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Quantification and source apportionment of the methane emission flux from the city of Indianapolis

M. O. L. Cambaliza [1*] • P. B. Shepson [1,2] • J. Bogner [3] • D. R. Caulton [1] • B. Stirm [4] • C. Sweeney [5,6] • S. A. Montzka [6] • K. R. Gurney [7] • K. Spokas [8] • O. E. Salmon [1] • T. N. Lavoie [1] • A. Hendricks [1] • K. Mays [1] • J. Turnbull [9] • B. R. Miller [6] • T. Lauvaux [10] • K. Davis [10] • A. Karion [5,6] • B. Moser [1] • C. Miller [1] • C. Obermeyer [1] • J. Whetstone [11] • K. Prasad [11] • N. Miles [10] • S. Richardson [10]

[1]Department of Chemistry, Purdue University, West Lafayette, Indiana, United States
[2]Department of Earth, Atmospheric and Planetary Science & Purdue Climate Change Research Center, Purdue University, West Lafayette, Indiana, United States
[3]Department of Earth & Environmental Sciences, University of Illinois at Chicago, Illinois, United States
[4]Department of Aviation Technology, Purdue University, West Lafayette, Indiana, United States
[5]University of Colorado, Boulder, Colorado, United States
[6]NOAA/ESRL, Boulder, Colorado, United States
[7]School of Life Sciences, Arizona State University, Tempe, Arizona, United States
[8]U.S. Department of Agriculture, St. Paul, Minnesota, United States
[9]National Isotope Centre, GNS Science, Lower Hutt, New Zealand
[10]Department of Meteorology, The Pennsylvania State University, University Park, Pennsylvania, United States
[11]NIST, Gaithersburg, Maryland, United States
*mcambali@purdue.edu/mocambaliza@gmail.com

Abstract

We report the CH_4 emission flux from the city of Indianapolis, IN, the site of the Indianapolis Flux Experiment (INFLUX) project for developing, assessing, and improving top-down and bottom-up approaches for quantifying urban greenhouse gas emissions. Using an aircraft-based mass balance approach, we find that the average CH_4 emission rate from five flight experiments in 2011 is 135 ± 58 (1σ) moles s^{-1} (7800 ± 3300 kg hr^{-1}). The effective per capita CH_4 emission rate for Indianapolis is 77 kg CH_4 $person^{-1}$ yr^{-1}, a figure that is less than the national anthropogenic CH_4 emission (~91 kg CH_4 $person^{-1}$ yr^{-1}) but considerably larger than the global figure (~48 kg CH_4 $person^{-1}$ yr^{-1}). We consistently observed elevated CH_4 concentrations at specific coordinates along our flight transects downwind of the city. Inflight investigations as well as back trajectories using measured wind directions showed that the elevated concentrations originated from the southwest side of the city where a landfill and a natural gas transmission regulating station (TRS) are located. Street level mobile measurements downwind of the landfill and the TRS supported the results of aircraft-based data, and were used to quantify the relative contributions from the two sources. We find that the CH_4 emission from the TRS was negligible relative to the landfill, which was responsible for 33 ± 10% of the citywide emission flux. A regression of propane versus methane from aircraft flask samples suggests that the remaining citywide CH_4 emissions (~67%) derive from the natural gas distribution system. We discuss the combination of surface mobile observations and aircraft city-wide flux measurements to determine the total flux and apportionment to important sources.

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Introduction

Next to carbon dioxide, methane (CH_4) is the second most important anthropogenic greenhouse gas, contributing ~0.48 W m^{-2} (~17%) to the total direct radiative forcing by long-lived greenhouse gases (Myhre et al.,

2013). Most sources and sinks of methane are identified but their contributions to the total global emission remain uncertain (Kirschke et al., 2013). Major anthropogenic sources of CH_4 include fossil fuel production and transmission, agricultural activities (enteric fermentation, rice cultivation, biomass burning), and waste sources (landfills and waste water treatment plants) (Denman et al., 2007; Dlugokencky et al., 2011). The natural sources of methane are dominated by wetland emissions with small but important contributions from termites and oceans. Global inverse modeling efforts (Mikaloff Fletcher, et al., 2004; Chen and Prinn, 2005; Neef et al., 2010) show that the total emission from anthropogenic sources currently accounts for ~60% of the source budget. CH_4 is a very potent greenhouse gas whose emissions are 28 to 34 times more effective than an equivalent emission of carbon dioxide in trapping heat in the atmosphere (when integrated over a 100-year time frame) (Myhre et al., 2013). Because of its relatively short lifetime (9.1 – 11.2 years, Myhre et al., 2013) and its large global warming potential, reduction in its emission may have a significant effect on the climate system in the near term (Montzka et al., 2011).

CH_4 emission estimates on the national, regional and city-scale are still poorly quantified. Top-down inverse modeling of European CH_4 emissions for 2001 to 2006 showed that total anthropogenic emissions from Northwest Europe were 40% higher than values compiled by the United Nations Framework Convention on Climate Change and 21% greater than estimated by EDGARv4.0 (Emissions Database for Global Atmospheric Research) emission inventory (Bergamachi et al., 2010). Using atmospheric CH_4 observations and a high resolution transport model, Miller et al. (2013) showed that current inventories from the US Environmental Protection Agency (US EPA) and EDGAR underestimate the national anthropogenic emission rate by a factor of 1.5 to 1.7. Similar results were also obtained for urban city centers, which were shown to be significant sources of CH_4 emissions with magnitudes much larger than reported in bottom up approaches (Mays et al., 2009; Wunch et al., 2009). Mays et al. (2009) further demonstrated that the measured CH_4 emissions from the city of Indianapolis do not correlate with emissions from mobile combustion sources. These results suggest that (1) there are poorly characterized emission sources in urban environments that are thereby underreported in emission inventories, or alternatively, (2) there may be unidentified sources of CH_4 in urban city centers. There is a need to identify and quantify the magnitude of various anthropogenic sources contributing to the total emissions from urban environments, as this information will aid in evaluating the success of future emission reduction strategies.

The CH_4 emission flux from urban environments has been previously estimated using a variety of approaches: standard eddy covariance (EC) measurement in Florence, Italy (Gioli et al., 2012), aircraft-based mass balance approach in Indianapolis, IN, USA (Mays et al., 2009), a combination of ground-based concentration and stable isotope measurements in Krakow, Poland (Kuc et al., 2003), and London, United Kingdom (Lowry et al., 2001), as well as the utilization of correlation slopes between CH_4 and carbon monoxide (CO) and/or carbon dioxide (CO_2) quantified using remote sensing techniques or flask sampling in the South Coast Air Basin, CA, USA (Wunch et al., 2009; Hsu et al., 2010; Wennberg et al., 2012; Peischl et al., 2013). The CH_4 emission from the Los Angeles South Coast Air Basin, for example, was quantified by Wunch et al. (2009) using spectroscopic measurements of column abundances of CH_4 and CO_2 together with the known bottom up emission inventory for CO_2. Hsu et al. (2010) similarly used correlations of CH_4 and CO in flask samples along with the inventory for CO to estimate CH_4 emissions. This method of determining the CH_4 emission rate relies on the accuracy of the emission inventories for the correlated species (CO or fossil fuel CO_2), and for CO_2, on the assumption that the total measured incremental CO_2 is essentially equal to the fossil fuel CO_2.

Fluxes from small area sources of CH_4 (e.g. landfills) have been previously measured using the static chamber method, micrometeorological technique, vertical radial plume mapping (VRPM), tracer correlation method, and aircraft-based mass balance approach (Hovde et al., 1995; Czepiel et al., 1996a; Bogner et al., 1999; 2011; Mosher et al., 1999; Tregoures, et al., 1999; Galle et al., 2001; Spokas et al., 2006; Borjesson et al., 2009; Scheutz et al., 2009; Abichou et al., 2012; Goldsmith et al., 2012 Peischl et al., 2013; Cambaliza et al., 2014). Some area sources can have considerable spatial heterogeneity and large areal extent. Landfills, for example, are complex sources because CH_4 emissions from landfill cover soils can vary over several orders of magnitude due to the texture and thickness of cover soils as well as seasonal climate and soil microclimate dependencies for gaseous transport and methanotrophic oxidation (Czepiel et al., 1996b; Bogner et al., 1997; Chanton and Liptay, 2000; Albanna et al., 2007; Lee et al., 2009; Chiemchaisri et al., 2011; Rachor et al., 2013). Landfill CH_4 emissions are also dependent on the direct effect of engineered biogas extraction systems (Abichou et al., 2006; Perdikea et al., 2008; Bogner et al., 2011). Thus, for landfill sites, small-scale enclosures or chambers are useful for determining spatial variability of fluxes, but a large number of chambers must be deployed to obtain an average whole site emission rate.

The tracer flux technique enables measurement of the emission relative to a known flux by releasing tracer gas at the location of the source at a known mass emission rate, and measuring the tracer and target gas. The enhancement in concentration of CH_4 and the tracer above background are measured downwind at a distance where the source and tracer gases are observed to be well-mixed. The presence of other major

CH_4 sources surrounding the area of interest makes it challenging to use the tracer technique, as it becomes difficult to simulate and distinguish the emission from two or more sources.

The VRPM method involves the use of an open path optical remote sensing technique (e.g. tunable diode laser absorption spectroscopy (TDL) or Fourier transform infrared spectroscopy (FTIR)) to measure the path integrated concentrations along five radial distances in a vertical plane downwind of the source (Hashmonay et al., 2001; Abichou et al., 2012; Goldsmith et al., 2012). The integrated concentration is multiplied by the component of the wind normal to the vertical plane to calculate the advected flux flowing through the crosswind plane. The application of this technique is restricted to flat horizontal surfaces, as irregular or rough topography of the source can cause unusual wind vectors that render the data unusable (Goldsmith et al., 2012). Another challenge with the technique is the determination of the effective surface area contributing to the derived emission rate especially for sources with large areal extent such as landfills (Abichou et al., 2012; Goldsmith et al., 2012).

The standard EC method is a robust measurement technique that requires very fast instrumentation able to capture the smallest flux-carrying eddies. The EC method is most accurate when the source is homogeneous and the terrain of the measurement site is flat over extended distances. The application of the EC approach becomes difficult for landfill sites where emissions are spatially heterogeneous (Tregoures et al., 1999). Furthermore, the results from tower EC calculations apply only for a small footprint. Hence, several towers are needed across a city to obtain an integrated emission estimate from an urban environment.

In this work, we utilize an aircraft-based mass-balance approach (Mays et al., 2009; Cambaliza et al., 2014) for quantifying area and city-wide surface methane fluxes. It is an attractive method in that the approach can readily cover the entire footprint of the city, that the mobility of the platform allows for the characterization of the boundary layer depth and variability, and that the uncertainties can be assessed and quantified (Cambaliza et al., 2014). We report the total CH_4 emission from the city of Indianapolis from several flight experiments with the additional goal of identifying and quantifying the contributions from the most important CH_4 sources within the urban environment. We note that Indianapolis (39.77°N, 86.16°W, Figure 1) is the city of focus of the Indianapolis Flux Experiment (INFLUX, http://sites.psu.edu/influx/), a collaborative study designed to develop, assess, and improve top-down and bottom-up approaches for quantifying greenhouse gas emissions in urban environments. The aircraft-based results presented here are complemented by surface mobile measurements to identify and determine the magnitude of specific sources of CH_4 within the city. Estimates of the citywide CO_2 emission flux for several flight experiments were reported in a separate manuscript (Cambaliza et al., 2014) and compared with bottom-up estimates for fossil fuel CO_2 from Hestia (Gurney et al., 2012). The Hestia project is a bottom-up approach for quantifying the hourly CO_2 emissions of all on-site fossil fuel sources at a finer scale of building and street level for the entire urban landscape, making use of traffic data, power generating and local air pollution reporting data, as well as a building energy simulation model (Gurney et al., 2012) (see also http://hestia.project.asu.edu/).

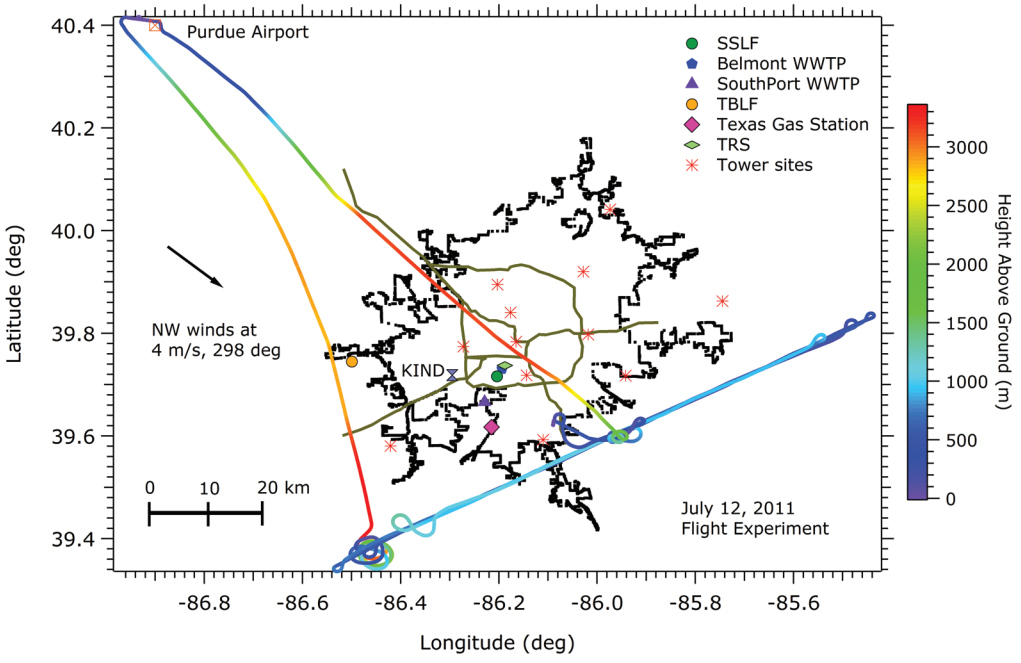

Figure 1

Experimental flight path on 12 July 2011 as a function of height above the ground.

Prevailing winds were from the northwest with an average magnitude of 4 m s^{-1}. Also shown in the figure are the sites of CH_4 sources within and just outside Indianapolis (black outline): Southside Landfill (SSLF), Belmont wastewater treatment plant (WWTP), Southport WWTP, and a landfill designated as TBLF. For reference, we also include the location of the Indianapolis international airport (KIND) as well as the network of 12 INFLUX tower sites. The dark green outline represents the major highway arteries traversing Indianapolis (I-65, I-70, and IN-465).

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Methods

Aircraft-based measurements of greenhouse gases and meteorological parameters

CH_4 and CO_2 emissions from the city of Indianapolis were quantified using an aircraft-based platform combined with a mass balance approach. Several flight experiments were conducted downwind of the city using Purdue University's Airborne Laboratory for Atmospheric Research (ALAR) (http://science.purdue.edu/shepson/research/bai/alar.html), a light aircraft with a compartment space of ~ 1 m^3. The twin-engine Beechcraft Duchess is equipped with (1) a global positioning and inertial navigation system (GPS/INS), (2) a Best Air Turbulence (BAT) probe for wind measurements (Garman et al., 2006), (3) a cavity ring-down spectroscopy system for in-situ, real-time CO_2, CH_4, and H_2O measurements, (4) an in-flight CO_2/CH_4 calibration system, and (5) a programmable flask package (PFP) system for discrete sampling of ambient air.

Ambient concentrations of CO_2, CH_4, and H_2O were measured at 0.5 Hz using a Picarro cavity ring-down spectrometer (CRDS) model G2301-f (Crosson 2008; Chen et al., 2010; Rella et al., 2013; Karion et al. 2013). Ambient air was pulled from the nose of the aircraft through 5-cm diameter PFA Teflon tubing at a flow rate of 1840 L min^{-1} using a high-capacity blower located at the rear of the aircraft. Using 0.64-cm o.d. Teflon inlet tubing, the CRDS continuously sampled from the 5-cm PFA Teflon line at a flow rate of 450 mL min^{-1} corresponding to a residence time of 6 s. Inflight calibrations for CO_2 and CH_4 were conducted using three NOAA/ESRL reference cylinders with the following mole fractions: 378.49, 408.83, and 438.29 ppm for CO_2, and 1803.0, 2222.2, and 2599.5 ppb for CH_4. The one sigma measured precisions at 0.5 Hz for CO_2 and CH_4 were 0.1 ppm and 2.6 ppb, respectively (Cambaliza et al., 2014).

Winds were obtained at 50-Hz using the BAT probe, a nine-port pressure differential probe that extends from the nose of the aircraft (Garman et al., 2006; 2008). The measured pressure variations across the hemisphere of the probe are combined with 50-Hz inertial data from the GPS/INS system to obtain the three-dimensional wind vectors. Air temperature was also measured using a microbead thermistor located at the center of the probe.

Discrete grab samples of ambient air were also collected from the aircraft using a PFP (Karion et al., 2013). Two flasks were filled simultaneously during sampling to provide sufficient air for the analyses. Flasks were analyzed at NOAA/ESRL and University of Colorado Institute for Arctic and Alpine Research (INSTARR) Laboratory (http://instaar.colorado.edu/research/labs-groups/stable-isotope-laboratory/) for a set of trace gases and stable isotopes (Montzka et al., 1993; Vaughn et al., 2004; Conway et al., 2011; see also http://www.esrl.noaa.gov/gmd/ccgg/aircraft/analysis.html for a description of the measurement analysis of the 55 species that include greenhouse gases, halocarbons and hydrocarbons, and stable isotopes). The 14C content of CO_2 is determined by first extracting CO_2 from whole air samples at the University of Colorado INSTARR Laboratory, then prepared to graphite and measured by accelerator mass spectrometry (AMS) at either University of California Irvine (http://www.ess.uci.edu/group/ams/home) or GNS Science (http://www.gns.cri.nz/Home/Services/Laboratories-Facilities/National-Isotope-Centre) (Turnbull et al., 2007). In this analysis, thirty flasks were analyzed at NOAA/ESRL for propane (C_3H_8), CH_4, and acetylene (C_2H_2).

Experimental flight design

Prior to each flight experiment, the prevailing wind direction was determined using the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT, Draxler and Rolph, 2012). The aircraft was oriented perpendicular to the wind direction and traversed constant altitude horizontal transects downwind of Indianapolis at various altitudes up to near the top of the convective boundary layer (CBL). All flight experiments were conducted between 11:00 and 16:00 hours (local time) when the CBL is most likely to be fully developed and consistent in height during the whole mass balance experiment. The city of Indianapolis (Figure 1) is about 70 km wide in the North - South direction, and about 50 km wide in the East–West direction. Thus, the horizontal transects were about 80 to 100 km long to sample beyond the extent of the city plume. This extended flight path allowed for the simultaneous determination of the regional background CO_2 and CH_4 concentrations at the transect edges where the downwind air was isolated from the urban emissions. Two and a half to three hours of flight measurements were used to generate a two-dimensional rastered plane downwind of the city of Indianapolis. The depth of the CBL was determined from vertical profiles of water, potential temperature, and variance of the vertical wind speed. Two sets of vertical profiles (before and after the horizontal transects) were typically flown during which the aircraft ascended and descended in a spiral flight path from close to the surface up to 4000 m above ground level (a.g.l.) in the free troposphere. Here, we discuss the results from five mass balance flight experiments in Indianapolis in 2011 (01 March, 29 April, 01 June, 30 June, and 12 July 2011).

Flux calculation

For the flux calculation, the downwind measurements of CH_4, CO_2, temperature, pressure, and perpendicular wind speeds were interpolated in a two-dimensional gridded plane using the kriging approach (Matlab-based

EasyKrig3.0, Chu, 2004; Mays et al., 2009; Cambaliza et al., 2014). The mean background CO_2 and CH_4 mole fractions, obtained from the edges of the 2D gridded plane, were subtracted from the interpolated 2D mixing ratio. The net molecular concentration (measured concentration – background concentration) at each grid cell (mol m^{-3}) was determined using the ideal gas law and the interpolated pressure and temperature matrices. The gridded differential concentration matrix was then multiplied by the mean gridded wind speed perpendicular to the 2D plane at each altitude to determine the net mass flow across the grid cells. The city-wide emission rate, F (mol s^{-1}), was finally calculated by integrating the net mass flow in the horizontal and vertical directions using equation 1,

$$F = \int_0^{z_H} \int_{-x}^{x} \left([C]_{ij} - [\bar{C}]_b \right) \bullet U_{\perp ij} \, dx dz \quad (1)$$

where z_H is the CBL depth, $-x$ and $+x$ are the effective horizontal boundaries of the city determined from projecting the city limits onto the horizontal transect plane, and dx and dz are the horizontal and vertical grid spacings (m), respectively. $U_{\perp ij}$ is the perpendicular component of the horizontal wind speed, $[C]_{ij}$ and $[C]_b$ are the gridded and mean background molecular CH_4 concentrations, respectively, and i and j are the horizontal and vertical grid cell indices, respectively.

Investigation of CH_4 sources via upwind trajectory flights and surface mobile measurements

In all 2011 flight experiments, downwind CH_4 enhancements of about 50 to 100 ppb above background were observed along the horizontal flight transects, e.g. from -8 km to +2 km (Figure 2A; Section 3). Two methods were used to investigate the potential sources of the CH_4 plume as well as their magnitudes. First, the plume was followed upwind during one flight experiment and was determined to originate from the southwest side of the city. Second, mobile surface measurements were conducted within the city of Indianapolis using a Mobile Surface Laboratory (MSL) on separate sampling days from the five flight experiments. In this experiment, a Ford Taurus station wagon was instrumented with a CRDS system (Picarro, Inc., model G2301-f) for CO_2, CH_4, and H_2O measurement identical to the system installed in ALAR, a vacuum pump, and a GPS system for mapping and storing the coordinates of the measurement, all mounted in a secure rack. A 1000-W inverter connected to the vehicle's 12V battery was used to power all devices. The GPS antenna was mounted on the roof of the vehicle. A 1.2-m, 0.635-cm o.d. length of PFA Teflon tubing was mounted at approximately 0.3 m above the roof of the vehicle and served as the inlet for the CRDS system. The residence time in the inlet line was 10 s. During the earlier implementation of the project (2012 to early 2013), the wind direction was obtained from the Indianapolis international airport meteorological station prior to each "drive-around"

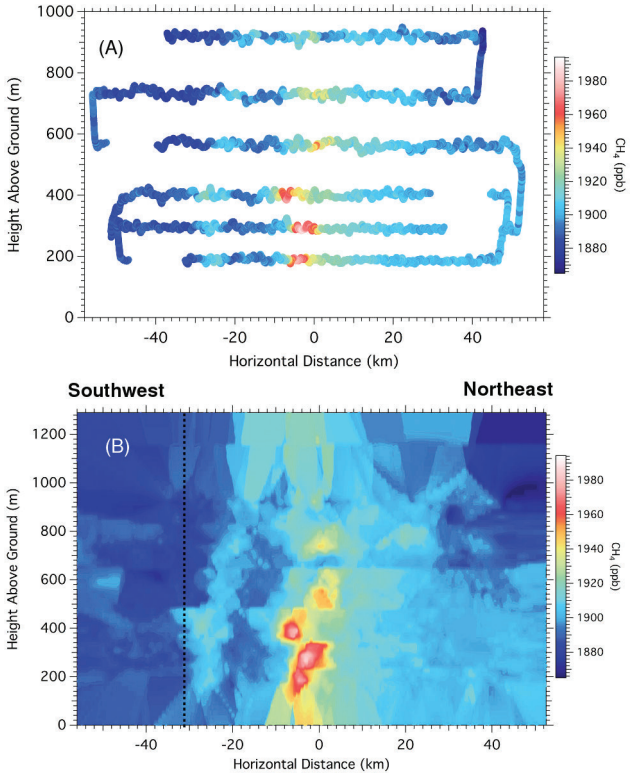

Figure 2

(A) Raw and (B) Kriged horizontal transect distribution of CH_4 on 12 July 2011.

The CH_4 distribution was shown as a function of height above the ground and horizontal distance. The CBL depth was 1290 m. The black broken line represents the boundary of the projected city width on the southwest side of the horizontal transects. The city limits (-x to +x) were -31 km to +46 km.

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measurement, and the drive path was chosen to be perpendicular to the prevailing wind direction. However, during the summer of 2013, we used a sonic anemometer (Campbell Scientific, CSAT3) installed on the roof of the vehicle, and winds were determined when the vehicle was stationary. The locations of known and potential sources of CH_4 within the city of Indianapolis (e.g. landfill, wastewater treatment plants, manufacturing plants, waste transfer station) were mapped to determine specific target measurement locations. In addition, we surveyed the city for other CH_4 sources by covering ~ 5000 road kilometers, systematically driving on roads that were nominally perpendicular to the wind direction.

Comparison of measured and modeled emissions from the Southside landfill with other Indiana landfills

As there was a large landfill located within the metropolitan area of Indianapolis, for comparison purposes we also applied our aircraft-based mass balance technique to other Indiana landfills and modeled the emissions from all sites using a new, field-validated process-based model (California Landfill Methane Inventory Model v 5.4, http://www.ars.usda.gov/services/software/download.htm?softwareid=300). CALMIM (Spokas and Bogner, 2011) is a field-validated one-dimensional transport and oxidation model that calculates the annual CH_4 emission based on: (1) surface area and properties of cover materials, (2) percent surface area of each landfill cover type with engineered gas recovery, and (3) meteorological factors (precipitation and temperature) that affect the microbial CH_4 oxidation in each cover type. CALMIM models 1-D diffusion of CH_4 and oxygen (O_2) through landfill cover soils inclusive of methanotrophic oxidation for 2.5 cm depth increments and 10-min time steps over a typical annual cycle (Bogner et al., 2011; Spokas et al., 2011; Spokas and Bogner, 2011). Embedded globally-validated (0.5 deg x 0.5 deg.) USDA climate and soil microclimate models (Global RAINSIM; Global TEMPSIM; SOLARCALC; STM2) enable site-specific calculations for both oxidized and unoxidized CH_4 emissions for cover soils entered by the user. CALMIM includes the major dependencies of emissions on the extent and properties of cover soils, extent of gas recovery, and seasonal oxidation.

The Indianapolis Southside landfill (SSLF, http://www.ssidelandfill.com/), a privately owned solid waste facility, is the only open landfill serving the city. To assess the quality and results of our method for estimating the CH_4 emission from SSLF, which is surrounded by other sources, we compared the measured and modeled emission rate from this solid waste facility with the measured and modeled emissions from four other Indiana landfill sites with various landfill capacities (Table 1). Unlike the SSLF that is located within the city boundary, the other four solid waste facilities are relatively isolated from other CH_4 sources making it easier to accurately quantify CH_4 emissions from these landfills.

The magnitude of landfill CH_4 emissions depends on the thickness and composition of cover soils, seasonal climate and soil microclimate affecting both gaseous transport and methanotrophic oxidation in cover soils (Chanton and Liptay, 2000), and the implementation of engineered gas collection and control systems (Spokas et al., 2011). The five Indiana landfill sites (Table 1) have different landfill footprints, soil cover properties, and soil microclimates, as the facilities are located in different parts of the state. Thus to uniformly compare the five sites, we model the site-specific emission using CALMIM. For this simulation for the SSLF and other Indiana sites, we used information furnished by the site operators. We also assume that the daily filling area (open face) was underlain by previous cells which were fully methanogenic. This assumption is based on the standard engineering practice at U.S. landfills to strip off the intermediate cover soil before filling a new landfill cell on top of the older cell. The older cell would have been previously filled for about 3 – 5 years, depending on the site, and methanogenesis would thus be fully established. Hence we distinguished between daytime "open face" and nighttime "daily cover" conditions. Table S1 provides the site-specific landfill data

Table 1. Description of Indiana landfills

County	Landfill designated name[a]	Total landfill surface area (x1000 m^2)	Flight expt. date/s	Estimated CH_4 emission (mol s^{-1})
Hendricks	TBLF	702	30 Aug 2012	17[b]
Marion	SSLF	923	See Table 2	45[c]
Newton	NCLF	795	16 June 2011, 03 May 2012	80[d]
Randolph	RFLF	455	07 July 2011	24
Shelby	CLF	324	04 May 2011	8.5

[a]We assign code names to the landfills to withhold their identities.
[b]Mean emission flux obtained from measurements at two downwind distances from the landfill site (see Cambaliza et al., 2014).
[c]Mean SSLF CH_4 emission flux from five Indianapolis flight experiments shown in Table 2.
[d]Mean emission flux obtained from measurements at two downwind distances for two flight dates, yielding four independent CH_4 flux estimates (see Cambaliza et al., 2014).
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that were used as inputs in the model. In this analysis, we calculated the site-specific CH_4 emissions for two cases, i.e., including and excluding soil CH_4 oxidation (denoted as "with" and "without" oxidation).

To quantify the CH_4 emission from the landfills, we used the same aircraft-based mass balance approach employed for the city of Indianapolis. For two of the four landfill sites (denoted as TBLF and NCLF in Table 1), we flew several transects at various altitudes in the CBL at two different downwind distances from the facility to quantify the precision of the aircraft mass balance approach. Under steady conditions such as constant, sustained winds and boundary layer depths as well as constant emission from the source, the measured emission fluxes from two or more downwind distances should ideally be identical (Cambaliza et al., 2014). Thus, a difference in the emission rates is a measure of the combined influences of various parameters (variability of the atmospheric boundary layer conditions, interpolation errors, sampling statistics, and instrument limitations), and is therefore a measure of the precision of the approach. We find that for small area and point sources (landfill and power plant), the variability in the estimated emissions ranged from 10% to 50%, with an average precision of 30% (Cambaliza et al., 2014), which is at the low end of reported uncertainties of previous studies that made use of an aircraft-based mass balance approach to quantify the emissions from urban environments or small area sources (see Cambaliza et al., 2014, and references therein).

Results and discussion

Citywide CH_4 emission estimates

We calculated the citywide CH_4 emission flux for several flight experiments downwind of Indianapolis conducted on the following dates in 2011: 01 March, 29 April, 01 June, 30 June, and 12 July. Figure 1 shows a sample flight path from the 12 July 2011 flight experiment in which several downwind horizontal transects were flown perpendicular to the wind direction (mean wind speed and direction of 4 m s^{-1} and 298°, respectively). Two vertical profiles were also flown to 4000 m a.g.l. in a spiral flight pattern. The black outline represents the boundary of the city while the green lines are the major highway arteries traversing the city. We also show in Figure 1 the location of important point and area sources of CH_4 located within and just outside the city boundaries such as the SSLF, Belmont Wastewater Treatment Plant (WWTP), Southport WWTP, a natural gas transmission regulating station (TRS), and a landfill that is designated as TBLF. For reference, the locations of the Indianapolis international airport (KIND) as well as the twelve INFLUX tower sites are also included (Miles et al., 2013).

Figure 2 shows the raw and interpolated horizontal transect concentrations of CH_4 as a function of altitude and horizontal distance for the 12 July 2011 flight experiment for which we saw enhancements in the CH_4 plume between 20 to 100 ppb above background. The black broken line (Figure 2B) at -31 km represents the boundary of the projected city width in the southwest side of our flight transects. Our horizontal transects were just long enough to encompass the projected city width on the northeast side of the transect. Using the interpolated perpendicular winds and the background concentration, the calculated citywide CH_4 emission rate for this day is 198 moles s^{-1}. Table 2 and Figure 3 summarize the citywide CH_4 emission for the five

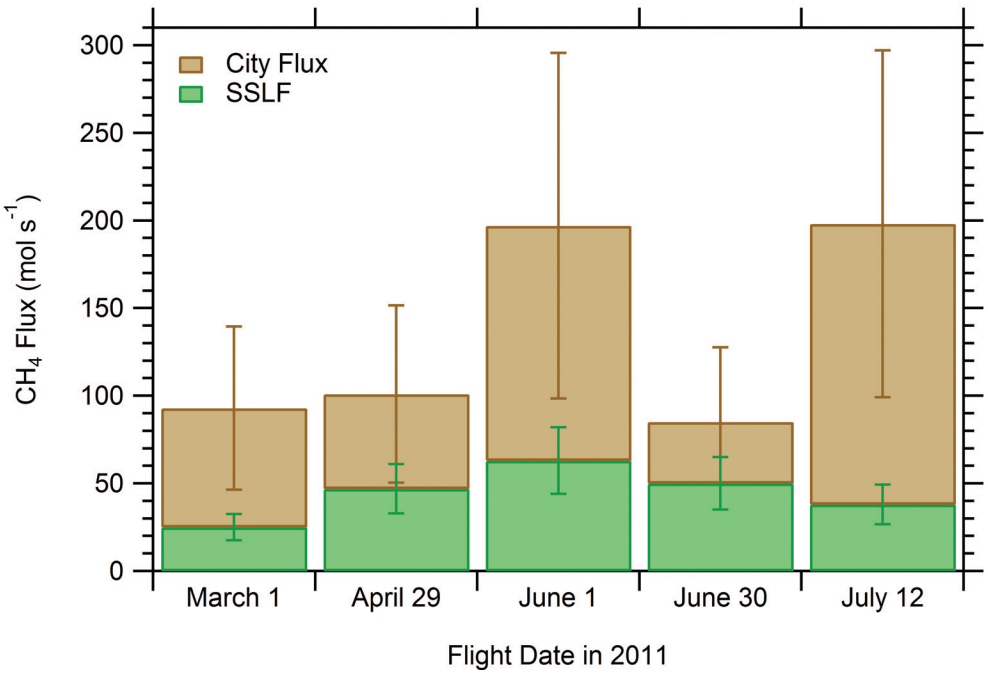

Figure 3

Citywide CH_4 emission derived from the aircraft-based mass balance approach for five 2011 flight experiments.

Also shown in the figure are the measured contributions from SSLF. Error bars correspond to ± 50 % uncertainty on the total flux, and ± 30 % on the SSLF contribution (Cambaliza et al., 2014).

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Table 2. Summary of CBL depths, mean wind speed and direction, citywide CH_4 emissions, and contribution from SSLF and TRS for several Indianapolis flight experiments in 2011

Flight date in 2011	CBL depth (m)	Mean wind speed ($m\ s^{-1}$), and wind direction (deg)	Citywide CH_4 emission ($mol\ s^{-1}$)	SSLF + TRS CH_4 emission($mols\ s^{-1}$)	Percent (%) contribution from SSLF+TRS
01 Mar 2011	525	5.9, 200	93	25	27
29 Apr 2011	1110	4.0, 332	101	47	47
01 June 2011	1720	5.9, 283	197	63	32
30 June 2011	1350	2.9, 206	85	50	59
12 July 2011	1290	4.0, 298	198	38	19

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flight experiments in 2011. In a separate manuscript (Cambaliza et al., 2014), we examined the uncertainties in our aircraft-based mass balance approach, and estimated the uncertainty of the citywide emissions to be ± 50% with the largest uncertainties contributed by the variability in the background concentrations and boundary layer depth. From the five 2011 flight experiments, the mean citywide CH_4 emission is 135 ± 58 (1σ) moles s^{-1} (7780 ± 3340 kg hr^{-1}) (Table 2).

As previously mentioned, the citywide CO_2 emissions were also determined for several flight experiments (Cambaliza et al., 2014; Mays et al., 2009). We thus apply an independent approach to determine the average citywide CH_4 emission based on the mean emission ratio between the total CH_4 and CO_2 mass balance emission estimates together with the Hestia bottom-up CO_2 emission for Indianapolis. The total uncertainty in the Hestia Indianapolis fossil fuel CO_2 emissions is -15%, +20% at the 95% confidence interval (Cambaliza et al., 2014).

The use of the CH_4:CO_2 flux ratio (or CH_4 emission factor) to obtain an independent CH_4 flux estimate essentially minimizes the uncertainties associated with the aircraft-based mass balance approach such as uncertainties in boundary layer depth, wind speed, and sampling errors that affect the flux determinations for both species. The accuracy of the result then relies to a large extent on the uncertainty of the bottom-up Hestia estimate of the citywide fossil fuel CO_2 flux. The use of emission factors to obtain total average emission rate has been previously described (Wunch et al., 2009; Hsu et al., 2010; Turnbull et al., 2011; Miller et al., 2012; Wennberg et al., 2012; Peischl et al., 2013). In these previous works, the scaling ratio of the concentration enhancements of two trace gases (e.g. CH_4 and CO, or CH_4 and fossil fuel CO_2) referenced to the bottom up inventory of one of the pollutants (e.g. CO) is used to obtain the emission rate of the trace gas of interest. This concentration enhancement ratio approach assumes that the trace gases are non-reactive within the time scale of dispersion and that they are well-mixed before they reach the point of observation (Hsu et al., 2010). The first assumption is valid because CH_4, CO and CO_2 are long-lived gases. The latter assumption makes the concentration ratio approach reasonable to use even when the trace gases originate from different sources. Based on observations, we find that the molar ratios of CO_2 and CH_4 were not always homogeneously mixed in the vertical direction downwind of Indianapolis. This is most likely due to the close proximity of our crosswind horizontal transects to various sources in the city (Cambaliza et al., 2014). Thus, for this analysis, we have employed the flux ratio approach rather than the concentration ratio method, as the former is reasonable to use whether or not the boundary layer is well mixed because it takes into consideration the determined total emission rate of the two greenhouse gases.

Table 3 and Figure 4 show the total CH_4 and CO_2 mass balance emission fluxes for several INFLUX flight experiments as well as the 2008–2009 Indianapolis mass balance experiment results from Mays et al. (2009). We also report the Hestia fossil fuel CO_2 flux in Table 3 corresponding to the flight dates and the mass balance CH_4/CO_2 emission flux ratios. We note that our CO_2 measurements include both fossil fuel and biogenic CO_2 with a potentially significant influence from biogenic CO_2 during the growing season. Thus to avoid biases in our results, we determine the average CH_4 to CO_2 flux ratio only for the non-growing season where the fluxes are believed to be purely anthropogenic in origin. This assumption is supported by radiocarbon measurements (^{14}C in CO_2) that show that the total CO_2 is mainly fossil-fuel derived during the non-growing season in Indianapolis (Turnbull et al., 2014, submitted). Using the flux ratios reported in Table 3 but not including the summer time results, i.e. 01 June, 30 June, and 12 July 2011 flight experiments, the mean flux ratio from 11 flight experiments is determined to be 0.0096 ± 0.0068 (1σ). Multiplying this figure by the average Hestia fossil-fuel citywide CO_2 emissions for the non-growing season (10059 moles s^{-1}), we obtain an average citywide CH_4 flux emission of 97 ± 68 (1σ) moles CH_4 s^{-1} for 11 flight experiments. For comparison, an orthogonal distance regression through the same data (Figure 4) results in a slope of 0.0096 ± 0.0034 (1σ), yielding an identical emission rate of 97 ± 34 (1σ) moles CH_4 s^{-1}. These results are statistically indistinguishable from the mean CH_4 flux directly obtained from the 2011 mass balance experiments (135 ± 58 moles s^{-1}) (P = 0.22 by t test).

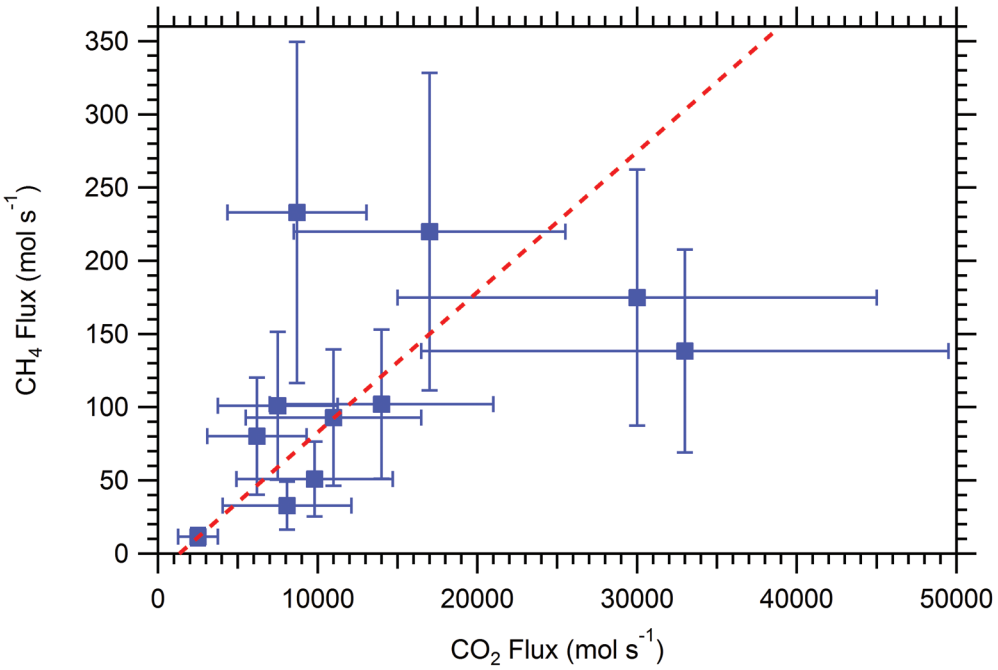

Figure 4

Citywide CH_4 flux versus CO_2 flux for 11 Indianapolis flight experiments summarized in Table 3.

Data presented do not include summertime estimates. The dotted red line represents the orthogonal distance regression with a slope of 0.0096 ± 0.0034 (1σ) mole CH_4 per mole CO_2 (Pearson r = 0.5), which yields a total citywide emission of 97 ± 34 (1σ) moles s^{-1} when multiplied by the mean Hestia emission flux (10059 moles s^{-1}) corresponding to the 11 flight experiments. The resulting intercept (-12.6 ± 18.3 (1σ) mole CH_4 s^{-1}) is not statistically different from zero.

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Identification of specific CH_4 sources

A noticeable feature in the horizontal transect distribution for July 12, 2011 (Figure 2) is the 50 to 100 ppb enhancement between -8 km to +2 km along the transect. We observed this distinct feature for all 2011 flight experiments considered in this analysis (see, for example, Cambaliza et al. (2014) for the 01 March, 29 April, and 01 June 2011 CH_4 horizontal transect distributions as well as Figure S1 for the 30 June 2011 flight experiment). During the 01 March 2011 flight experiment, we followed the plume upwind to the observed source region, the southwest side of Indianapolis, which is the industrial section of the city that is

Table 3. CO_2 emission fluxes estimated from the aircraft mass-balance method and bottom-up Hestia approach. Also reported are the mass balance CH_4 emission fluxes and the CH_4/CO_2 flux ratios[a]

Flight date	Mass balance CO_2 flux (mol s^{-1})	Mass balance CH_4 flux (mol s^{-1})	Hestia CO_2 fossil fuel flux (mol s^{-1})	CH_4/CO_2 flux ratio
28 Mar 2008	8080	33	11222	0.0041
02 Apr 2008	2500	12	9354	0.0047
14 Apr 2008	9800	51	8324	0.0052
15 Apr 2008	14000	102	9308	0.0073
21 Apr 2008	6200	80	6084	0.0129
23 Nov 2008	33000	140	7607	0.0042
20 Dec 2008	30000	170	11552	0.0058
07 Jan 2009	8700	230	12742	0.0268
01 Mar 2011	11000	93	12122	0.0085
29 Apr 2011	7500	101	10751	0.0135
01 Jun 2011[b]	26000	197	12134	0.0076
30 Jun 2011[b]	12000	85	8464	0.0071
12 July 2011[b]	49000	198	12221	0.0040
08 Nov 2012[c]	17000	220	11584	0.0129

[a]Uncertainties in the mass balance approach and Hestia county-level fossil fuel CO_2 flux are ± 50% and (-15%, +20% at 95% C.I.), respectively.

[b]Summertime mass balance flight experiments that were not included in the analysis in Figure 4 to avoid the potential contribution from biogenic CO_2 during the growing season.

[c]It was not possible to obtain the SSLF contribution from this INFLUX flight experiment, as the landfill plume was well-mixed with the city plume most likely due to the combined effect of relatively slower winds (2.9 m s^{-1}, 232°) and considerable downwind distance of the horizontal transects from the landfill (~40 km). Hence this INFLUX flight is used in Figure 4 but not included in the analysis in Table 2.

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home to the SSLF, a natural gas TRS, and the Belmont WWTP. Figure S2A shows the overall flight path for the 01 March 2011 flight experiment colored by the CH_4 concentrations, as well as a close-up view of the track following the plume upwind (Figure S2B). As we flew upwind (Figure S2B) and approached the southwest region of the city, particularly the landfill, we observed elevated concentrations of CH_4 (200 to 300 ppb above background concentrations). And as we passed the landfill and flew northwest of the solid waste facility (Figure S2A) away from the plume, the observed concentrations returned to background levels. By flying upwind and following the plume to the source, the identification of the local landfill area as the source was unambiguous. From that inflight investigation, we determined that the southwest side of the city is indeed the general area contributing to the observed elevated concentration in the downwind horizontal transect measurements.

To determine the location and identity of large CH_4 sources from the southwest side of the city observed from aircraft data, we conducted surface mobile measurements in Indianapolis. As previously stated, we conducted multiple plume traverses downwind of known sources by driving on roads that were roughly perpendicular to the prevailing wind direction. We also systematically surveyed the city for other CH_4 sources that may not be quantified in the inventories using the same driving approach, i.e., choosing roads that were nominally perpendicular to the wind direction. The upwind flight path in Figure S2 clearly shows that the landfill is a source of elevated methane. However, the landfill is right next to a WWTP and a TRS. All three sources appear to be a single strong source from a distance of at least 10 km during aircraft measurements. Hence, it was necessary to interrogate the sources using street-level measurements, as it is not possible to partition or resolve the CH_4 peaks to various potential sources using only the aircraft data. Figure 5A shows the CH_4 enhancements (concentrations above background) from one of five mobile surveys on Harding Street conducted on 21 Jan 2013 specifically targeting the southwest side of the city when the prevailing winds were from the west. From our extensive citywide ground sampling that involved driving upwind and downwind of potential sources, we consistently measured CH_4 enhancements downwind of SSLF and the TRS, for multiple wind directions, clearly indicating that the two most significant sources in the city's southwest side are these two sources (Figure 5), with no significant contribution from the wastewater treatment plant. However, we note the presence of shallow organic rich Devonian shale (Indiana Geological survey Bedrock Geology of Marion County, http://igs.indiana.edu/MarionCounty/BedrockGeology.cfm, accessed 25 April 2014) that is exposed at the Harding Street Quarry, which is located southeast of the landfill (Figure 5). The landfill and the quarry are both west of Harding Street, and are separated by the White River (Figure 5). A number of papers in the literature discuss the potential for biogenic CH_4 production from Devonian shales at shallow depths when the exposed organic rich shale is underwater and presumably in anaerobic condition (Martini et al., 1996; 1998; 2003). Thus, the Harding Street Quarry may potentially be a source of CH_4, as quarries tend to fill with water during extraction. This requires further investigation since the southwest section of the landfill overlaps with the northern area of the quarry. In the absence of emission verification from the quarry and the presence of a large CH_4 peak downwind of the landfill (Figure 5), we assume no significant emission from the quarry, as shown in Figure 5.

Determination of combined and individual contributions from SSLF and TRS

To quantify the contributions from SSLF and TRS to the citywide flux, we first determined their combined contribution from aircraft data by assuming that grid cells in the interpolated CH_4 matrix (Figure 2B) were attributable to these sources if the grid cell concentration was at least two standard deviations greater than the mean city CH_4 concentration, and correlated with back trajectory analysis to the SSLF/TRS sources. The mean city CH_4 concentration, for example, was 1897 ± 11 (1σ) ppb for the 12 July 2011 flight experiment. We applied this criterion to the five flight experiments considered in this analysis and found that the combined contributions from SSLF and TRS ranged from 19% to 47% of the total flux, with a mean contribution of 33 ± 10%, equivalent to 45 ± 14 (1σ) moles s^{-1} (Table 2). In this analysis, we used the mean city CH_4 concentration for the background.

To determine the individual contributions from SSLF and TRS, we measured the total enhancement (area under the curve in Figure 5B, ppm-degree) of the two distinct plumes from the surface mobile measurements (Figure 5B) and determined their relative contributions to the total enhancement. We then assumed that the relative contributions of these two sources were proportional to their relative contributions to the combined SSLF+TRS emission flux determined from aircraft measurements. However, the downwind distances of these two sources from Harding Street are not equal (~40 m for the TRS and ~1300 for SSLF), and thus there is differential dispersion affecting the two plumes. Because the dispersion of the plume is a function of downwind distance, we used a Gaussian plume model to apply a correction to the relative flux contribution of TRS by determining its effective plume area under the curve at 1300 m, the same distance as SSLF. We

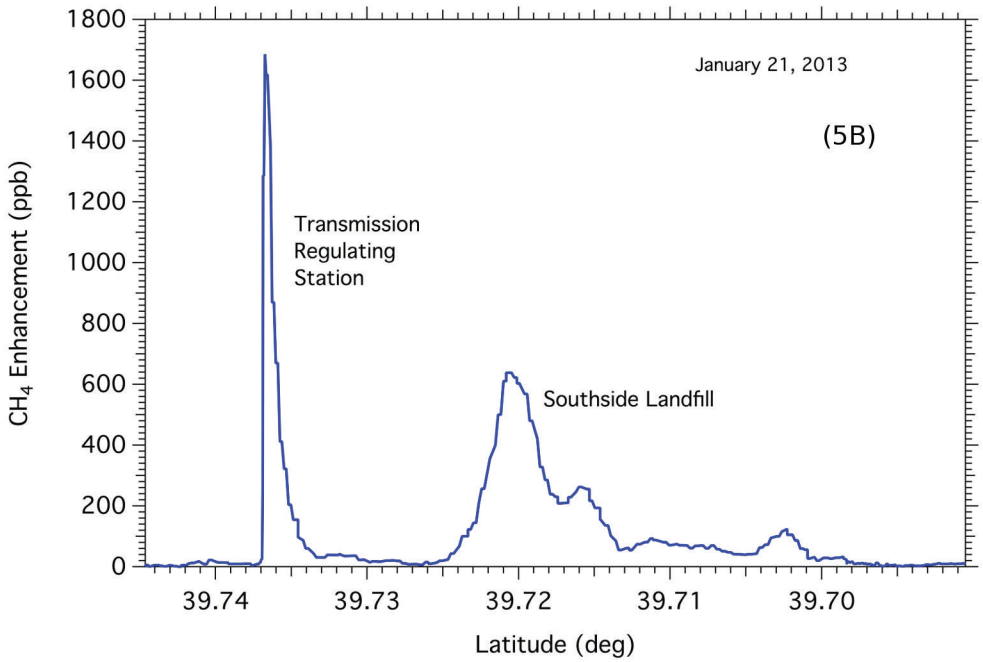

Figure 5 (A and B)

Observed CH_4 enhancements directly downwind of SSLF and a transmission regulating station on Harding St.

The CH_4 enhancement data were obtained during a surface mobile measurement on 21 January 2013. Prevailing winds were westerly.

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discuss in detail the derivation of the correction factor using the Gaussian plume model in the supplementary information (Text S1) and summarize in Table 4 the CH_4 flux contributions from SSLF and the natural gas TRS to the large enhancement from this section of the city. A total of five traverses from three sampling days (15 Oct 2012, 12 Nov 2012, and 21 Jan 2013) were used in our analysis (Table 4). We show in Table 4 the TRS CH_4 plume at downwind distances of 40 m and 1300 m. After applying the correction, we find that the average CH_4 emission flux from the TRS was 20 ± 17 (1σ) millimoles s^{-1}, a negligible quantity relative to that from the landfill. Thus, the observed excess CH_4 from aircraft data and the corresponding average flux of 45 ± 14 moles s^{-1} was almost entirely attributable to the landfill. This magnitude was about one-third of the whole citywide emission.

Table 4. CH_4 flux contributions from SSLF and a transmission regulating station (TRS) on Harding St. and Raymond St. intersection[a]

Date and traverse no.	Area SSLF at 1300m (ppb-deg)	Area TRS at 40m (ppb-deg)	Area TRS at 1300m (ppb-deg)	Total area at 1300m (ppb-deg)	Fractional contribution SSLF	Fractional contribution TRS	Flux SSLF (mol s^{-1})	Flux TRS (mol s^{-1})
21 Jan 2013, 1st	4.197	1.620	0.0022	4.199	0.9995	0.0005	44.976	0.024
21 Jan 2013, 2nd	4.274	0.629	0.0009	4.275	0.9998	0.0002	44.991	0.009
21 Jan 2013, 4th	5.459	1.462	0.0020	5.461	0.9996	0.0004	44.984	0.016
12 Nov 2012	1.559	1.247	0.0017	1.561	0.9989	0.0011	44.951	0.049
15 Oct 2012	4.178	0.295	0.0004	4.178	0.9999	9.6E-05	44.996	0.004
Average							44.980	0.020
Standard deviation							0.017	0.017

[a]A total of five traverses along Harding St. were used for the calculation: three out of four traverses on 12 Jan 2013 (third traverse was not long enough to sample the plumes from the two sources), and one traverse each on 12 Nov and 15 Oct 2012.

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Given that CH_4 is an important component of natural gas, it is informative to quantify the energy equivalent of 45 moles s^{-1} (equivalent to 1060 L s^{-1} assuming that the atmospheric pressure and temperature are 1013 mbars and 288 K, respectively), the mean landfill fugitive emissions determined from five flight experiments. The national average electric energy generated per unit of fuel used is 125 kWh per 28,300 L (1000 cubic ft) of natural gas (U.S. Energy Information Administration, http://www.eia.gov/tools/faqs/faq.cfm?id=667&t=6, accessed 18 October 2014). Given that the 2012 mean annual electricity consumption for an Indiana household was 11,960 kWh (U.S. Energy Information Administration, http://www.eia.gov/electricity/sales_revenue_price/xls/table5_a.xls, accessed 01 September 2014), a CH_4 emission of 45 ± 14 moles s^{-1} (assumed constant), when captured and converted to electric power energy, is equivalent to the electric power needs of 13,000 ± 4060 (1σ) households (see Text S1 for detailed calculation). We note that the 2012 electricity consumption of an Indiana household is higher than the US national average, which is 10,840 kWh (U.S. Energy Information Administration, http://www.eia.gov/tools/faqs/faq.cfm?id=97&t=3, accessed 01 September 2014).

Comparison of CH_4 emissions from SSLF with other Indiana landfills

The quantification of the emission flux from the landfill is challenging, given the fact that it is surrounded by other potential sources in the city. To assess the quality of our estimated CH_4 emission for SSLF (and hence, our approach to derive its magnitude), we compared its measured and modeled emissions with those of four other Indiana landfills that cover a range of landfill properties, both larger and smaller than SSLF. The four Indiana landfills are isolated solid waste facilities, the emissions from which can be more accurately quantified with the aircraft-based mass balance approach. We note that during the 03 May 2012 flight experiment downwind of NCLF, the wind was from the southwest and CH_4 plumes from seven dairy farms were also intercepted during measurement. We partitioned the total calculated CH_4 flux to remove the dairy component and isolated the contribution from the landfill (see Cambaliza et al., 2014, for detailed discussion). Table 1 summarizes the landfill site descriptions, dates of the aircraft-based mass-balance experiments, and the estimated CH_4 emissions. For this analysis, we use CALMIM, which simulates CH_4 emissions from daily, intermediate, and final site-specific landfill cover designs (Spokas and Bogner, 2011). Figure 6 shows a comparison of the landfill CH_4 emissions derived from the aircraft-based mass balance approach and the CALMIM monthly-averaged model results with and without soil microbial oxidation. Uncertainty in the mass balance derived emissions from small area/point sources (±30% as discussed in section 2.5 and in Cambaliza et al., 2014) was previously estimated from aircraft-based mass balance experiments designed to quantify the precision of the approach. In those experiments, emission fluxes from small area/point sources (CH_4 and CO_2 fluxes from two isolated landfills and a power generating station, respectively) were determined from separate flux calculations at multiple downwind distances (Cambaliza et al., 2014). Error bars in the CALMIM monthly-averaged results represent the standard deviation of the 10-minute time step modeled surface emissions. The model does not include non-diffusive emissions such as potential leakages from the gas extraction system, cover fissures, or edge leakages since current US landfill regulations require regular quarterly monitoring and remediation of such leaks. However, as we discussed in the Methods (*Comparison of measured and modeled emissions from Southside landfill with other Indiana landfills*), we did model daytime emissions from the working face with the assumption that this area overlies an older layer of cells with fully methanogenic waste. Differences between the CALMIM modeled emissions and mass balance estimates may arise from (1) non-diffusive emission contributions and (2) year-to-year differences in oxidation since CALMIM models "typical annual climate" using 30-year meteorological data. In Figure 6, we find that the observed CH_4 emissions are statistically indistinguishable from the CALMIM model results for both cases. The linear least squares fit forced through zero for the "without oxidation" and "with oxidation" cases are: y = 1.27

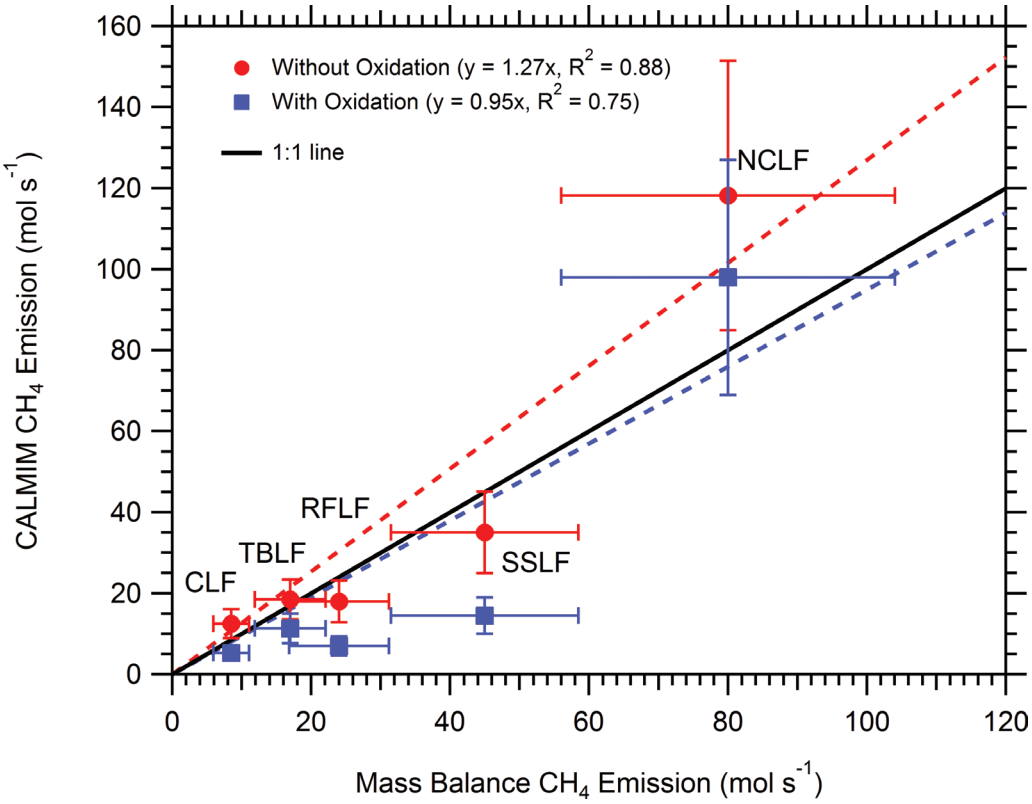

Figure 6

Linear least squares regression of the CALMIM monthly-averaged model results versus the mass balance CH_4 emission flux estimates for the five Indiana landfills.

Uncertainty in the mass balance estimates is ± 30%, and was previously determined from aircraft-based experiments designed to quantify the precision of the flux measurement approach for small area/point sources (Cambaliza et al., 2014). Error bars in the CALMIM monthly-averaged results represent the standard deviation of the 10-minute time step modeled surface emissions. Broken lines correspond to the linear least squares fit forced to zero for both cases.

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(±0.16)x (1σ) (r^2 = 0.88), and y = 0.95 (±0.20)x (1σ) (r^2 = 0.75), respectively. While the mass balance results for four sites were higher than the CALMIM-modeled results, the observations for the NCLF were lower than modeled by CALMIM. With respect to this apparent inconsistency, it is important to emphasize that the CALMIM-modeled results are conservatively-modeled "typical annual emissions" based on 0.5 degree climate models for site latitude/longitude, site-specific cover materials (with variable areas, composition, and thickness) and status of engineered gas recovery. In comparison, the mass balance field result captures a snapshot of emissions within the seasonal variability modeled by CALMIM. Moreover, in general, higher mass balance results compared with CALMIM can also be attributed to fugitive leakages not accounted for by diffusive emissions modeled by CALMIM. However, it is quite likely that the residuals reflect a random combined short-term variability in the emissions, and the precision of the measurement. With respect to expected seasonal variability for each site, Figure S3 shows the modeled monthly CALMIM emissions at each of the five landfill sites with and without oxidation, and with the individual monthly standard deviations. Without the NCLF data point in Figure 6, the two slopes are 0.82 and 0.36 (r^2 values of 0.8 and 0.28 for the "without oxidation" and "with oxidation" cases, respectively), respectively, with the SSLF data point essentially on the line. The "with" and "without" soil microbial oxidation cases represent the two baselines for CH_4 emissions through the landfill cover soil. We note that the climate variability affecting gaseous transport in individual site-specific cover materials is identically represented in both the oxidized and non-oxidized CALMIM runs, with the only difference being that oxidation is "turned off" for the non-oxidized analysis (Spokas et al., 2011). As stated in the methods section, soil gas CH_4 transport is explicitly modeled according to climate (soil moisture and temperature) for individual 10-min time steps and 2.5 cm depth increments over the typical annual cycle. The assumptions for oxidation are that, for individual cover materials, oxidation rates are scaled to maximum oxidation rates for a given temperature and pressure (Spokas and Bogner, 2011). Given the range of uncertainties between the modeling and measurement approaches, the relatively high linearity between the two methods suggests that the measured emission rate from the South Side Landfill is not out of line, for either regression estimate (Figure 6). This, and the ground transect method, leads us to conclude that our SSLF flux determination is not appreciably biased by other significant sources nearby, and that the uncertainty is comparable to that for the other landfills.

CH_4 emissions from the natural gas distribution system

While the SSLF is an important small area source, the majority (~67%) of the CH_4 emitted in Indianapolis is from other sources. Mays et al. (2009) previously determined, by relating the whole city CH_4 flux to traffic flow data, that mobile combustion sources are insignificant, and as stated above, as is the wastewater treatment plant. Our systematic surface-based survey of the city revealed no identifiable source of CH_4 other

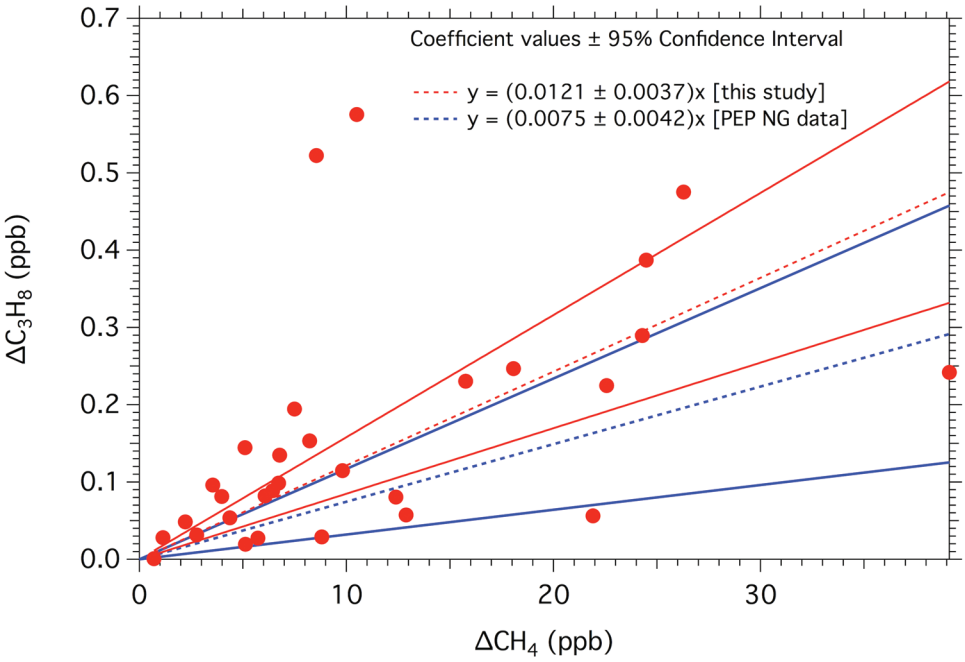

Figure 7

Observed propane versus methane enhancement from aircraft flask measurements (Pearson r = 0.51).

Flask data were obtained downwind of the city but outside the landfill plume for several flight experiments. Also shown is the mean ratio of propane to methane from the processed natural gas (NG) composition data reported by Panhandle Eastern Pipeline (PEP) corresponding to our flight dates. The solid lines correspond to the upper and lower 95% confidence limits.

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than leaks in the natural gas distribution system. Thus, we hypothesize that in fact all of the non-landfill CH_4 sources in the city derive from leakage from the natural gas distribution system. To test this hypothesis, we compare the $(C_3H_8)/CH_4$ ratio obtained from analysis of our aircraft flask samples, acquired within the CBL downwind of the city but only for samples outside the landfill plume (Figure 7). We plot the enhancement in each hydrocarbon (Figure 7) relative to observed background concentrations taken from the flask with the lowest C_3H_8 and CH_4 concentrations. Our C_3H_8 data are corrected for the very small contribution from automobile emissions, using measurements of C_2H_2 in the same flask samples and C_3H_8 and C_2H_2 emission data directly obtained from light-duty gasoline vehicles tested using a chassis dynamometer (Pang et al., 2014). We calculated the corresponding C_3H_8:C_2H_2 emission ratio (0.045 moles C_3H_8 per mole C_2H_2, see Text S1 for calculation) and applied this quantity to our flask data to obtain the vehicle combustion contribution (~7% of total C_3H_8), and subsequently, the natural gas contribution assuming that the total C_3H_8 obtained in flask samples is comprised essentially of the natural gas and vehicle emission contributions, as there are no other known significant sources. We note that the slope of the linear least squares regression of C_3H_8 versus C_2H_2 enhancements obtained from Indianapolis (2.24 ± 0.84 ppb C_3H_8 /ppb C_2H_2) is not statistically significantly different from the emission ratios at the 95% CL obtained from Los Angeles (1.91 ± 1.32 ppb C_3H_8 /ppb C_2H_2) (Borbon et al., 2013), and from New York and Boston cities (2.19 ± 1.29 ppb C_3H_8 /ppb C_2H_2) (Warneke et al., 2007) (Figure S4), suggesting similar sources of C_3H_8 and C_2H_2 from these urban environments.

We compared the slope of the C_3H_8 to CH_4 regression (Figure 7) with the mean C_3H_8 to CH_4 ratio from processed natural gas composition data reported by Panhandle Eastern Pipeline (PEP), the primary supplier of natural gas to Indianapolis, for their four transmission segments corresponding to the experimental flight dates (http://peplmessenger.energytransfer.com/ipost/PEPL/gas-quality/daily-average-quality-info-by-date, accessed 02 December 2013). We find that the slope of the C_3H_8 versus CH_4 flask data (0.0121 ± 0.0037) is not significantly different from the mean C_3H_8/CH_4 ratio from natural gas composition data from PEP (0.00745 ± 0.00245) at the 95% confidence level. This result suggests that all of the remainder, i.e. 67% of the total emission of CH_4 for the city of Indianapolis, derives from the natural gas distribution system, with an average emission rate of 90 ± 54 (1 σ) moles s^{-1}. Indeed, our mobile surface lab measurements found leaks that are associated with the natural gas distribution system ranging from ≤ 500 ppb to ≥ 10,000 ppb enhancements above background. This natural gas leakage flux (90 ± 54 moles s^{-1}) is an electric power consumption (3.1±(1.9) × 10^8 kWh yr^{-1}) equivalent to that of 26,000 ± 16,000 households.

Indianapolis per capita CH_4 emission rate

As mentioned above, the mean total CH_4 emission for the city determined from several 2011 flight experiments is 135 ± 58 moles s^{-1} (7800 ± 3300 kg hr^{-1}). This emission rate yields a per capita CH_4 emission of 165 ± 71 (1σ) μmoles $person^{-1}$ s^{-1} (9.5 ± 4.1 g $person^{-1}$ hr^{-1}) given that the 2010 population of Indianapolis was 820,000. However, SSLF (http://www.ssidelandfill.com/, accessed 20 September 2014) serves other nearby towns outside Indianapolis that comprise a population of ~240,000 in 2010. The list of these towns

and their 2010 population are provided in Table S2. Thus, the mean landfill emission flux of 45 moles s^{-1} can be apportioned accordingly: 77% contribution from Indianapolis, which corresponds to an emission rate of ~35 moles s^{-1}, and 23% from other towns yielding a corresponding emission of ~10 moles s^{-1}. This will then impact the whole-city CH_4 per capita emission and slightly reduces the figure to 152 ± 65 (1σ) μmoles person^{-1} s^{-1} (9 ± 4 g person^{-1} hr^{-1}). For comparison, the CH_4 emission from the Los Angeles megacity was estimated to be 750 to 870 moles s^{-1} (43,000 to 50,000 kg hr^{-1}) from 2007 – 2010 (Wunch et al., 2009; Hsu et al., 2010; Wennberg et al., 2012). Given that the population of Los Angeles in 2010 was approximately 3.8 million, this yields a per capita emission of 200 to 230 μmoles person^{-1} s^{-1} (12 to 13 g person^{-1} hr^{-1}). It is interesting to note that while the CH_4 emission rate of Los Angeles is about a factor of 6 larger than Indianapolis, the per capita emission rate from these two urban environments are not statistically different from each other.

Conclusions

We quantified the CH_4 emission from the city of Indianapolis and found a mean of 135 ± 58 moles s^{-1} from several mass balance flight experiments in 2011. We find that the SSLF contributes a mean emission rate of 45 ± 14 moles s^{-1}, which is equivalent to about one-third of the citywide emission. Because this only open landfill inside the city also serves nearby towns surrounding the city, the Indianapolis per capita emission is calculated as 152 ± 65 μmoles person^{-1} s^{-1}, equivalent to 4800 moles CH_4 person^{-1} year-^{1} (77 kg CH_4 person^{-1} year^{-1}). This figure is less than the 2010 national per capita anthropogenic CH_4 emission (~ 5700 moles CH_4 person^{-1} year^{-1} equivalent to 91 kg CH_4 person^{-1} year^{-1}), given that the US anthropogenic CH_4 emission for 2010 is 28.2 Tg CH_4 (US EPA, Inventory of Greenhouse Gas Emissions and Sinks: 1990 – 2011) and the 2010 US population was ~310 million (US Census Bureau, US and World Population Clock, http://www.census.gov/popclock/, accessed 25 September 2013). For comparison, the approximate global anthropogenic per capita emission is 2990 moles CH_4 person^{-1} yr^{-1} (48 kg CH_4 person^{-1} yr^{-1}), estimated from the global anthropogenic CH_4 emission of ~340 Tg CH_4 yr^{-1} (Dlugokencky et al., 2011) and the world population of 7.1 billion (Smith, 2011). Thus, on a per capita basis, CH_4 emission from Indianapolis is less than the national figure but significantly larger than the global number.

The C_3H_8 versus CH_4 regression suggests that all of the remainder, i.e. 67% of the citywide CH_4 emission, derives from leakages in the natural gas distribution system. Given that CH_4 is a potent greenhouse gas and an important energy commodity, there is clearly a need to identify and quantify the individual sources contributing to that 90 ± 54 moles s^{-1} derived from natural gas sources (e.g. transmission regulating stations, distribution lines, residential leaks). To this end, a comprehensive surface mobile survey of the city is needed, a necessary first step to identify specific natural gas sources. In addition to this qualitative method, complementary approaches such as the tracer-correlation method, as well as transport and dispersion models in combination with street level mole fraction and meteorological data, are needed to accurately quantify contributions from sources such as transmission regulating stations, closed landfills, and wastewater treatment plants that have modest source strengths relative to the operating solid waste facility. Component level understanding can then lead to appropriate inventory development and emissions mitigation approaches. These efforts are currently works in progress for the city of Indianapolis and will enable the development of a set of source-specific prior emission fluxes useful for top-down whole-city inverse modeling efforts.

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Contributions

- Conception and design: MOLC, PBS, DRC, BS
- Acquisition of data: MOLC, PBS, OES, TNL, BM, CM, AH, KM, CO, KRG, EC, JT
- Analysis and interpretation of data: MOLC, JB, KS, DRC, CS, SAM, BRM, KRG
- Drafting and/or revising the article: MOLC, PBS, DRC, JB, CS, AK, JT, SAM, KP, TL, KD, JW, NM, SR
- Final approval of the version to be published: MOLC, PBS

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Competing interests

The authors have declared no competing interests.

Supplementary material

- **Figure S1. Downwind CH_4 horizontal transect distribution as a function of altitude for 30 June 2011.**
  The black broken lines represent the boundary of the project city width on the downwind horizontal transects. The –x to +x city limits are -38 km to +21 km.

- **Figure S2. Measurement flight path on 01 March 2011 color coded by the observed methane concentration.**
  The plot in (A) shows the entire flight path including the downwind horizontal transpects while the image in (B) shows the close up view of a section of the flight path as the airplane flew upwind to interrogate the source of observed enhancement in the downwind horizontal transect.

- **Figure S3. CALMIM modeled monthly CH_4 emissions with standard deviations for the five Indiana landfills.**
  The seasonal trends of the modeled CH_4 emissions for both the oxidized and un-oxidized cases are shown for (A) CLF, (B) NCLF, (C) RFLF, (D) SSLF, and (E) TBLF.

- **Figure S4. Observed propane versus acetylene enhancements obtained from flask measurements downwind of the city of Indianapolis.**
  Also shown are the C_3H_8:C_2H_2 emission ratios from Los Angeles city (Borbon et al., 2013), and from New York City and Boston (Warneke et al., 2007). These emission ratios are not statistically significantly different from the slope of the regression line obtained for Indianapolis at the 95% confidence interval. The solid lines represent the lower and upper 95% confidence limits.

- **Table S1. Summary of CALMIM 5.4 inputs. To model emissions from the daytime open face, we assumed a final cover profile (as underlying waste was fully methanogenic) overlain by 6″ sand. Nighttime daily cover emissions as shown in table.**

- **Table S2. List of towns surrounding Indianapolis that are also served by SSLF.**

- **Text S1. Quantification and source apportionment of the methane emission flux from the city of Indianapolis**

Data accessibility statement

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