition, weather stations and 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, Bourne, MA). In general, soil temperature and soil moisture at California landfills exhibit seasonal changes related to contrasting winter wet-season and summer dry-season conditions. In landfill cover soils, the soil temperature variability is typically damped with depth similar to nonlandfill soils; however, at deeper depths near the top of the underlying waste there is a secondary temperature increase reflecting elevated waste temperatures of $>23^\circ\text{C}$ (Yesiller et al., 2005, 2008; Spokas et al., 2009; Spokas and Bogner, unpublished data, 2011). This study concentrated on the variability of emissions among the various cover types during wet-season (March) and dry-season (August) conditions; hence, transient emissions associated with leakages from landfill gas piping and soil desiccation cracks, which under existing regulations must be remediated quarterly as part of routine maintenance, were not part of this study.

To quantify CH_4 oxidation, stable carbon isotopes for CH_4 (and selected samples for CO_2) were analyzed at Florida State University. Samples included the final (30-min maximum) chamber samples, the composite source gas from the landfill gas recovery system, and soil gas from shallow and deep probes (10- to 25-cm depth and >25 cm, respectively). Methane oxidation was determined using previously published methods (Liptay et al., 1998; Chanton and Liptay, 2000) where the percent oxidation is determined from the $\delta^{13}\text{C}$ of the anoxic CH_4 vs. the emitted CH_4 (using either a final chamber sample or a soil gas probe sample). Additional details regarding the isotopic methods and their application are given in the Supplemental Information. Previous literature has demonstrated that when diffusion is the primary flux mechanism, use of the chamber CH_4 underestimates oxidation (Chanton et al., 2008); thus, the chamber results are systematically more conservative than the probe results.

Results and Discussion

We believe that this is the first study to systematically quantify greenhouse gas emissions from daily, intermediate, and final cover soils at the same sites, as well as emissions from uncov-

ered "fresh" refuse during the working day. The experimental design was consistent with the primary importance of engineered controls (gas recovery systems and compacted cover soils with variable thickness) and the secondary importance of natural climatic and soil microclimate variability. In this study, we also included field measurements of N_2O and CO_2 (dark respiration) emissions to provide improved understanding of other greenhouse gas emissions related to C and N cycle processes in landfill cover soils. As discussed in previously published literature, conditions over short vertical distances range from strictly anaerobic in the buried refuse to highly aerobic in the upper portion of the cover materials (Bogner et al., 1999).

Source Gas

Unpublished data from the California Department of Resources Recycling and Recovery indicate that $>90\%$ of the waste in place in the 370 permitted California landfills has an engineered landfill gas collection system, which provides a primary control on emissions (Spokas et al., 2006). More than 70 California sites currently utilize the gas for energy, primarily with onsite electrical generation using engines or turbines. Sites without utilization combust the gas in a high-temperature enclosed flare. California landfilling operations typically include a time series of engineered cells with each being a few hectares in area and several meters thick; subsequent cells are added both adjacent to and overlying the formerly active cells. Each cell includes an engineered high-density polyethylene (HDPE) liner with an overlying gravity drainage system for liquids recovery and treatment. The landfill gas collection and control systems include horizontal collectors installed concurrently with filling, vertical wells installed when cells reach final grade, and additional gas collection from leachate underdrain systems.

The source gas characteristics were defined by sampling the composite wellfield gas and individual wells near the chamber deployment areas. Averaged across all field campaigns (± 1 SD), the Marina well gas had CH_4 394 (± 92) mL L^{-1} and Scholl Canyon had CH_4 323 (± 93) mL L^{-1} . Many California landfill gas collection systems are systematically operated at vacuums sufficient to minimize emissions—thus, CH_4 values are typically less than the 500 to 600 mL L^{-1} CH_4 of generated landfill gas due to dilution with atmospheric air, and CO_2 mixing ratios are also proportionally reduced. Note that both sites had relatively high SDs for CH_4 , which can be attributed to frequent wellfield monitoring and adjustments during normal operations. Both sites had similar CO_2 mixing ratios (Marina 252 [± 61] mL L^{-1} , Scholl Canyon 258 [± 47] mL L^{-1}) but different N_2O signatures (Marina 1.25 $\mu\text{L L}^{-1}$, Scholl Canyon $<0.20 \mu\text{L L}^{-1}$), which bracket average atmospheric N_2O values of approximately $0.32 \mu\text{L L}^{-1}$ (Forster et al., 2007).

Cover Soil Properties and Emissions

In addition to gas recovery, a second level of control for gaseous emissions is the type of engineered cover material, including no cover on "fresh" refuse placed during the working day, very thin "daily" cover, thicker "intermediate" cover, and thick "final" cover placed when portions of the site reach final grade. The landfill cover soils consist of recycled, compacted local soils or permitted alternative cover materials (such as shredded or composted garden waste, biosolids compost). Functional soil

microbial ecosystems develop over time in the landfill cover materials; often these are dominated by methanotrophy. However, based on very limited field data, cover materials can also have N₂O and CO₂ (dark respiration) emission rates in final soil covers that can be similar to grassland or forest ecosystems (Bogner et al., 1999), or, alternatively, have elevated N₂O emissions associated with biosolids-amended covers (Börjesson and Svensson, 1997). Importantly, large areas with thinner intermediate cover materials, but mature methanogenesis in the underlying refuse, may be maintained for several years before placement of final covers; these areas have the potential for substantial CH₄ emissions. Because California landfills constitute a highly managed “end point” system compared with natural wetlands and agricultural systems (Bogner et al., 2000), the climate-related effects constitute a third level of control on gaseous emissions after engineered gas recovery and cover systems, especially with respect to the seasonal effects of soil moisture and temperature on transport and CH₄ oxidation. Typically, California is characterized by coastal Mediterranean to semi-arid climatic conditions, with most of the rainfall and cooler temperatures, plus more abundant natural vegetation, concentrated in the winter-to-spring wet season.

Table 1 summarizes major physical and chemical properties of the cover soils. In California, unlike at many other U.S. sites, HDPE geomembranes are not typically used as part of final cover designs. The cover soils at both sites consisted primarily of sand-dominated and sandy loam soils derived from

local sources with moderate pH, variable percent total carbon, and low to moderate cation exchange capacity. Soil thicknesses ranged from 30 cm for daily cover to >2 m for final covers. Extremely high bulk densities were achieved for the highly compacted Scholl Canyon intermediate and final cover materials, where field moisture contents were relatively low (<20% v/v) and below field capacity (20–35% v/v).

For fresh refuse, monitored at the end of the working day (no cover soil), there were no statistically significant seasonal trends nor site differences during the four field campaigns; hence, those data were composited. Fluxes (± SD; range) from fresh refuse averaged 0.053 g m⁻² d⁻¹ for CH₄ (± 0.034; 0.000569–0.107), 135 g m⁻² d⁻¹ for CO₂ (± 117; 12.6–390), and 0.063 g m⁻² d⁻¹ for N₂O (± 0.059; 0.005–0.181). The high CO₂ fluxes from fresh refuse can be attributed to the rapid onset of aerobic microbial activity in the recently deposited waste, with the higher end of the range similar to the lower end of the range for emissions from windrow composting of garden waste, or 250 to 2900 g CO₂ m⁻² d⁻¹ (Andersen et al., 2010). Somewhat surprisingly, the average N₂O and CH₄ fluxes were similar but with a much wider range for CH₄. The N₂O fluxes can be attributed to the daily burial of fresh refuse with readily degradable fractions (food waste; some plant debris) containing both high moisture and readily available N. The rapid onset of restricted aeration conditions after daily burial results in N₂O generation and emissions, under conditions that can be compared with the interior portions of windrow waste composting operations (Beck-Friis et al.,

Table 1. Selected physical and chemical properties of cover soils at Marina and Scholl Canyon Landfills, California. Data represent field values for sieved samples (<2 mm) at time of sampling (March 2007). Bulk densities for Scholl Canyon intermediate and final cover soils deeper than 10 cm were >2.0 g cm⁻³.

Depth	Total C	Total N	Sand	Clay	pH (1:1)	CEC†	Surface bulk density	Field moisture content	Field capacity moisture content
cm	%					meq 100 g ⁻¹	g cm ⁻³	% v/v	
Marina Landfill, Monterey County									
Daily cover (30-cm average thickness)									
0–30	0.2	0.04	90	8	7.4	6.1	1.35	5	13
Intermediate cover (50-cm average thickness)									
0–10	2.6	0.3	74	10	7.8	18.2	1.60	4	23
10–50	1.5	0.1	68	15	7.1	NA‡	NA	NA	NA
Final cover (200-cm average thickness)									
0–10	4.4	0.4	76	10	6.7	21.2	1.40	19	35
10–50	1.1	0.1	65	25	NA	NA	NA	NA	NA
50–90	1.0	0.2	55	35	NA	NA	NA	NA	NA
90–120	0.7	0.1	10	75	NA	NA	NA	NA	NA
120–200	NA	NA	NA	NA	NA	NA	NA	NA	NA
Scholl Canyon Landfill, Los Angeles County									
Daily cover (30-cm average thickness)									
0–30	1.1	0.09	84	4	7.4	18.5	1.55	5	19
Intermediate cover (100-cm average thickness)									
0–10	0.32	0.04	60	14	8.3	18.7	2.0§	3	31
10–20	0.31	0.04	60	21	NA	NA	NA	NA	NA
20–100	NA	NA	NA	NA	NA	NA	NA	NA	NA
Final cover (>275-cm thickness)									
0–10	1.0	0.1	60	14	8.2	19.1	2.0§	3	32
10–275	NA	NA	NA	NA	NA	NA	NA	NA	NA

† CEC, cation exchange capacity.

‡ NA, data not available.

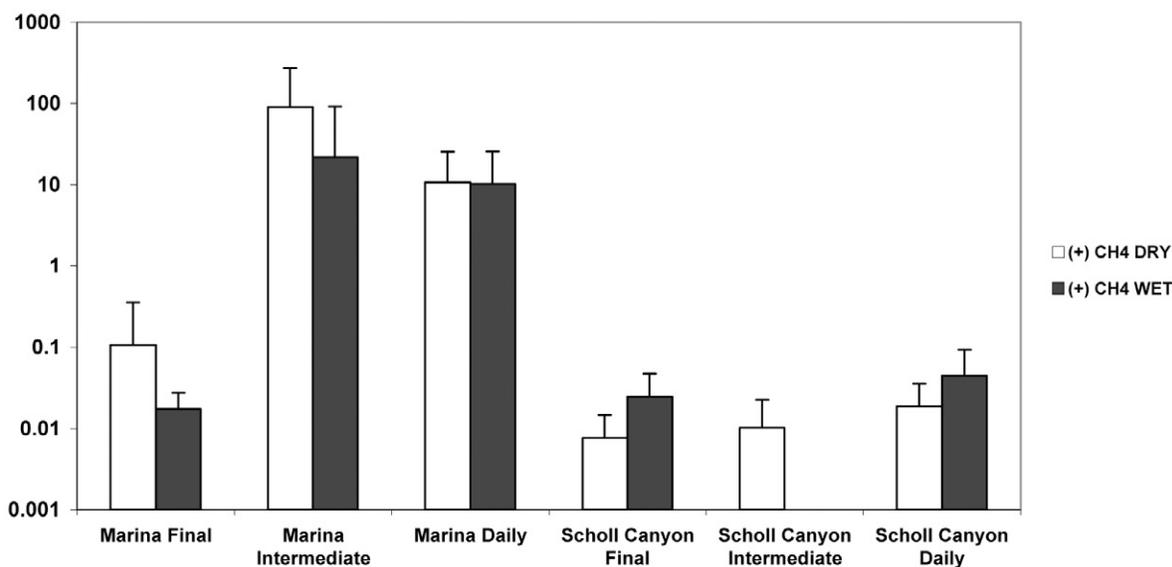
§ Gravel (>2-mm fraction) may have influenced values.

2000; Andersen et al., 2010). Methane emissions are dependent on CH₄ generation, oxidation, and emissions processes in previously buried methanogenic waste, including waste in underlying cells, accounting for the high variability in observed emissions. Largely because of physical difficulties with seating the chamber bases in fresh refuse, approximately 33% of the fresh refuse fluxes had $r^2 < 0.9$ and were rejected, the highest percentage for any cover type.

Overall, average emissions of CH₄, CO₂, and N₂O from the various cover soils varied across several orders of magnitude. Consistent with previous literature, high variability was also documented within cover-specific and season-specific data sets, in large part due to the complexity of gaseous transport, oxidation, and emissions processes in each cover type as influenced by seasonal soil moisture and temperature in this Mediterranean

climate. Because California has distinctive wet (winter–spring) and dry (summer–fall) seasons, the field data for fluxes under wet- and dry-season conditions were respectively composited for the daily, intermediate, and final covers at each of the two field sites (Fig. 1a–1d). The March field campaigns coincided with the end of the winter wet season when soil moisture conditions are highest; the August field campaigns occurred at the end of the summer dry period when soil moisture is minimal. Positive and negative CH₄ fluxes were plotted separately, because these, respectively, represent the different processes of net CH₄ emissions to the atmosphere (positive fluxes) or oxidation of atmospheric CH₄ (negative fluxes). In the case of negative CH₄ fluxes, these are readily distinguished in field data by a negative slope for the linear regression of CH₄ concentration vs. time. Overall, there were more than four orders of magnitude

a.



b.

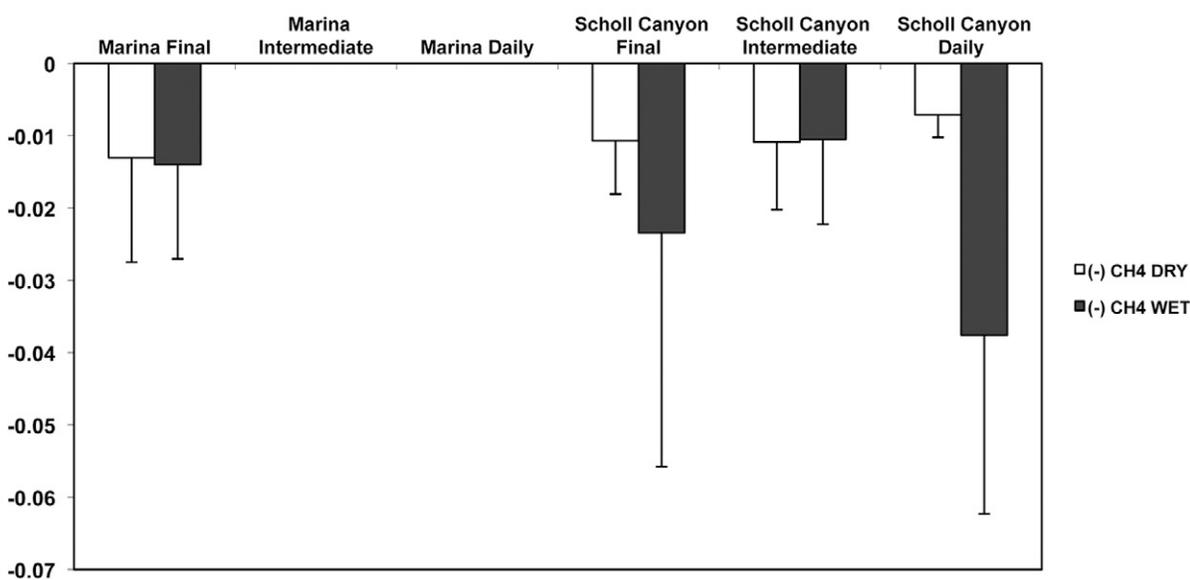


Fig. 1. Dry- and wet-season gaseous fluxes, mean values, g m⁻² d⁻¹ (+SD): (a) Positive (+) CH₄; (b) Negative (–) CH₄ (indicating uptake of atmospheric CH₄); (c) CO₂; (d) N₂O. Note: log scale for 1a and 1d. See Supplemental Table S1 for number of fluxes for each gas by cover type (e.g., where linear regression for plot of gas concentration vs. time had $r^2 > 0.9$).

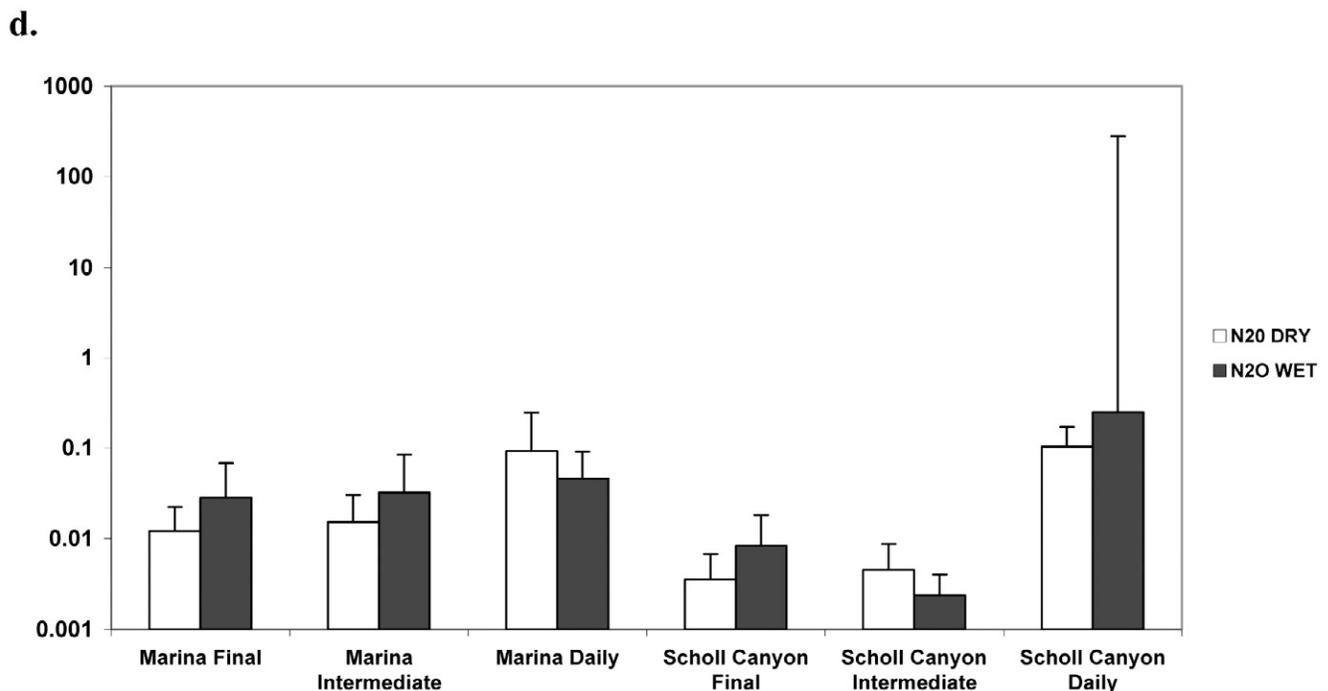
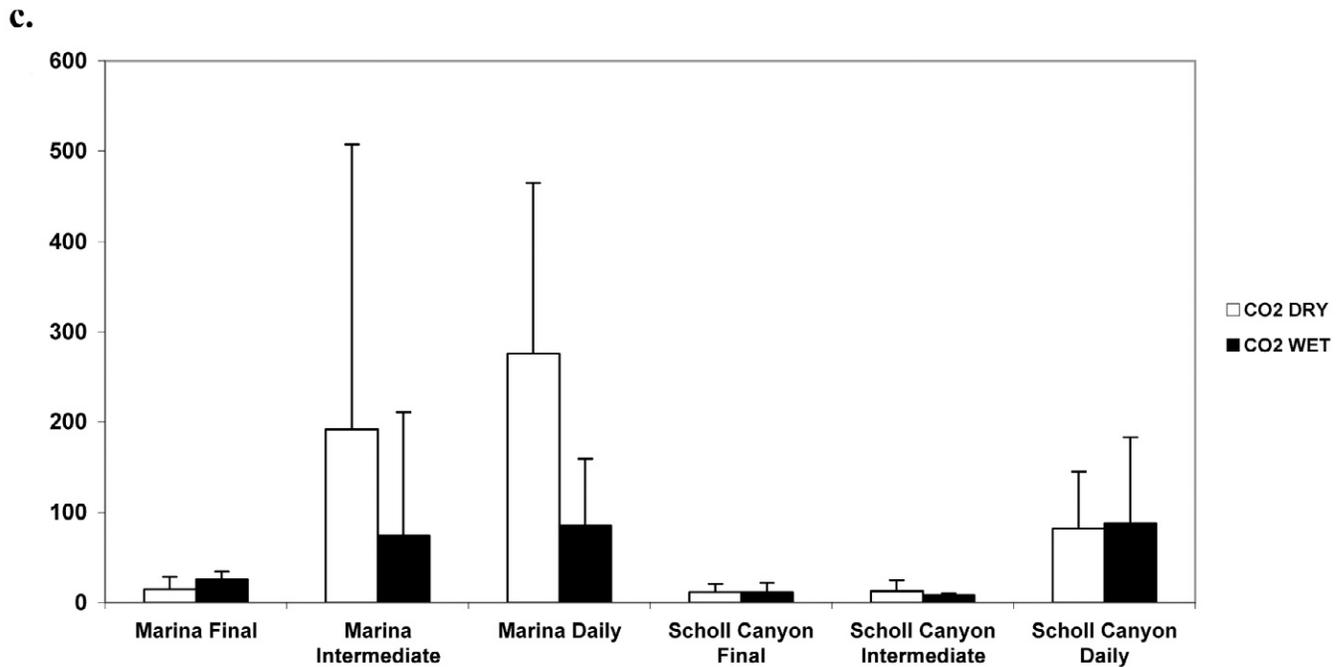


Fig. 1. Continued.

variability in the positive CH₄ fluxes and less than one order in the negative CH₄ fluxes. The maximum measured emissions in the entire study were from the Marina intermediate cover (MIC: one wet-season value of 353 g CH₄ m⁻² d⁻¹ and one dry-season value of 794 g CH₄ m⁻² d⁻¹). Average positive CH₄ fluxes for both the MIC and Marina daily cover (MDC) were significantly lower, ranging between 10 and 100 g CH₄ m⁻² d⁻¹ for both seasons. The average positive CH₄ emissions for the MDC and MIC were higher than the comparative emissions for any of the other covers (Marina final cover [MFC], Scholl daily cover [SDC], Scholl intermediate cover [SIC], Scholl final cover [SFC]) by two to four orders of magnitude. Moreover, as seen in the figures, there were no negative CH₄

emissions for the MDC and MIC for either season. Both of the thick (≥ 2 m) final cover materials (MFC and SFC) had positive CH₄ fluxes that averaged between 0.01 and 0.1 g CH₄ m⁻² d⁻¹. In general, the final cover fluxes are similar to maximum CH₄ fluxes from wetland ecosystems, which rarely exceed 0.1 g CH₄ m⁻² d⁻¹ (e.g., Jackowicz-Korczynski et al., 2010).

Addressing the negative CH₄ emissions (uptake of atmospheric CH₄), it is important to note that there were no positive CH₄ emissions for the SIC during the wet season. In general, the absolute values for the average negative CH₄ emissions for the Scholl site had similar ranges to the positive CH₄ emissions, suggesting that these soils could readily achieve oxidation rates similar to measured emission rates. As shown in Fig. 1b, the

average uptake rates ranged from about (-) 0.01 to 0.04 g CH₄ m⁻² d⁻¹. These rates are more than an order of magnitude larger than the maximum CH₄ uptake rate reported in a 3-yr study of California grasslands (0.0006 g CH₄ m⁻² d⁻¹ in Blankinship et al. [2010]), which might be considered a natural analog for California landfills which, if seeded, are typically revegetated using a grass-dominated mix. Other ecosystem studies have reported field measurements for average annual uptake on semiarid Mongolian steppe soils ranging from 0.0008 (grazed) to 0.0013 (ungrazed) g CH₄ m⁻² d⁻¹ (Chen et al., 2010), and the following values from a variety of latitudes from numerous investigations, as reported in Curry (2007) and references cited therein [all annual averages normalized to (-) g CH₄ m⁻² d⁻¹]:

Colorado grassland 0.0013 (maximum 0.0025)

Amazon pasture 0.00027, and forest 0.00085 (maximum 0.00095)

Finland forest 0.001 (maximum continental site 0.0021)

West Queensland, Australia 0.00012 (maximum 0.002)

Thus, the maximum values from nonlandfill ecosystems are generally an order of magnitude less than the average values reported for the California landfill sites (Fig. 1b). In landfill settings, the extremely wide ranges for (+) landfill CH₄ emissions and higher values for (-) emissions (uptake) reflect high rates in landfill settings for both of the competing processes of methanogenesis in the anaerobic refuse and methanotrophy in aerobic cover soils. These processes are linked by CH₄ transport affected by seasonally variable moisture and temperature. The higher negative rates compared with ecosystem studies can be largely attributed to extended exposure times under elevated CH₄ concentrations and the subsequent development of very high oxidation capacities (see discussion in Spokas and Bogner, 2011). As discussed earlier, other investigators have also observed negative CH₄ emissions from landfills as well as positive ranges spanning many orders of magnitude (Bogner et al., 1997a; Scheutz et al., 2009). Results from the two field sites in this study confirm that well-engineered and managed sites can also exhibit highly variable seasonal fluxes.

At the two California field sites, we attribute much of the variability in CH₄ emissions to variable CH₄ oxidation rates at nonoptimum temperatures and moistures. Complementary laboratory studies (Spokas and Bogner, 2011) have shown that optimum moisture potential for oxidation for these soils (at optimum temperature) was close to field capacity, with corresponding volumetric soil moisture contents ranging from 14 to 24% (v/v). During the dry season, the highest average surface soil moisture (volumetric, v/v) was 11 ± 6% for the SDC, while the average for the other cover types ranged from a low of 1 ± 3% for the MIC to a high of 8 ± 8% for the MDC. During the wet season, surface soil moistures for both daily covers and the MIC averaged 17 to 18% (v/v) ± 12 to 34, depending on cover type, while the average surface soil moisture for the MFC averaged 11 ± 7% (v/v) with much lower values of 3 ± 1% (v/v) and 5 ± 5% (v/v) for the SIC and SFC, respectively. Thus, all soils were below optimum soil moisture content during the dry season, while all except the two daily covers and the MIC were below optimum during the wet season. The optimum tempera-

tures for oxidation in these soils were 25–30°C, but average surface temperatures (0- to 5-cm depth) during the dry season exceeded the optimum for all of the Scholl cover soils, including SDC at 36.2 ± 1.2°C, SIC at 36.0 ± 11.3°C, and SFC at 35.9 ± 9.4°C (Spokas and Bogner, 2011). Also, surface temperatures ranged up to extremely high values during the dry season: SIC and SFC maxima were 62.1 and 71.0°C, respectively. Temperatures for the MIC and MFC soils averaged in the optimum range during the dry season (MIC 28.3 ± 4.4°C; MFC 25.6 ± 4.6°C). However, during the wet season, all of the Marina soils and the SIC temperatures averaged below the optimum (15.8–17.3 ± 0.1–1.1°C), while the SDC and SFC were in a more optimum range of 31.0 ± 1.7°C and 28.1 ± 7.3°C, respectively. Therefore, we conclude that none of the covers were routinely characterized by optimal temperatures and moisture contents for CH₄ oxidation during either season.

Other studies addressing landfill CH₄ oxidation have reported maximum rates of 120 to 230 g CH₄ m⁻² d⁻¹ from laboratory column studies (Kightley et al., 1995; deVisscher et al., 1999; Stein and Hettiaratchi, 2001; Pawłowska et al., 2003; Scheutz et al., 2003). In general, the rates reported for CH₄ oxidation in landfill soils are the highest in the literature (Hanson and Hanson, 1996). Previous literature documenting the percent CH₄ oxidation during transport through landfill cover soils, using the same stable carbon isotopic method used in this paper, has reported the full range of rates from negligible to 100% (Scheutz et al., 2009). Laboratory batch studies have previously reported maximum rates of 118 to 173 μg g⁻¹ dry soil h⁻¹ (Scheutz and Kjeldsen, 2004; Börjesson, 1997; Börjesson et al., 1998a,b). As discussed above, Spokas and Bogner (2011) found that a 60-d preincubation with CH₄ and adjustment of the soil moisture potential for Marina and Scholl Canyon soils to approximately the water holding capacity (33 kPa) resulted in consistent and significantly higher rates of oxidation (112–644 μg g soil⁻¹ d⁻¹), compared with the same soils at field moisture conditions, either with or without the 60-d preincubation. These are the highest rates reported in the literature to date for landfill soils and suggest that many previously published values from batch studies for landfill CH₄ oxidation “capacity” under various experimental conditions may, in fact, be underestimates of the true capacity.

For CO₂ fluxes, previously published ranges were 1 to 60 g CO₂ m⁻² d⁻¹ over an annual cycle for final cover soils at a seasonally cooler midwestern U.S. landfill (Bogner et al., 1999). The rates shown in Fig. 1c for the MFC, SFC, and SIC compare favorably to those rates. However, as shown in the same figure, both of the daily cover soils and the MIC exhibited higher rates of 50 to 250 g CO₂ m⁻² d⁻¹. For the daily covers, the rates are similar to the fresh refuse and attributable to high rates of aerobic respiration in the recently placed refuse. For the MIC, the very high CO₂ fluxes were further attributed to aerobic respiration in soils with total carbon >2%. In a recent study cited previously (Andersen et al., 2010), comparative emissions from windrow composting of garden waste were another order of magnitude higher (250–2900 g CO₂ m⁻² d⁻¹). The lower CO₂ fluxes from the other landfill soils are similar to (dark) respiration rates for California grassland soils (i.e., 8–20 g CO₂ m⁻² d⁻¹ for California sandy loam soils; Suarez and Simunek, 1998). Note that the higher CO₂ fluxes in the MDC and MIC

occurred during the dry season, suggesting CO₂ partitioning to soil moisture during the wet season and possibly increased flux of landfill gas CO₂ during dry periods.

Average N₂O fluxes (Fig. 1d) varied by almost two orders of magnitude with the highest values associated with the daily cover materials, which is attributable to the combination of high moisture content, available N, and restricted aeration in the recently placed refuse. Both the MIC and MFC exhibited higher N₂O fluxes than the SIC and SFC, which we attribute to the increased availability of N in the biosolids-amended Marina cover materials. Previous literature for N₂O fluxes from final cover soils has shown that a zone of high N₂O production can occur at mid- to bottom depths in thicker final cover soils (Bogner et al., 1999), as well as from final cover soils amended with high-N source materials such as biosolids (Börjesson and Svensson, 1997) or from “semi-aerobic” landfills (Tsujiimoto et al., 1994). Maximum values for N₂O fluxes from windrow composting operations (approximately 1–8 g m⁻² d⁻¹; Beck-Friis et al., 2000; Andersen et al., 2010) were an order of magnitude higher than the average N₂O emissions for the MDC and SDC (approximately 0.1 g m⁻² d⁻¹) but within the high variability for SDC shown in Fig. 1d. Overall, N₂O fluxes from ecosystems can vary by four to five orders of magnitude, depending on fertilization/available N, seasonal moisture, aeration status, and other variables, with the highest values approaching 1 g N₂O m⁻² d⁻¹ (Hellebrand et al., 2008; Rochette et al., 2008; Du et al., 2008; Livesley et al., 2008) and typically associated with abundant available N, restricted aeration, and higher water contents below full saturation. Similarly, the overall range shown in Fig. 1d is about five orders of magnitude with average values that vary by about two orders of magnitude.

There were generally no simple relationships between observed fluxes and average soil temperatures or moisture contents. During the dry season, the Scholl Canyon surface soils exhibited an immense temperature range (22.1–71.0°C), while the Marina soils had a smaller range of 14.1 to 48.0°C. For both soils, temperatures >40°C during the dry season resulted in severely reduced rates of CH₄ oxidation. During the wet season, the Marina soils and the SIC exhibited little temperature variability (overall range 15.7–18.9°C), while the SDC had higher variability (29.1–33.6°C) and the SFC had a much wider range of 14.2 to 35.2°C. Simple plots of gaseous fluxes vs. soil moisture did not typically indicate statistically significant relationships; however, the average wet-season N₂O fluxes

(g m⁻² d⁻¹) were exponentially related to average soil moisture (v/v) across all cover types (data not shown; $y = 0.0019e^{20.93x}$, $r^2 = 0.79$). Although the vast literature on N₂O emissions from agricultural and other ecosystems generally indicates increased N₂O production and emissions at higher water-filled pore space below full saturation, this relationship is complex and intimately related to soil physical and biochemical properties (pore volume/geometry, N sources, relative N₂O production/transport/consumption rates; e.g., Parton et al., 2001).

Geospatially Averaged Emissions

Areal emissions were determined using an inverse distance weighting procedure recommended for areas characterized by heterogeneous sources (Journal and Huijbregts, 1981). Table 2 compares the geospatially determined CH₄ emissions to the average (+) CH₄ emissions values shown previously in Fig. 1a. In general, the geospatially determined CH₄ fluxes for all covers are lower than the (+) CH₄ emissions values. Table 2 also indicates the percentage of each cover type with negative (–) CH₄ fluxes, indicating uptake of atmospheric CH₄. Most values range between about 10 and 40%, depending on cover type and season, reinforcing the variability in oxidation discussed above. It is important to stress that four of the six cover types had significant percentages of negative (–) CH₄ fluxes. Also note the wide range, from negligible percentages for MDC and MIC to 100% for SIC during the 2007 wet season. Using recommended criteria (Journal and Huijbregts, 1981; Spokas et al., 2003), the structure of the CO₂ and N₂O emissions data sets did not permit use of a standard geostatistical technique.

Methane Oxidation and Secondary Source of Methane in Cover Soils

While primarily relying on the conservative isotopic method discussed above using chamber samples to derive the percent oxidation, we also selectively determined percent oxidation using the δ¹³C CH₄ for soil gas probe samples (see Supplemental Information). The probe samples provide a less conservative (higher) value for oxidation (Chanton et al., 2008; Cabral et al., 2010). Because of the relatively high percentages of negative CH₄ fluxes (Fig. 1b), where the isotopic method for percent CH₄ oxidation using chamber samples cannot be applied, the probe data also provided a secondary method for quantification of CH₄ oxidation using isotopic fractionation.

Table 2. Geospatial CH₄ fluxes (using inverse distance weighting [IDW] with significance test) for individual field campaigns and percentage (%) of surface area with negative (–) CH₄ fluxes using IDW at Marina and Scholl Canyon landfills, California.

Date	Marina			Scholl Canyon		
	Final	Intermediate	Daily	Final	Intermediate	Daily
Geospatial CH₄ fluxes (mg m⁻² d⁻¹)						
March 2007	3.31	32.04	2.09E+02	3.31	–6.28	6.41
August 2007	2.06	5.32E+04	5.64E+02	3.86	2.26	15.07
March 2008	7.35	3.42E+04	1.02E+04	7.90	12.94	19.30
August 2008	99.43	2.38E+05	8.86E+03	–0.80	–3.18	22.27
Percentage (%) of surface area with negative (geospatial) CH₄ flux						
March 2007	21	0	0	21	100	<1
August 2007	42	0	0	5	38	0
March 2008	8	0	0	40	3	21
August 2008	2	0	0	24	41	<1

In addition, we also selectively determined the $\delta^{13}\text{C CO}_2$ for soil gas probe samples at both sites which, with the $\delta^{13}\text{C CH}_4$, can assist with determining other potential biogenic sources whose isotopic signature would differ from the source gas and the oxidized source gas.

Table 3 summarizes the percentage (%) CH_4 oxidation in cover soils at both sites using the isotopic technique applied to chamber and probe data; these data were separately composited for each site as the large data ranges did not support seasonal discrimination. The first two rows in the table summarize percent oxidation using chamber data for the positive CH_4 fluxes at both sites. While the two sites have almost identical minimums, the higher maximum and larger number of high values for Scholl Canyon elevated the average value to 52%, compared with 30% for Marina. For probe samples, however, also shown in Table 3, the average percent oxidation at Marina was almost 10% higher than for the chamber-based data, which is consistent with previous literature reporting higher values using probe data (Chanton et al., 2008). However, for Scholl Canyon, because of the inclusion of deeper probe samples with more limited diffusion of atmospheric O_2 , the average percent oxidation using probe data was lower than the Marina percentage. In general, the Scholl field data indicated a much wider range for percent oxidation than the Marina data. Clearly, improved elucidation of oxidation dynamics with systematic sampling through profile sequences in field settings would be a priority for future research.

Figure 2 compares the $\delta^{13}\text{C CO}_2$ to the $\delta^{13}\text{C CH}_4$ for the source gas and selected probe samples from both sites, including MDC (25 cm), MIC (12–43 cm), SDC (10 cm), and SFC (30–243 cm). The Marina well gas averaged $\delta^{13}\text{C CH}_4$ -58.37 (± 1.239) while the Scholl Canyon well gas averaged $\delta^{13}\text{C CH}_4$ -60.07 (± 0.6602). All data shown in the figure are from the two wet-season campaigns where we were investigating the potential for shallow secondary production of CH_4 in cover materials. The circled area in this figure indicates tightly clustered values for the composite wellfield source gas ($\delta^{13}\text{C CH}_4$ between -58 and -60 ; $\delta^{13}\text{C CO}_2$ of 10 to 20). The majority of the probe data indicate that CH_4 oxidation has occurred, which explains the data plotted in the right triangle in this figure. However, some probe values from the MDC, MIC, and SDC had highly negative $\delta^{13}\text{C CH}_4$ values and plotted in a separate field in this figure, namely the oval in the lower left. This field indicates that some CH_4 is being derived from a secondary source of biogenic CH_4 . Moreover, in further support of this hypothesis, other chamber and probe samples, for which the $\delta^{13}\text{C CO}_2$ was not determined, also had $\delta^{13}\text{C CH}_4$ in the range for the secondary source and not in the range for either the source gas or oxidized gas (data not shown). The highly negative $\delta^{13}\text{C CH}_4$ values for this secondary source are indicative of biogenic CH_4 production via H_2 reduction with CO_2 (Hackley et al., 1999). Thus, on the basis of the isotopic data, we conclude that shallow secondary production of CH_4 was seasonally occurring in cover soils at both sites, which further complicates CH_4 generation, transport, and oxidation dynamics in these soils. This secondary production of CH_4 in cover soils is likely a tran-

Table 3. Percentage (%) CH_4 oxidation at Marina and Scholl Canyon landfills, California, based on $\delta^{13}\text{C CH}_4$ of chamber and probe samples compared with source gas from landfill gas collection system. Number in parentheses (*n*) indicates number of values.

Location	Avg. % CH_4 oxidation (<i>n</i>)	SD	Min.	Max.
Chamberst				
Marina	30.0 (37)	12.0	10.4	42.0
Scholl Canyon	51.7 (6)	44.5	10.6	100.0
Probes‡				
Marina: daily and intermediate cover (10–50 cm)	39.7 (46)	21.6	0.9	81.4
Scholl: final cover (30–244 cm)	47.6 (33)	9.2	39.1	73.3

† Data composited for all cover types at each site, including all chamber data where isotopic technique could be applied (nonnegative CH_4 fluxes). Many negative and low positive CH_4 fluxes for Scholl Canyon precluded use of this technique on a larger number of samples.

‡ Includes all probe data where there was isotopic fractionation relative to source gas. The number of soil gas probe samples ≥ 25 cm was 20 for Marina and 15 for Scholl Canyon. All probe data were composited. See Supplemental Information for additional details regarding the methodology.

sient phenomenon with high spatial and temporal variability, which requires more detailed investigation in future studies.

Conclusions and Modeling Implications

This is the first study that has systematically investigated seasonal greenhouse gas emissions from fresh refuse (no cover) and the full range of cover materials at the same landfill sites. The high variability with respect to CH_4 emissions among and between the various cover materials indicates that landfill CH_4 emissions modeling at field scale for various purposes, including improved emissions inventories, requires the incorporation of critical site-specific management and soil microclimate variables not included in previous models (see discussion in Scheutz et al., 2009; Bogner and Spokas, 2011). In particular, seasonal CH_4 transport, oxidation, and “net” emission rates in cover soils are dependent not only on-site management deci-

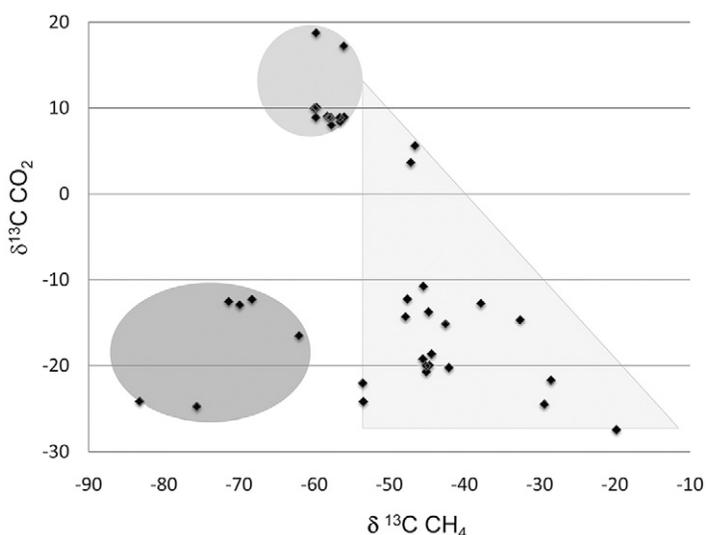


Fig. 2. Comparison of $\delta^{13}\text{C CO}_2$ and $\delta^{13}\text{C CH}_4$ for three differing isotopic signatures: unoxidized source gas from landfill gas recovery system (top circle), oxidized landfill gas collected in chambers and from soil gas probes in cover (right triangle), and a secondary source of unoxidized biogenic CH_4 (bottom oval).

sions (e.g., implementation of engineered gas recovery and cover thickness, texture, and density), but also on seasonal soil moisture and temperature variability over a typical annual cycle. Importantly, some cover materials are characterized by a high percentage of negative (–) CH₄ fluxes, indicating uptake of atmospheric CH₄ due to high seasonal capacities for CH₄ oxidation. Moreover, as indicated by the isotopic data, secondary production of CH₄ in cover soils is also possible, especially during the wet season. Average CO₂ emissions generally varied within the same order of magnitude across cover types. For fresh refuse and daily cover CO₂ fluxes, these values were consistent with (but lower than) emissions from aerobic composting of organic waste fractions. For intermediate and final covers, the CO₂ fluxes were consistent with dark respiration rates for grassland ecosystems. The N₂O emissions were similar to ecosystem ranges and consistent with a very limited number of previous studies where the highest rates were associated with biosolids-amended cover materials.

Based on the results of this study, one must call into question the routine use of existing regulatory and inventory models for landfill CH₄ emissions (e.g., IPCC, 2006) that rely solely on a hypothetical first-order kinetic model for CH₄ generation, but do not take into consideration either the properties of the cover materials or seasonal CH₄ oxidation in those cover soils. In limited field-scale comparisons to date (Spokas et al., 2006; also Fig. 2 in Bogner and Spokas, 2010; for well-monitored cells with detailed waste input information), the first-order kinetic models for theoretical CH₄ generation were shown to be good predictors for CH₄ recovery, but were extremely poor predictors for residual CH₄ emissions at sites which, like the majority of sites in California, have engineered gas extraction systems. Moreover, existing IPCC inventory methods (IPCC, 2006) typically allow only a 10% reduction in emissions due to CH₄ oxidation (based solely on one New Hampshire, USA, field site without gas recovery, as reported by Czepiel et al., 1996). Both a recent literature review (Chanton et al., 2009) and the field data reported in this study support a higher value for percent oxidation, even though the California temperature and moisture conditions were suboptimal over much of the year. In related work, we discuss the development, structure, and field validation of a new JAVA-based landfill CH₄ inventory model inclusive of seasonal oxidation for California landfill sites (Spokas et al., 2009; Bogner and Spokas, 2010; Spokas and Bogner, unpublished data, 2011). This model (CALMIM, for CA Landfill Methane Inventory Model) estimates site-specific landfill CH₄ emissions inclusive of seasonal CH₄ oxidation through any layered landfill cover soil at 2.5-cm depth increments and 10-min time increments, which are then summed to provide typical annual emissions. The model incorporates globally validated USDA models for climatology and soil moisture/temperature at the 0.5 by 0.5-degree latitude–longitude scale (global RainSIM, global TempSIM, SOLARCALC, and STM²) and couples those models to site-specific cover materials, extent of engineered gas extraction, and a one-dimensional model for diffusive CH₄ flux inclusive of oxidation. For diffusive flux, the driving force is the CH₄ gradient through the cover materials, which is a function of the type of cover (daily, intermediate, final), the extent of gas extraction under the cover materials, and methanotrophic oxida-

tion in the cover. Oxidation is scaled to maximum rates over the full range of annual temperature and moisture conditions as discussed in Spokas and Bogner (2011). Field validation for this model included the CH₄ emissions data discussed in this paper, as well as field data for CH₄ emissions and oxidation at three additional California sites.

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