ELEMENTA
Science of the Anthropocene

Cambaliza, MOL, et al 2017 Field measurements and modeling to resolve m^2 to km^2 CH_4 emissions for a complex urban source: An Indiana landfill study. *Elem Sci Anth,* 5: 36, DOI: https://doi.org/10.1525/elementa.145

RESEARCH ARTICLE

Field measurements and modeling to resolve m^2 to km^2 CH_4 emissions for a complex urban source: An Indiana landfill study

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Large spatial and temporal uncertainties for landfill CH_4 emissions remain unresolved by short-term field campaigns and historic greenhouse gas (GHG) inventory models. Using four field methods (aircraft-based mass balance, tracer correlation, vertical radial plume mapping, static chambers) and a new field-validated process-based model (California Landfill Methane Inventory Model, CALMIM 5.4), we investigated the total CH_4 emissions from a central Indiana landfill as well as the partitioned emissions inclusive of methanotrophic oxidation for the various cover soils at the site. We observed close agreement between whole site emissions derived from the tracer correlation (8 to 13 mol s^{-1}) and the aircraft mass balance approaches (7 and 17 mol s^{-1}) that were statistically indistinguishable from the modeling result (12 ± 2 mol s^{-1} inclusive of oxidation). Our model calculations indicated that approximately 90% of the annual average CH_4 emissions (11 ± 1 mol s^{-1}; 2200 ± 250 g m^{-2} d^{-1}) derived from the small daily operational area. Characterized by a thin overnight soil cover directly overlying a thick sequence of older methanogenic waste without biogas recovery, this area constitutes only 2% of the 0.7 km^2 total waste footprint area. Because this Indiana landfill is an upwind source for Indianapolis, USA, the resolution of m^2 to km^2 scale emissions at various temporal scales contributes to improved regional inventories relevant for addressing GHG mitigation strategies. Finally, our comparison of measured to reported CH_4 emissions under the US EPA National GHG Reporting program suggests the need to revisit the current IPCC (2006) GHG inventory methodology based on CH_4 generation modeling. The reasonable prediction of emissions at individual U.S. landfills requires incorporation of both cover-specific landfill climate modeling (e.g., soil temperature/moisture variability over a typical annual cycle driving CH_4 transport and oxidation rates) as well as operational issues (e.g., cover thickness/properties, extent of biogas recovery).

Keywords: landfill; methane emissions; aircraft-based mass balance; tracer correlation approach; CALMIM; methanogenic oxidation

Introduction

Atmospheric methane (CH_4) is now at its highest level during the last 800,000 years and, including interactions with ozone and water vapor, is responsible for 21% of the 2.3 W m^{-2} total positive radiative forcing since 1750 (IPCC, 2013). Despite more than three decades of literature addressing CH_4 emissions, our understanding of the regional magnitude and variability of emissions from multiple area and point sources remains relatively poor due to complex urban patchworks of industrial, energy, and waste management sources. Currently, with improved instrumentation choices and field measurement techniques during the last decade (Abichou et al., 2010, 2012; Babilotte et al., 2010; Cambaliza et al.,

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2014, 2015; Foster-Wittig et al., 2015; Goldsmith et al., 2012; Hashmonay et al., 2001; Mays et al., 2009; Mønster et al., 2014), we have tools to more quantitatively address diurnal, seasonal, and annual CH_4 emissions from specific sources. Realistically, considering the strengths, limitations, and uncertainties of each method, unraveling the temporal and spatial variability of emissions over a typical annual cycle from complex area sources can benefit from multiple techniques deployed simultaneously. Importantly, field-validated models are also needed to provide a framework for the expected annual cycle of emissions at each site, including unique site-specific dependencies on climate and management factors.

In this study, we quantify the CH_4 emissions from a landfill in central Indiana USA, using four field techniques and the CAlifornia Landfill Methane Inventory Model (CALMIM 5.4) (Spokas et al., 2011), which is a new field-validated, process-based model. The landfill site is about 30 km west of the Indianapolis city center, and an upwind CH_4 source to the city. Thus the current study also provides critical input to the accurate determination of Indianapolis CH_4 emissions, an important goal of the Indianapolis Flux Experiment project (INFLUX, http://sites.psu.edu/influx/; Cambaliza et al., 2014). The landfill site includes five adjacent areas with highly variable CH_4 emissions due to differing cover materials and gas management strategies. We applied multiple techniques to quantify CH_4 emissions at various temporal and spatial scales typical for closely-spaced urban CH_4 sources. Our goal was to improve understanding of temporal variability within the constraints of technique-specific limitations and uncertainties. The spatial scale of field measurements ranged from small static chambers ($<m^2$) to an aircraft-based mass balance technique ($>km^2$), with temporal scales ranging from hourly to monthly. As with other soil sources of CH_4 with large seasonal variability due to soil oxidation, the annual cycle of emissions for each cover area is challenging to quantify due to small-scale variability with rates ranging over several orders of magnitude (Scheutz, et al., 2009). Moreover, larger-scale field techniques (e.g., dynamic tracer techniques, tower-based and aircraft-based approaches) can struggle to quantify temporal variability for individual sources within a complex patchwork. Because it is rarely possible to deploy multiple techniques at large numbers of diverse sites, we also suggest that the lessons learned may be applicable to other closely-spaced agricultural, mining, energy, industrial and waste-related area sources of atmospheric CH_4. Such basic understanding is essential for developing improved urban-scale greenhouse gas (GHG) inventories to guide local and regional policymakers formulating GHG mitigation strategies.

Our specific objectives for this study were to: (1) quantify the spatial and temporal variability of emissions via direct field measurements using multiple techniques; (2) use modeling to project monthly average emissions with and without oxidation over a "typical annual cycle"; (3) identify the highest-emitting areas of the landfill footprint; and (4) address remaining questions and mitigation measures to reduce observed emissions.

Methods

Site description and summary of methods

The central Indiana landfill (IN-1) (**Figure 1**, located at 39.76679°N, 86.53031°W), has a total surface area of 0.702 km^2. At the time of our field investigations, the site was composed of the following areas with differing emissions signatures (**Figure 1** and Table S1, supplementary information): (1) a small working area with a very thin soil "daily cover" placed at the end of the working day, (2) an

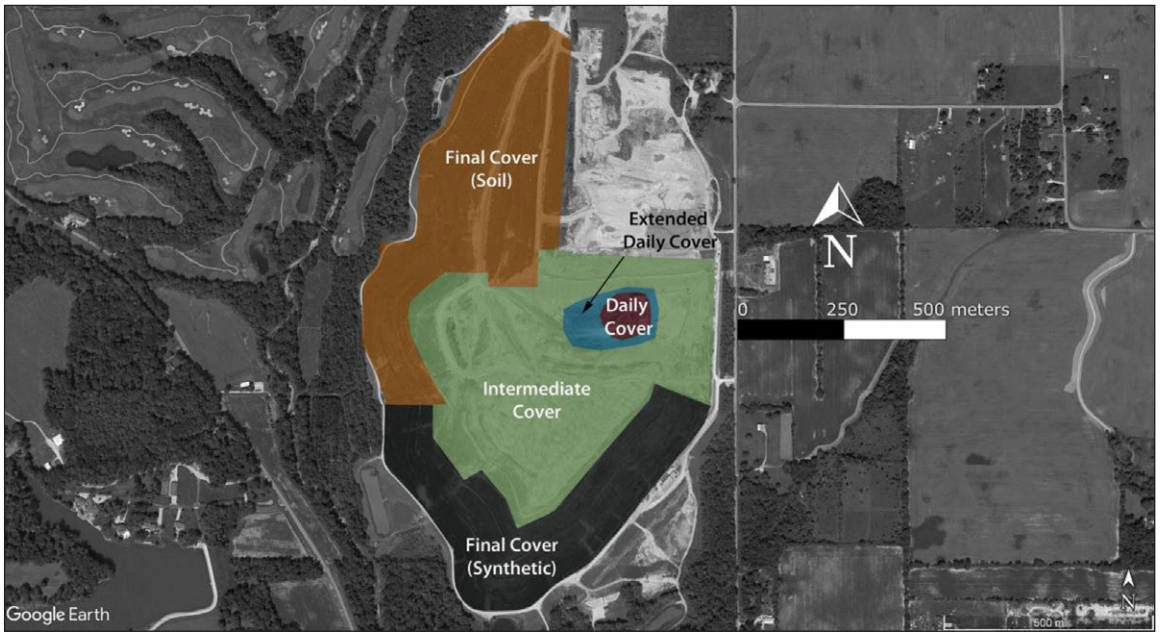

Figure 1: IN-1 landfill site in Indiana, USA. Aerial image (via Google Earth) of the IN-1 solid waste facility showing the various cover areas. The properties of the landfill cover types are provided in Table S1 in the supplementary information. DOI: https://doi.org/10.1525/elementa.145.f1

adjacent area with thicker longer-term "extended" daily cover soil, (3) a large area of thick intermediate cover soil where future vertical expansion of the landfill is planned, (4) an area which has reached final grade and has a very thick final soil cover with regulatory approval; and (5) a second area at final grade where the final cover has an approved regulatory design inclusive of a geomembrane. This wide variety of cover areas is typical for U.S. landfill sites. Detailed information for the various landfill cover areas is provided in the supplementary information. For IN-1, engineered gas recovery exists for areas (3) – (5). We examined the CH_4 emissions using two whole site measurement approaches (Aircraft Mass Balance and Tracer Correlation approaches) as well as two cover-specific measurement techniques (Static Closed Chamber method and Vertical Radial Plume Mapping). **Table 1** summarizes the field measurement approaches and dates for whole landfill and specific cover area campaigns. We also compared measured emissions to the 5-year record of reported emissions for this site under the U.S. EPA National Greenhouse Gas (GHG) Reporting Program. That program relies on the IPCC (2006) national GHG methodology for landfill CH_4 emissions, which is based on a calculation for total biogas generation from the buried waste in a given year, a variable "% collection efficiency" which is applied where engineered gas recovery exists under differing cover soils, and a variable allowance for methanotrophic oxidation (0–35% depending on cover materials).

Aircraft-based mass balance experiments

The aircraft-based mass balance (AMB) approach for quantifying the emissions of area and point sources relies on several horizontal transects flown at various altitudes within the convective boundary layer downwind from the source (Cambaliza et al., 2014, 2015; Mays et al., 2009). By flying perpendicular to the prevailing wind direction at ~16-km long horizontal transects, the IN-1 CH_4 plume is intercepted, and quantified across the crosswind two-dimensional plane.

Two field campaigns were conducted at IN-1. During the 03 July 2014 campaign, the wind direction was NNW (340° ± 18°, 1σ) with a magnitude of 6.2 ± 2.0 (1σ) ms^{-1}. Horizontal transects were flown at 4.5-km and 6-km downwind distances from the site. During the 30 August 2012 field measurement, the wind direction was SE (140° ± 22°, 1σ) with a magnitude of 5.3 ± 1.8 (1σ) m s^{-1}. Horizontal transects were flown at 3-km and 6-km downwind. We note that the results for the 30 August 2012 flight experiment have been previously reported by Cambaliza et al. (2014, 2015) and used together with small area flight measurements to determine the precision of the AMB approach. The flight paths for the two experimental days are shown in Figures S1 and S2 in the supplementary information.

Concentrations of CH_4, carbon dioxide (CO_2), and water vapor (H_2O) were measured at 0.5 Hz using a cavity ring-down spectrometer (CRDS, Picarro Inc. Santa Clara, CA, USA; model G2301-f) (Chen et al., 2010; Crosson, 2008; Karion et al., 2013). We performed inflight calibrations for CO_2 and CH_4 using three NOAA/ESRL calibration cylinders with the following mole fractions that span the expected mixing ratios from emissions sources: 378.49, 408.83, and 438.29 ppm for CO_2, and 1803.0, 2222.2, and 2599.5 ppb for CH_4 (Cambaliza et al., 2014, 2015). The measurement precision for CO_2 and CH_4 at 0.5 Hz during in-flight calibrations was 0.1 ppm (1σ) and 2.6 ppb (1σ), respectively (Cambaliza et al., 2014). Wind vectors were measured at 50 Hz using a Best Air Turbulence (BAT) probe, a nine-port pressure probe extending from the nose of the aircraft (Garman et al., 2006, 2008). The measured pressure differentials across the probe are combined with 50 Hz inertial data from the GPS/INS system to obtain the three-dimensional wind fields. Located in the center of the hemispherical probe is a sensor for ambient temperature measurements. The measured horizontal distributions of CH_4, pressure, temperature, and perpendicular wind speeds were interpolated across the two-dimensional planes using a kriging approach (Chu, 2004), and the net molecular concentration (mol m^{-3}) across each grid cell is determined using the ideal gas law and the interpolated pressure and temperature. The background CH_4 concentration (Table S2, supplementary information) was obtained from the edges of the gridded two-dimensional matrix outside the landfill boundary as described in Cambaliza et al. (2014). The background CH_4 mole fractions were essentially constant and indistinguishable for the two downwind distances of the 30 August 2012 (1930.6 ± 8.6 ppb and 1923.6 ± 7.8 ppb for the 3 and 6 km, respectively) and 03 July 2014 flight experiments (1896.3 ± 1.2 ppb and 1896.2 ± 1.3 ppb for the

Table 1: Summary of measurements conducted on various IN-1 landfill cover areas using the different measurement approaches: Tracer Correlation Approach (TCA), Aircraft Mass Balance (AMB) Approach, Vertical Radial Plume Mapping (VRPM), and Static Closed Chamber. DOI: https://doi.org/10.1525/elementa.145.t1

Measurement Technique	Measurement Dates	Whole Site	Daily Cover[a]	Intermediate Cover	Final Cover
TCA	2009 – 2012[b]	X			
AMB	30 Aug 2012, 03 July 2014	X			
VRPM	Mar, May 2009			X	X
Static Closed Chamber	May, Aug 2012		X	X	

[a]"Extended" (longer term) daily cover area only. See text.

[b]Exact dates and number of TCA measurements are shown in Table S3 in the supplementary information.

4.5 and 6 km, respectively). The emissions rate (mol s^{-1}) is determined by integrating the flux elements across the gridded two-dimensional plane, using the measured wind speed perpendicular to the plane and the difference between the gridded and background CH_4 concentration. The calculated single-measurement precision of this AMB approach for quantifying emissions of relatively small area sources but with large source strengths (e.g. landfills and power plants) is ±30% (Cambaliza et al., 2014). Figures S3 and S4 (supplementary information) show the measured and interpolated CH_4 data for the flight experiments in 2012 and 2014, respectively.

Tracer correlation approach

The tracer correlation approach (TCA) quantifies whole site CH_4 emissions by releasing a tracer gas at a known emissions rate. Enhancements of CH_4 and the tracer gas above background are simultaneously measured downwind of the source where the two gases are observed to be well-mixed (Babilotte et al., 2010; Czepiel et al., 2003; Foster-Wittig et al., 2015; Galle et al., 2001; Mønster et al., 2014; Mosher et al., 1999). Each TCA measurement at the IN-1 site made use of 2–3 cylinders of atomic absorption grade acetylene (C_2H_2, 99.6% purity) using either a triangular or a linear release geometry from the flat area at the top of the landfill. The cylinders were typically spread out so that the release points maximize the landfill top coverage. The C_2H_2 release rate was 20 L min^{-1} with a total release time of approximately 3 hours (Foster-Wittig et al., 2015). Concentrations of CH_4 and C_2H_2 were simultaneously measured at a sample flow rate of 2 L min^{-1} (Picarro CRDS Model G1203; Picarro Inc. Santa Clara, CA, USA) with typical field precisions of 0.088 ppb and 0.463 ppb for C_2H_2 and CH_4, respectively, at the data acquisition frequency of 0.5 Hz (see supplemental information of Foster-Wittig et al., 2015). The accuracy of the CRDS mole fraction measurements were determined using periodic comparisons with gas standards (Linde Gas USA, OH, USA) at 2 ppm CH_4 and 100 ppb C_2H_2 levels during in-field check procedures prior to trace gas release (Foster-Wittig et al., 2015 supplementary information). The performance of the CRDS was stable to within ±10% accuracy check. The CRDS, control computer, and calibration system were housed on a mobile platform (GMC Sierra 4 × 4 pickup truck) fitted with a vibration isolated instrument rack. Wind speed, wind direction, and atmospheric turbulence properties were monitored on the landfill using a three-dimensional (3D) sonic anemometer mounted 3-m above ground level. Measurements of CH_4 and C_2H_2 were conducted at sufficient distances downwind of the source to allow the respective plumes to become well-mixed. Thus, at sufficient downwind distances, the multiple C_2H_2 point sources that span the landfill appear to be one large area source with the same width as the landfill, and the widths of CH_4 and C_2H_2 plumes are identical. For their study spanning 15 sites in 8 states, Foster-Wittig et al. (2015) found that the separation distance between the downwind observation location at the plume center and the centroid of the C_2H_2 tracer release geometry was on average 1.82 ± 0.63 km (1 σ) and ranged from 0.62 to 5.4 km to achieve acceptable transects with well-mixed plumes.

The whole-site CH_4 emissions rate was calculated using both (1) the unconstrained slope of the best-fit line between CH_4 and C_2H_2 concentrations, and (2) the ratio of the time-integrated areas of CH_4 and C_2H_2, multiplied by the tracer release rate and the ratio of the molecular weights of CH_4 and C_2H_2 (Foster-Wittig et al., 2015). Detailed description of the two methods is discussed in the supplementary information. TCA data were collected during multiple campaigns from 2009–2012, resulting in a total of 94 data points that satisfy the (80, 20) criteria discussed by Foster-Wittig et al. (2015), i.e., the R^2 correlation coefficient of the CH_4 vs C_2H_2 best fit line is greater than 0.80 and the percentage difference between the emissions rates estimated from the two methods is less than 20%. Herein we report CH_4 emissions from the plume integration ratio approach (method 2 above).

CH_4 emissions measurements from specific cover areas

U.S. landfills are highly-engineered and include daily, intermediate, and final cover areas with distinctive CH_4 emissions signatures due to differing thickness and physical characteristics, temporal depth-dependent changes in soil moisture and temperature, and the variable extent of engineered gas recovery. As discussed below, both static closed chambers and vertical radial plume mapping (VRPM) were deployed to quantify emissions from the extended daily and intermediate cover areas expected to be significant contributors, on a unit area basis, to the total site emissions (Bogner et al., 2011).

Static closed chamber methods. CH_4 emissions from the intermediate cover (May 2012) and longer-term daily cover (August 2012) were determined using a static closed chamber technique (Bogner et al., 2011; Spokas and Bogner, 2011). The stainless steel chambers consisted of two parts: 1) a cylindrical base with a beveled lower edge to push into the cover soil and an upper trough filled with distilled water; and 2) a hemispheric chamber placed into the trough and secured with hand clamps. During short monitoring periods (<30 min), 5-ml timed samples were withdrawn from a septum port at the top of each chamber and injected into autosampler vials (Agilent) flushed with He. In addition, soil gas CH_4, CO_2, O_2, and N_2 soil gas concentration profiles were determined using additional 5 mL samples collected by syringe from a portable soil gas probe (U.S. Geological Survey) and also injected into autosampler vials. The probe had a removable Ultra-Torr top fitting (Swagelok) with a septum port and was purged between samples at successive depths. During May 2012, at "near-well" and "between-well" locations, chamber fluxes and soil gas profiles were determined for an intermediate cover area with vertical gas extraction wells, using a stratified random sampling design (Bogner et al., 2011; Spokas and Bogner, 2011). In August 2012, emissions and soil gas profiles were determined for the longer term "extended" daily cover area immediately adjacent to the current filling area (**Figure 1**). Each of 6 randomized soil gas profiles was paired with 3 static chamber fluxes, and additional randomized soil gas profiles and soil gas concentration profiles were also completed. Three of the static chambers

were replicated four times over an 18-hour period. All gas samples were sent to the USDA-ARS laboratories (in St. Paul, MN) for analysis of CH_4 and other gases (CO_2, N_2O) using gas chromatography (GC) and GC with mass spectrometry (GC/MS) instrumentation as described in previous publications (Bogner et al., 2011; Spokas et al., 2011; Spokas and Bogner, 2011). Only the CH_4 results are discussed herein; minimum detectable flux for CH_4 was 12 mg m^{-2} d^{-1}.

Vertical radial plume mapping. The vertical radial plume mapping (VRPM) method makes use of an optical remote sensing instrument (ORSI) to quantify the path integrated CH_4 concentrations in a perpendicular plane downwind of the source at five radial distances (Abichou et al., 2010, 2012; Goldsmith et al., 2012; Hashmonay et al., 2001). The advected mass emissions rate (g min^{-1}) is determined by multiplying the component of the local wind normal to the vertical plane with the integrated CH_4 enhancements. For VRPM measurements conducted at IN-1, the ORSI was a tunable diode laser absorption spectrometer (TDLAS). The VRPM emissions shown here were measured from the intermediate and final cover areas in March and May 2009 and were previously reported by Goldsmith et al. (2012). The sources of uncertainties in the VRPM method were initially examined by Abichou et al. (2010). While the degree of uncertainty of the VRPM-determined emissions is still not well-defined, Abichou et al. (2010) found that a variability of ±30° in the winds perpendicular to the VRPM plane can lead to a 20% uncertainty in the flux while the various methods to estimate the landfill area contributing to the surface flux can introduce an additional uncertainty of 10% to 30% depending on the stability class.

CH_4 emissions modeling using CALMIM 5.4 model

CALMIM is a site-specific inventory model for annual landfill CH_4 emissions which incorporates soil profile properties and microclimate modeling coupled to globally-validated 0.5° scale climate models (Global TempSIM; Global RainSIM; SOLARCALC; STM^2 Soil Temperature/Moisture; Spokas et al., 2011). Based on 1-D diffusion for CH_4 and O_2 at 2.5-cm depth increments and 10-min time-steps, CALMIM is a freely available JAVA tool that models a typical annual cycle for CH_4 emissions from site-specific daily, intermediate, and final landfill covers at any landfill site worldwide. (http://www.ars.usda.gov/services/software/download.htm?softwareid=300). CH_4 oxidation is scaled to maximum rates based on extensive supporting laboratory studies that quantified rate dependencies on soil temperature and moisture potential (Spokas and Bogner, 2011). CALMIM also includes a subtraction for O_2 demand for normal soil respiration (Spokas et al., 2011). Compliant with IPCC "Tier III" criteria, CALMIM was originally developed for application to an improved GHG inventory for California with intensive field-validation at two California sites (Monterey County and Los Angeles County) and limited field validation at three additional California sites (Bogner et al., 2011; Spokas et al., 2011). During 2011–2014, multiple programmatic and structural improvements were made, including multi-platform capabilities (PC, MAC, UNIX). An independent international field validation was completed using 40 covers at 29 sites located on 6 continents (Bogner et al., 2014) and CALMIM was subsequently applied to a new California inventory for landfill CH_4 emissions (Spokas et al., 2015). Table S1 summarizes major CALMIM inputs for the various cover soils at the IN-1 site.

CH_4 emissions modeling based on IPCC (2006) under current U.S. EPA Mandatory Reporting Rule (MRR)

The current IPCC (2006) national GHG inventory methodology for landfill CH_4 relies on a first order kinetic model where the biogas generation rate is assumed to peak in the year of waste placement and decline thereafter. The annual rate of biogas generation is estimated from the degradable organic carbon content of individual waste components (IPCC, 1996, 2006). The kinetic constant [k, units of (1/t)] is assumed to be related to climate. Emissions are estimated from the difference between modeled biogas generation and estimated or measured biogas recovery after applying a measured or default (50%) mixing ratio for the CH_4 content of the biogas. In the IPCC method, emissions can also be reduced by an additional 10% to account for CH_4 oxidation in cover soils. The 10% is based on the first paper in the literature estimating annual landfill CH_4 oxidation at a small New Hampshire (USA) landfill without an engineered gas recovery system (Czepiel et al., 1996). Importantly, as there were few field measurements for landfill CH_4 emissions when this method was adopted (1996), the historic "field validation" of the IPCC and LandGEM models was limited to comparisons between modeled biogas generation and measured biogas recovery (see Peer et al., 1993; Oonk and Boom, 1995; van Zanten and Scheepers, 1995; Oonk, 2010, 2012; and references cited therein).

More recently, the U.S. EPA has required individual landfills to estimate CH_4 emissions under the Mandatory Reporting Rule (MRR) for GHG emissions using several required HH- formulas incorporating IPCC (2006) equations (See Subpart HH- for Municipal Solid Waste Landfills at http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98_main_02.tpl). In lieu of measured LFG recovery, and in spite of the fact that LFG recovery efficiency has been rarely measured at field scale (e.g., Spokas et al., 2006), a "recovery efficiency" factor is applied where engineered gas recovery exists. Briefly, assigned LFG recovery factors vary from 60% (areas with daily cover only) to 75% (areas with either intermediate cover or final soil cover <3 ft thick) to 95% (areas with a final soil cover >3 ft thick or a geomembrane composite cover) [See Subpart HH-Municipal Solid Waste Landfills-Calculating Collection Efficiency for Use in Equations HH-7 and HH-8, e-GGRT RY2013.01]. Under the MRR, there is also a sliding scale for % oxidation assignments including 0% (areas with a geomembrane composite cover overlain by <12 inches soil), 10% (default for other areas), and up to a maximum of 35% for areas with ≥24 in soil covers depending on *estimated* emission rates [See Table HH-4 to Subpart HH of Part 98 – Landfill Methane Oxidation Fractions]. Herein we will compare total

measured emissions to the total 2010–2014 modeled CH_4 generation, the current measured CH_4 recovery, and the estimated emissions under the current reporting requirements summarized above.

Results and discussion

Whole-site emissions: temporal variability and uncertainty

Figure 2 shows the time series distribution of whole-site CH_4 emissions determined using the TCA and AMB data. The corresponding TCA mean and standard deviations (1σ) together with the measurement dates and the number of points are provided in Table S3 (supplementary information) while the AMB estimates for the two downwind distances corresponding to the two flight dates are reported in Table S2 in the supplementary information. **Figure 2** shows that many of the whole-site field campaigns were conducted during months that were relatively convenient for outdoor field measurements, i.e., a notable absence of measurements during the colder winter (January and February), and early spring months (March and April) was observed. To determine the annual cycle of observed emissions, multi-year TCA measurements for a particular month were combined in a percentile plot shown in **Figure 3**. The monthly average CALMIM modeled results with their standard deviation are also plotted in this figure; these are based on 30-year average weather data for the site latitude and longitude, as well as the site-specific cover soils (Spokas et al., 2011). For the intermediate and extended daily cover soils, site-specific model inputs included the average measured soil gas CH_4 and O_2 mixing ratios at the base of cover.

Landfill CH_4 emissions are composed of CH_4 diffusing through cover soils, as well as any fugitive emissions through cracks in cover materials and leakages associated with cell boundaries or the gas recovery infrastructure. Multi-year measurements (2009 – 2012) from the TCA approach (**Figure 3**) display a relatively large range in the observed emissions as shown by the box and whisker plot with median values (red line) ranging from 8 to 13 mol s^{-1}. We calculated the standard deviations of successive TCA multi-passes downwind of the source, i.e., for sequential measurements conducted within 30 min to 2 hrs on the same day, and found the variability ranging from 11% to 55% with an average precision of 25%. For comparison, the two aircraft experiments yielded mean AMB emissions estimates of 17 mol s^{-1} for the 30 August 2012 experiment (Cambaliza et al., 2014, 2015) and 7 mol s^{-1} for the 03 July 2014 flight measurement. The mean AMB emissions were calculated from the independent measurements at two downwind distances from the landfill as discussed in the methods section. The individual results from the two downwind measurements corresponding to the two flight campaigns are shown in Table S2 in the supporting information. These mean results are not statistically different from the TCA median values given the range of uncertainties of the two approaches. As stated in the Methods section, uncertainty in the AMB estimates is ±30%, which corresponds to the single point method precision for area/point sources (Cambaliza et al., 2014). This uncertainty is a measure of the combined influences of various parameters such as the variability of the atmospheric boundary layer conditions, interpolation errors, sampling statistics, and instrument limitations (Cambaliza et al., 2014, 2015).

The TCA median values ranging from 8 – 13 mol s^{-1} indicate significant CH_4 emissions. For comparison, following the procedure of Cambaliza et al. (2015), if captured and converted to electrical energy, this corresponds to the annual energy requirements of 2300 to 3800 Indiana households. Given the fact that IN-1 is upwind of Indianapolis when winds are westerly, it is essential to account for its contribution to city CH_4 emissions.

Both the TCA and AMB results provide useful estimates of the whole site CH_4 emissions. However, they have limited

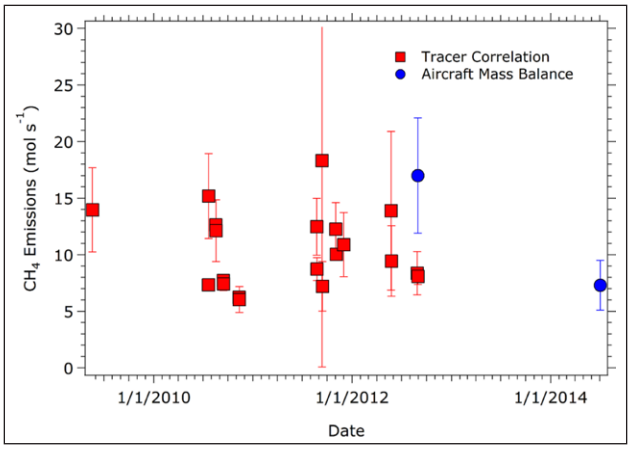

Figure 2: Time series distribution of IN-1 whole-site CH_4 emissions. The TCA (tracer correlation approach) data were taken from 2009 – 2012 (See Table 1 for the corresponding months) while the AMB (aircraft mass balance) estimates corresponded to flight dates conducted on 30 August 2012 and 03 July 2014. DOI: https://doi.org/10.1525/elementa.145.f2

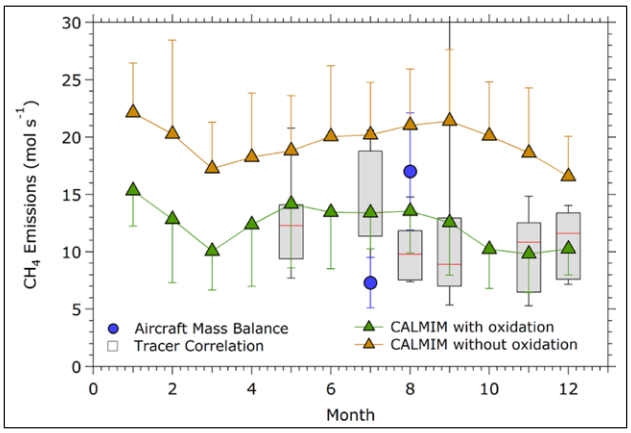

Figure 3: Monthly measured and modeled CH_4 emissions from the central IN landfill. The TCA box and whisker plot has the following percentile values: box bottom: 25th, box top: 75th, whisker bottom: 10th, whisker top: 90th, and red line: median. The CALMIM modeled results are shown for two cases: with and without oxidation. Also shown are the AMB emissions estimates on 30 August 2012 and 03 July 2014. DOI: https://doi.org/10.1525/elementa.145.f3

temporal duration. They provide a snapshot of emissions over certain hours on several days within a given month. Hence, models such as CALMIM are useful for investigating the seasonal variability of emissions. The CALMIM monthly-averaged modeled emissions shown in **Figure 3** were determined for two cases, i.e., inclusive and exclusive of soil microbial oxidation, which represent endpoints for the two major theoretical baselines for CH_4 emissions through landfill cover soils. Error bars in the CALMIM results correspond to the standard deviation of the [10-min time step] modeled surface emissions for a given cover soil for a given month using 30-year default weather data. We find that the CALMIM modeled emissions with oxidation lie at or below the 75th percentile of the TCA observations. Turning off the process of oxidation in CALMIM yields upper bound modeled emissions that lie within the 75th to the 90th percentile of the TCA measurements. The mean annual modeled emissions with oxidation is determined to be 12 ± 2 mol s^{-1}, which is not statistically different from the range of TCA median values or the AMB estimated emissions.

CH_4 emissions through each individual landfill cover soil is regulated by a combination of several factors: the presence of engineered gas extraction systems, properties of the landfill cover materials (e.g. texture, thickness), and seasonal climate effects on both gaseous transport and methanotrophic CH_4 oxidation (Spokas et al., 2011). The annual cycle of emissions (**Figure 3**) is characterized by spatial variability between the various cover soils and temporal variability due to the dependency of both gaseous transport and CH_4 oxidation on soil temperature and moisture (Spokas and Bogner, 2011). We note that anoxic CH_4 production in the buried waste is largely unaffected by ambient temperatures, due to the insulating properties of most engineered cover soils (Chanton and Liptay, 2000), whereas CH_4 oxidation rates in the aerobic cover soils are highly variable over a typical annual cycle as the result of variable soil temperature and moisture conditions. Spokas and Bogner (2011) have shown that CH_4 oxidation rates are highest at soil temperatures ranging from 25–35°C and soil moisture potentials near field capacity. In **Figure 3**, CALMIM modeled results inclusive of oxidation show elevated emissions of 15 mols s^{-1} in January followed by a decline for the rest of the winter months with a dip in March to about 10 mol s^{-1}. The modeled emissions increase in April and May, and stabilize at a relatively constant and elevated value of 15 mol s^{-1} from June to August, then decrease to ~10 mols s^{-1} from September to December. We note that seasonal trends in whole-site landfill emissions are dependent both on seasonal site-specific microclimate (referenced to latitude and longitude) as well as site engineering/operational practices, especially the thickness and properties of cover materials and the extent of biogas recovery under each cover.

Area-specific emissions: spatial variability and uncertainty

Figure 4 compares monthly modeled and measured CH_4 emissions from the various cover areas. As in **Figure 3**, CALMIM-modeled CH_4 emissions rates are plotted with and without soil oxidation. We note that very low unit emissions are associated with the thicker final (**Figure 4A**) and intermediate covers (**Figure 4B**), as both have engineered gas recovery systems and significant seasonal oxidation. The thinner extended daily cover area without gas recovery (**Figure 4C**) has higher unit emissions due to reduced soil oxidation during the cold winter months (DJFM) and higher soil gas CH_4 gradients. The different landfill cover materials and thicknesses combined with the meteorological factors influencing oxidation obviously affect the surface emissions, resulting in unique emissions signature from each area. Superimposed on the modeled results are the measured static closed chamber and VRPM emissions. In general, the observations lie within the bounds of the two baseline modeled cases (with and without oxidation), suggesting reasonable agreement between the modeled and measured results. However, significant variability was observed for the VRPM data. We note that Goldsmith et al. (2012) reported the standard error and the number of measurements conducted downwind of the intermediate and final cover areas of IN-1. To be consistent with the reporting of the uncertainties for all measurement approaches, we calculated the standard deviation of the VRPM measurements. We note that due to the large variability in the observed VRPM results for the intermediate cover, the standard deviations are reported right next to the marker label rather than displayed as error bars. One challenge with the VRPM method is that it works best in topographically flat areas; thus non-uniform landfill topography creates variable wind vectors that affect the accuracy of the approach (Goldsmith et al., 2012). Furthermore, an associated uncertainty with determining the area contributing to flux can, under optimum conditions, contribute 10 – 30% uncertainty (Abichou et al., 2010), or even more under typical field conditions.

We note that CH_4 emissions from the small daily cover area (**Figure 4D**) were not measured during our field campaigns: static chambers, VRPM, and TCA could not be practically implemented due to the boundary conditions, irregular topography, and highly variable surface conditions. In the absence of measurements for this cover, we report the modeled results. For this area, CALMIM was used in two ways: one simulation for daytime hours when there was no cover (simulated with 2 cm of sand) and secondly, the nighttime flux using the designated daily cover. **Figure 4D** shows the very high combined daytime and nighttime emissions rates from this area that are at least 10 times larger than the extended daily cover and intermediate/final covers. These modeled results are reasonable since at this particular site, the working area directly overlies thick sections of fully methanoganic older waste without intervening intermediate cover or any biogas recovery infrastructure. This is a typical practice when a landfill is expanded vertically for a new layer of cells at a topographically higher elevation. In general, the pre-existing intermediate cover is stripped and stockpiled for future use. Then the new cell directly overlies a thick sequence of fully methanogenic older waste, often without biogas recovery infrastructure. This practice preserves landfill volume for new waste placement and retards ponding of infiltration. We find that the annual average of the

combined daytime and nighttime CH_4 emissions with oxidation from the small daily cover area was 11 ± 1 mol s^{-1} (2200 ± 250 g m^{-2} d^{-1}). This corresponds to ~90% (**Figure 5**) of the annual average CH_4 emissions for the entire site. **Figure 5** shows the percentage emissions rate contribution from each landfill cover area together with the corresponding area footprint. Considering that the small active filling area occupies only 2% of the total landfill footprint, its contribution to emissions is almost 300% greater than the other covers on an area basis. Even though we were unable to directly measure emissions from this area, we noted very high near-surface atmospheric CH_4 values immediately adjacent to, downslope, and downwind from this area in connection with the chamber measurements on the adjacent "extended daily cover" area. Table S4 (supplementary information) compare the initial chamber atmospheric concentrations of several gases in the upland, upwind intermediate cover area, and in the extended daily cover, which was downslope and downwind from the daily filling area. The initial chamber (air) CH_4 values from the extended daily cover ranged from 8 – 357 ppm and averaged 53 ppm (Table S4, supplementary information), thus indicating high atmospheric CH_4 enrichment. As shown in **Figure 4**, expected CH_4 oxidation is negligible in this very thin daily cover.

Comparison of measured CH_4 emissions to reported emissions under U.S. EPA National GHG Reporting Program using IPCC (2006)

The reported whole landfill CH_4 emissions (mol CH_4 s^{-1}) for IN-1 under the US EPA National Greenhouse Gas Reporting Program for the years 2010 – 2014 are shown in **Table 2**. HH-1 refers to total modeled CH_4 generation using IPCC (2006) based on a first order kinetic model and the estimated degradable organic carbon content of the waste. HH-6 includes subtractions for measured CH_4 recovery and 10% oxidation for 2010–2012 but up to 35% oxidation for 2013–2014 for thicker clay covers. Table S5 (supplementary information) provides the various landfill site conditions and the corresponding oxidation rate to use in the HH-6 calculations. HH-8 includes recovery adjusted for collection efficiencies of up to 95% for thicker

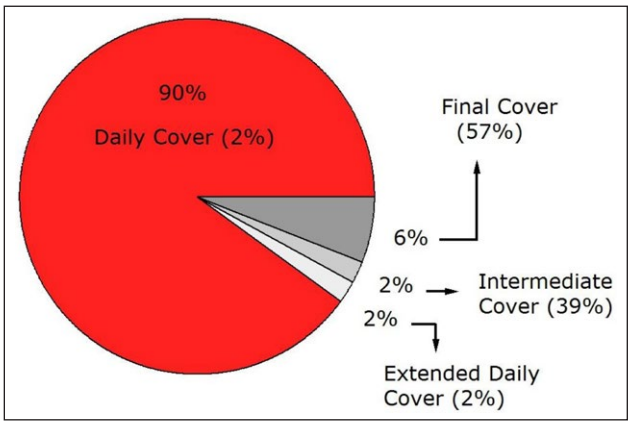

Figure 5: CH_4 emissions contribution from IN-1 landfill covers. The percentage value refers to the emissions contribution of each landfill cover while the corresponding fractional area footprint is provided inside the parenthesis. The daily cover CH_4 emissions (11 ± 1.2 mol s^{-1}) contribute about 90% to the total modeled landfill emissions rate (12 ± 2 mol s^{-1}). DOI: https://doi.org/10.1525/elementa.145.f5

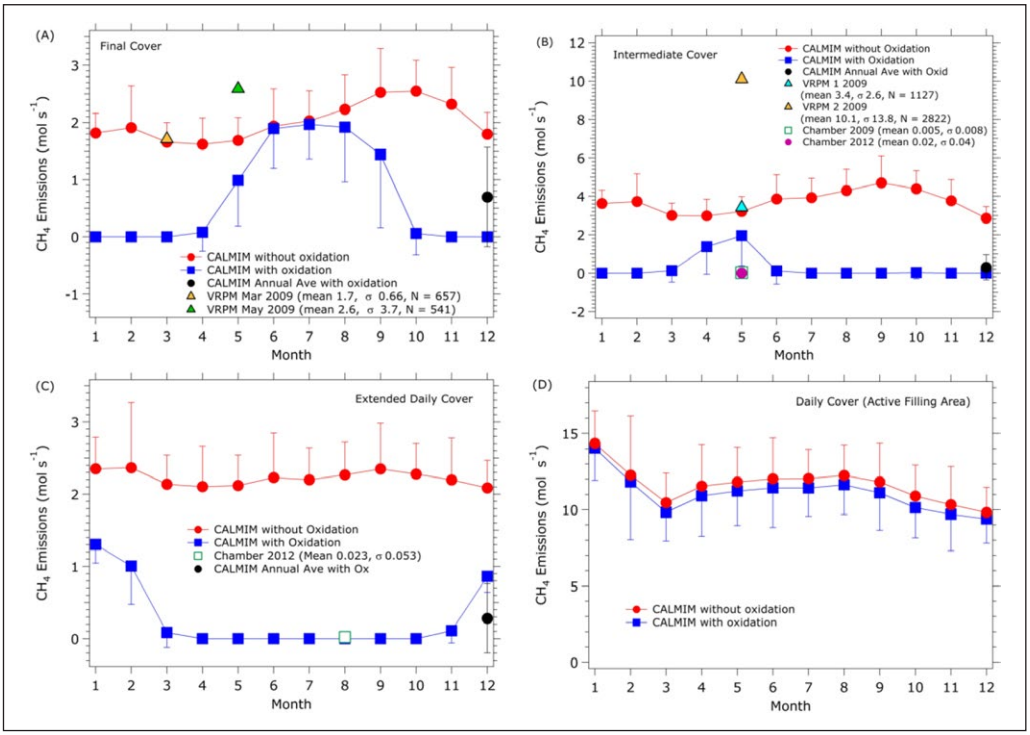

Figure 4: Measured and modeled average monthly CH_4 emissions (mol s^{-1}) for individual cover areas. (A) Final Cover (soil only, negligible emissions from final cover with geomembrane), **(B)** Intermediate Cover, **(C)** Extended Daily Cover, **(D)** Active Filling Area (thin overnight daily cover). DOI: https://doi.org/10.1525/elementa.145.f4

Table 2: Reported whole landfill CH_4 emissions (mol CH_4 s^{-1}) for IN-1 under the U.S. EPA National Greenhouse Gas Reporting Program for the years 2010 – 2014. DOI: https://doi.org/10.1525/elementa.145.t2

Year	HH-1[a]	HH-6[b]	HH-8[c]
2010	64.0	16.4	10.5
2011	65.4	19.3	8.4
2012	50.9	9.5	6.0
2013	62.9	15.5	6.1
2014	64.2	18.1	5.9

[a]HH-1 is the total modeled CH_4 generation using IPCC (2006).

[b]HH-6 includes subtractions for measured CH_4 recovery and 10% oxidation for 2010–2012 but up to 35% oxidation for 2013–2014 for thicker clay covers.

[c]HH-8 includes recovery adjusted for collection efficiencies of up to 95% for thicker clay covers with low estimated CH_4 flux rates to the base of the cover. These are the reported values to the GHGRP.

clay covers combined with lower estimated CH_4 flux rates to the base of the cover. It is important to note that the values reported to the GHGRP also reflect changes in individual cover areas during 2010–2014.

Comparing these values to the measured results and CALMIM-modeled results in **Figures 2** and **3**, the HH-8 values fall at or below the lower 10% percentile for the TCA results, as well as generally below the AMB results and the average CALMIM-modeled results with oxidation. We attribute this discrepancy to the empirical nature of the HH- calculations, which do not take into consideration the temporal site-specific climate effects on both gaseous transport and CH_4 oxidation rates in individual cover materials, which are considered by CALMIM. We also question the HH-1 methane generation calculation based on a single first order kinetic equation which is currently applied to all global landfills (IPCC, 2006). This equation assumes that the annual total CH_4 generation rate per unit mass of waste peaks in the year of waste placement and exponentially declines thereafter. Neither the theoretical generation nor the % recovery efficiency can be routinely validated by field data and, indeed, has been rarely quantified at field scale (Spokas et al., 2006). Recently, as discussed in Spokas et al. (2015), 2010 field data for 129 well-managed California landfills indicated a robust linear relationship ($r^2 = 0.85$) for *measured* landfill CH_4 recovery compared to *measured* waste-in-place (approx. 125 Nm^3 CH_4 per million Mg landfilled waste). As the California dataset included open/closed sites, small/large sites, old/new sites in all climatic regions of the state, this simple relationship contradicts the HH-1 first order kinetic equation where biogas generation peaks in the year of disposal with an exponential decline thereafter (See additional discussion in Spokas et al., 2015; Bogner et al., 2016). Moreover, under current regulations, U.S. landfills are required to perform routine quarterly monitoring for CH_4 concentrations near the ground surface followed by remediation and re-testing where elevated concentrations are observed, thus periodically checking for CH_4 leakages. Finally, if we consider the 2010–2014 data for all of Waste Management's U.S. landfill sites (>200 sites, 942 data points 2010–2014), approximately 85% of those data points do not fit the historic assumed recovery efficiency range of 75–100% of theoretical biogas generation (IPCC, 2006), with approximately 15% > 100% of theoretical generation and the remainder below 75% .

Remaining challenges and uncertainties

U.S. landfills are highly engineered and managed facilities with gaseous emissions regulated under the Resource Conservation and Recovery Act (RCRA), the Clean Air Act, the Greenhouse Gas Reporting Program, and other Federal, state, and local legislation. Typically, sites include a relatively small (few ha) active filling area with daily cover placement at the end of the working day. However, unit emissions (g CH_4 $m^{-2}d^{-1}$) from that area can drastically differ depending on whether the active area overlies (1) new waste only, with relatively low emissions per unit area; (2) older underlying cells containing fully methanogenic waste but with an intervening intermediate cover, resulting in higher emissions from the combined waste as opposed to the new waste only; or (3) older deeper cells where the intermediate cover has been stripped away prior to the development of the active area, resulting in very high unit emissions from the combination of older and younger waste. In addition, another consideration is whether engineered gas recovery exists below (horizontal collectors) or adjacent to (vertical wells) the daily filling area; if gas recovery exists, emissions are significantly reduced. For the future, field measurement of daytime vs. nighttime emissions for the active filling area should be a research priority. However, emissions measurements for this area pose significant challenges, as this small, sometimes highly-emitting area is often immediately adjacent to areas where unit emissions can be orders of magnitude lower. New measurement approaches (e.g., Gålfalk et al., 2016) may assist with future field campaigns by comparing relative emissions signatures from closely-spaced areas under favorable conditions. For CALMIM modeling, we note that current CALMIM defaults for the daily cover assume new waste only (Bogner et al., 2011; Spokas et al., 2011). However, one can readily model scenarios (2) or (3) by assuming a default intermediate, final, or custom soil gas profile, as appropriate. Importantly, for this Indiana site, scenario (3) was the appropriate choice. Our study

showed that while it is important to determine the whole-site CH_4 emissions, it is equally important to quantify the contributions from the various cover areas to identify the most significant contributor(s) and plan mitigation strategies accordingly. In this study, the small daily filling area, which comprises only 2% of the total landfill area, contributed about 90% of the annual average CH_4 emissions.

Our results also lead to reflection on improved quantification of emissions from large open dumpsites in developing countries which may lack engineered controls for waste placement, liquids, and gases. Such sites may have very thin cover soils characterized by very high unit emissions (e.g., Morris, 2001). Improved quantification of fluxes from such sites should be a priority, and "whole landfill" AMB approaches may be the appropriate technique, as the lack of engineered cover soils, irregular topography, and complex meteorology can constrain the use of TCA, static closed chamber or VRPM methods.

Finally, our comparison of observed to reported CH_4 emissions under the US EPA National GHG Reporting program underscores the need to update the current IPCC (2006) inventory methodology based on estimated CH_4 generation to a more direct approach based on explicit modeling of CH_4 emissions. Using CALMIM, for example, modeling relies on climate and process drivers known from literature (cover soil characteristics, seasonal climatic variability in soil moisture and temperature, specific site operational practices) and is consistent with field-measured results as shown in this study. Given the differences between measured and reported CH_4 emissions using the two approaches, there is a need to consider moving towards a Tier 3 field-validated emissions-based inventory methodology as discussed in this study.

Data Accessibility Statement

Data from this study is available upon request from the corresponding author, and will be made publicly available on the INFLUX website: http://sites.psu.edu/influx/ by 30 June 2017.

Supplemental Files

The supplemental files for this article can be found as follows:

- **Text S1.** Resolving m^2 to km^2 CH_4 emissions for a complex urban source: An Indiana landfill study. DOI: https://doi.org/10.1525/elementa.145.s1
- **Table S1.** Properties of the various IN-1 landfill cover types used as inputs to CALMIM 5.4. DOI: https://doi.org/10.1525/elementa.145.s2
- **Table S2.** AMB Background CH_4 mixing ratio and estimated CH_4 emission rate corresponding to two downwind distances from IN-1 during the 30 August 2012 and 03 July 2014 flight experiments. DOI: https://doi.org/10.1525/elementa.145.s3
- **Table S3.** Mean and Standard deviations of the TCA estimated whole-site CH_4 emissions (mol s^{-1}) from 2009 – 2012. DOI: https://doi.org/10.1525/elementa.145.s4
- **Table S4.** Comparison of upwind and downwind CH_4 Mixing ratios at ground level at IN-1. DOI: https://doi.org/10.1525/elementa.145.s5
- **Table S5.** HH-6 to Subpart HH of Part 98 – Landfill Methane Oxidation Fractions. DOI: https://doi.org/10.1525/elementa.145.s6
- **Figure S1.** Experimental flight path as a function of altitude on 30 August 2012 for sampling the CH_4 plume from IN-1. DOI: https://doi.org/10.1525/elementa.145.s7
- **Figure S2.** Experimental flight path as a function of altitude on 03 July 2014 at the IN-1 Landfill site. DOI: https://doi.org/10.1525/elementa.145.s8
- **Figure S3.** Horizontal distributions of the methane plume on 30 August 2012. DOI: https://doi.org/10.1525/elementa.145.s9
- **Figure S4.** Horizontal distributions of the methane plume on 3 July 2014. DOI: https://doi.org/10.1525/elementa.145.s10

Acknowledgements

MOLC, PBS and BHS are grateful to the Purdue University Jonathan Amy Facility (JAFCI) for technical support in this project. JEB also thanks Margaret Corcoran, Dept. EAES UIC graduate student for field assistance (now at Oak Ridge Institute for Science and Education at US EPA Great Lakes National Program Office, Chicago).

Funding information

The aircraft-based mass balance experiments designed and conducted by MOLC, PBS, and BHS are part of the INFLUX project that is funded by the National Institute of Standards and Technology (NIST). JEB also acknowledges the Environmental Research & Education Foundation, Raleigh, NC for University of Illinois at Chicago (UIC) funding under UI contract G5534, and the UIC Reimbursable Cooperative Agreement No. 58-3640-1-194 between UIC and USDA.

Competing interests

The authors have no competing interests to declare.

Author contibutions

- Contributed to conception and design: MOLC, JEB, PBS, RBG, TAH, KAS, BHS, MC
- Contributed to acquisition of data: MOLC, JEB, PBS, RBG, TAH, BHS, MC
- Contributed to analysis and interpretation of data: MOLC, JEB, TAH, KAS, MC
- Drafted and/or revised the article: MOLC, JEB, PBS, KAS
- Approved the submitted version for publication: MOLC, JEB, PBS, RBG, TAH, KAS, BHS, MC

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How to cite this article: Cambaliza, MOL, Bogner, JE, Green, RB, Shepson, PB, Harvey, TA, Spokas, KA, Stirm, BH and Corcoran, M 2017 Field measurements and modeling to resolve m^2 to km^2 CH_4 emissions for a complex urban source: An Indiana landfill study. *Elem Sci Anth,* 5: 36, DOI: https://doi.org/10.1525/elementa.145

Domain Editor-in-Chief: Detlev Helmig, University of Colorado Boulder, US

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Knowledge Domain: Atmospheric Science

Part of an *Elementa* Special Feature: Quantification of urban greenhouse gas emissions: The Indianapolis Flux experiment

Submitted: 09 December 2016 **Accepted:** 10 May 2017 **Published:** 04 July 2017

ELEMENTA Science of the Anthropocene

Elem Sci Anth is a peer-reviewed open access journal published by University of California Press.

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