Background Heterogeneity and Other Uncertainties in Estimating Urban Methane Flux: Results from the Indianapolis Flux (INFLUX) Experiment

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Abstract

As natural gas extraction and use continues to increase, the need to quantify emissions of methane (CH₄), a powerful greenhouse gas, has grown. Large discrepancies in Indianapolis CH₄ emissions have been observed when comparing inventory, aircraft mass-balance, and tower inverse modeling estimates. Four years of continuous CH₄ mole fraction observations from a network of nine tower-based cavity ring-down spectrometers measuring atmospheric CH₄ mole fractions at 39 to 136 m above ground as part of the Indianapolis Flux Experiment (INFLUX) are utilized to investigate four possible reasons for the abovementioned inconsistencies: (1) differences in definition of the city domain, (2) a highly temporally variable and spatially non-uniform CH₄ background, (3) temporal variability in CH₄ emissions, and (4) the presence of unknown CH₄ sources. Reducing the Indianapolis urban domain size to be consistent with the inventory domain size decreases the CH₄ emission estimation of the inverse modeling methodology by about 35% and thereby lessens the discrepancy by bringing total city flux within an error range of one of the inventories. Nevertheless, the inverse modeling estimate still remains about 40% higher than the inventory value. Hourly urban background CH₄ mole fractions are shown to be heterogeneous and temporally variable. Statistically significant, long-term biases in background mole fractions of 2-
5 ppb are found from single point observations from most wind directions. Random errors in single point background mole fractions observed for a few hours are 20-30 ppb, but decrease substantially when data are averaged over multiple days. Boundary layer budget estimates suggest that Indianapolis CH$_4$ emissions did not change significantly when comparing 2014 to 2016. However, it appears that CH$_4$ emissions may follow a diurnal cycle with daytime emissions (12-16 LST) approximately twice as large as nighttime emissions (20-5 LST). The strongest CH$_4$ source in Indianapolis is the South Side Landfill. Other point sources, perhaps leaks from the natural gas distribution system, are localized and transient, and do not appear to be a consistently large source of CH$_4$ emissions in Indianapolis. Long-term averaging, spatially-extensive upwind mole fraction observations, mesoscale atmospheric modeling of the regional emissions environment, and careful treatment of the times of day and areal representation of emission estimates is recommended for precise and accurate quantification of urban CH$_4$ emissions.

1 Introduction

From the beginning of the Industrial Revolution to 2011, atmospheric methane (CH$_4$) mole fractions increased by a factor of 2.5 due to anthropogenic processes such as fossil fuel production, waste management, and agricultural activities (Ciais et al., 2013). The increase in CH$_4$ is a concern as it is a potent greenhouse gas (GHG) with a global warming potential 28-34 times greater than that of CO$_2$ over a period of 100 years (Myhre et al., 2013). The magnitudes of component CH$_4$ sources, and the causes of variability in the global CH$_4$ budget, however, are not well understood although there is some evidence that biogenic emissions may play an important role in the recent CH$_4$ increases (Nisbet et al., 2016; Saunois et al., 2016). Improved understanding of CH$_4$ emissions is needed (National Academies of Sciences and Medicine, 2018).
In particular, the estimates of continental U.S. anthropogenic CH₄ emissions disagree. Inventories from Environment Protection Agency (EPA) and Emissions Database for Global Atmospheric Research (EDGAR) in 2008 reported emission values of 19.6 and 22.1 TgC y⁻¹ (Miller et al., 2013). However, top-down methodologies using aircraft and inverse modeling framework found emission values of 32.4 ± 4.5 TgC y⁻¹ for 2004 and 33.4 ± 1.4 TgC y⁻¹ for 2007-2008 respectively (Kort et al., 2008; Miller et al., 2013). Underestimation of natural gas (NG) production and agricultural sources are possible reasons for this disagreement (Miller et al., 2013; Brandt et al., 2014; Jeong et al., 2014). Efforts to reconcile GHGs emissions estimates using atmospheric methods and inventory assessment have succeeded (Schuh et al., 2013; Zavala-Araiza et al., 2015; Turnbull et al., 2019) when careful attention is given to the details of each method, and targeted atmospheric data are available. A recent synthesis of emissions from the U.S. NG supply chain demonstrated similar success and concluded that current inventory estimates of emissions from U.S. NG production are too low and that emission from NG distribution is one of the greatest remaining sources of uncertainty in the natural gas supply chain (Alvarez et al., 2018).

Due to the uncertainties in CH₄ emissions from NG distribution it is natural that urban emissions are of interest as well. For example, studies indicate that ~60-100% of Boston CH₄ emissions are attributable to the NG distribution system (McKain et al., 2015; Hendrick et al., 2016). Recent studies of urban CH₄ emissions indicate that the California Air Resources Board (CARB) inventory tends to underestimate the actual CH₄ urban fluxes possibly due to fugitive emissions that result from the large NG infrastructures common to the urban environments (Wunch et al., 2009; Jeong et al., 2016; Jeong et al., 2017). The accuracy and precision of atmospheric estimates of urban CH₄ emissions are limited by available atmospheric observations (Townsend-Small et al., 2012), potential source magnitude variability with time (Jackson et al., 2014; Lamb...
et al., 2016), errors in atmospheric transport modeling (Hendrick et al., 2016; Deng et al., 2017; Sarmiento et al., 2017;), and complexity in atmospheric background conditions (Cambaliza et al., 2014; Heimburger et al., 2017). In this work, detailed analysis of urban CH$_4$ mole fractions is performed in the city of Indianapolis to better understand the aforementioned uncertainties of urban CH$_4$ emissions.

The Indianapolis Flux Experiment (INFLUX; Davis et al., 2017) is a testbed for improving quantification of urban GHGs emissions and their variability in space and time. INFLUX (http://influx.psu.edu) is located in Indianapolis partly because of its isolation from other urban centers and the flat Midwestern terrain. It includes a very dense GHGs monitoring network, comprised of in situ aircraft measurements (Heimburger et al., 2017; Cambaliza et al., 2014), in situ observations from communications towers using cavity ring-down spectroscopy (Richardson et al., 2017; Miles et al., 2017), and automated flask sampling systems for quantification of a wide variety of trace gases (Turnbull et al., 2015). Meteorological sensors include a Doppler lidar providing continuous boundary layer depth and wind profiles, and tower-based eddy covariance measurements of the fluxes of momentum, sensible and latent heat (Sarmiento et al., 2017). The network is well suited for emissions estimates using top-down methods such as tower-based inverse modeling (Lauvaux et al., 2016) and aircraft mass balance estimates (Cambaliza et al., 2015).

Recently Lamb et al. (2016) compared Indianapolis CH$_4$ emissions estimates from inventory, aircraft mass balances, and inverse modeling. The results revealed large mean differences among the city fluxes estimated from these methods (Fig. 1). In general, the inventory methods arrived at lower estimates of emissions compared to the atmospheric, or top-down approaches. CH$_4$ fluxes calculated using the aircraft mass balance technique varied considerably
between flights, more than would be expected from propagation of errors of the component measurements (Cambaliza et al., 2014; Lamb et al., 2016). The atmospheric inverse estimate was significantly higher than the inventory and some of the aircraft-derived values.

Biogenic emissions from the city are dominated by a landfill close to downtown, and these emissions are thought to be fairly well known. Uncertainty in emissions is driven by the uncertainty in thermogenic emissions, which are hypothesized to emerge largely from the NG distribution system (Mays et al., 2009; Cambaliza et al., 2015; Lamb et al., 2016). This uncertainty has not yet been resolved. In this study, we explore potential explanations for the discrepancies in CH$_4$ emissions estimates from Indianapolis and posit methods and recommendations for the study of CH$_4$ emissions from other urban centers.

We examine four different potential explanations for the CH$_4$ flux discrepancies reported in Lamb et al. (2016): (1) inconsistent geographic boundaries, (2) heterogeneity in the urban-scale CH$_4$ background, (3) temporal variability in urban emissions, and (4) CH$_4$ sources that are not accounted for in the inventories. Well-calibrated CH$_4$ sensors on the INFLUX tower network (Miles et al., 2017) collected continuous CH$_4$ observations from 2013 to 2016 and provide a unique opportunity to explore these issues.

2 Methods

2.1 Experimental site

This study uses data from a tower-based GHG observational network located in the city and surrounding suburbs of Indianapolis, Indiana, in the Midwestern U.S. Prior studies have used varying definitions for the region of Indianapolis (Cambaliza et al., 2015, Lamb et al., 2016).
this work, we follow Gurney et al. (2012) and define Indianapolis as the area of Marion County. The flat terrain of the region simplifies interpretation of the atmospheric transport. The land-surface heterogeneity inherent in the urban environment (building roughness, spatial variations in the surface energy balance) do have a modest influence on the flow within the city and the boundary layer depth difference between the urban and rural areas (Sarmiento et al., 2017).

2.2 INFLUX tower network

The continuous GHG measurements from INFLUX are described in detail in Richardson et al. (2017). The measurements were made using wavelength-scanned cavity ring down spectrometers (CRDS, Picarro, Inc., models G2301, G2302, G2401, and G1301), installed at the base of existing communications towers, with sampling tubes secured as high as possible on each tower (39 – 136 m above ground level (AGL), Miles et al., (2017)). A few towers also included measurements at 10 m AGL and one or two intermediate levels. While INFLUX tower in-situ measurements began in September 2010, here we focus on the CH₄ measurements from 2013 – 2016. From June through December 2012, there were two or three towers with operational CH₄ measurements. By July 2013, five towers included measurements of CH₄, and throughout the majority of the years 2015 – 2016 there were eight INFLUX towers with CH₄ measurements (Fig. 2).

By May 2013, the inflow to all CRDS instruments was dried. Prior to deployment and following any manufacturer repairs, the instruments were calibrated for slope and offset in the Pennsylvania State University calibration laboratory (Richardson et al., 2017) using three to five NOAA-calibrated tanks. At each site, one or two NOAA-calibrated tanks were sampled daily for 10 min as field offset calibration points. In this study we used hourly means of CH₄, which were...
reported on the WMO X2004A scale. Flask to in-situ comparisons and round-robin style testing indicated compatibility across the tower network of 0.6 ppb CH$_4$ (Richardson et al., 2017).

2.3 Weather data

Wind data was measured at the Indianapolis International Airport (KIND), Eagle Creek Airpark (KEYE), and Shelbyville Municipal Airport (KGEZ). The data used are hourly values from the Integrated Surface Dataset (ISD) (https://www.ncdc.noaa.gov/isd) and 5-minute values directly from the Automated Surface Observing System (ASOS). A complete description of ASOS stations is available at http://www.nws.noaa.gov/asos/pdfs/aum-toc.pdf. The accuracy of the wind speed is ±1 m/s or 5% (whichever is greater) and the accuracy of the wind direction is 5 degrees when the wind speed is ≥ 2.6 m/s. The anemometer is located about 10 meters AGL. The wind data reported in ISD are given for a single point in time recorded within the last 10 minutes of an hour and are closest to the value at the top of the hour.

The planetary boundary layer height (BLH) was determined from a Doppler lidar deployed in Lawrence, IN, about 15 km to the northeast of downtown. The lidar is a Halo Streamline unit, which was upgraded to have extended range capabilities in January 2016. The lidar continuously performs a sequence of conical, vertical-slice, and staring scans to measure profiles of the mean wind, turbulence, and relative aerosol backscatter. All of these measurements are combined using a fuzzy-logic technique to automatically determine the BLH continuously every 20-min (Bonin et al., 2018). The BLH is primarily determined from the turbulence measurements, but the wind and aerosol profiles are also used to refine the BLH estimate. The BLHs are assigned quality-control flags that can be used to identify times when the BLH is unreliable, such as when the air is exceptionally clean, the BLH is below a minimum detectable height, or clouds and fog that
attenuate the lidar signal exist. Additional details about the algorithm and the lidar operation for
the INFLUX project are provided in Bonin et al. (2018). Doppler lidar measurements are available
at https://www.esrl.noaa.gov/csd/projects/influx/.

2.4 CH₄ Sources

Only a few known CH₄ point sources exist within Indianapolis (Cambaliza et al., 2015, Lamb et
al., 2016). The Southside Landfill (SSLF), located near the center of the city, is the largest point
source in the city with emissions of about 28-45 mol/s, accounting for 22% to 63% of total Marion
County CH₄ emissions (Cambaliza et al., 2015; Maasakkers et al., 2016; Lamb et al., 2016). Other
city point sources are comparatively small; the wastewater treatment facility located near SSLF
contributes approximately 4-10% to city CH₄ totals or about 3-7 mol/s, and the transmission-
distribution transfer station at Panhandle Eastern Pipeline (also known as a city gate and further in
this study abbreviated as PEP) is estimated to be about 0.5-1% or 1 mol/s. The remaining CH₄
sources, mainly from NG and livestock, are considered to be diffuse sources and are not well
known. Potential sources of emissions related to NG activities include gas regulation meters,
emissions from transmission and storage, and Compressed Natural Gas (CNG) fleets. These
diffuse NG sources account for 21-69% or 20-64 mol/s of the city emissions (Cambaliza et al.,
2015; Maasakkers et. al. 2016; Lamb et al., 2016). Livestock emissions for Marion County are
estimated to be around 3% or 1.5 mol/s.

2.4 Urban methane background

Both aircraft mass balance and inverse modeling methodologies rely on the accurate estimation of
the urban CH₄ enhancement relative to the urban CH₄ background in order to produce a reliable
flux estimate (Cambaliza et al., 2014; Lamb et al., 2016). The CH₄ mole fraction enhancement is defined as,

\[ C_{\text{enhancement}} = C_{\text{downwind}} - C_{bg} \]  

(1)

where \( C_{\text{downwind}} \) is the CH₄ mole fraction measured downwind of the source and \( C_{bg} \) is the CH₄ background mole fraction, which can be measured upwind of the source, but this is not necessary.

Background, as defined in this body of literature, is a mole fraction measurement that does not contain the influence of the source of interest, but which is measured simultaneously. Because choosing the background involves a degree of subjectivity (Cambaliza et al., 2014; Heimburger et al., 2017) we consider how this choice may influence emission estimates and introduce error, both random and systematic, using data from the INFLUX tower network.

Using tower network data from November, 2014 through the end of 2016, two CH₄ backgrounds are generated based on two different criteria. Both criteria identify a tower suitable to serve as a background for each of the eight wind directions (N, NE, E, SE, S, SW, W, NW), where an arc of 45° represents a direction (e.g. winds from N are between 337.5° and 22.5°).

Criterion 1 is based on the concept that the lowest CH₄ mole fraction measured at any given time is not affected by the city sources and therefore is a viable approximation of the background methane mole fractions outside of the city (Miles et al., 2017; Lauvaux et al., 2016). Given this assumption, the tower with the lowest median of the CH₄ enhancement distribution (calculated by assuming the lowest measurement among all towers at a given hour as a background) for each of the wind directions over the November, 2014 through December, 2016 time period is chosen as a background site (Miles et al., 2017). Criterion 2 requires that the tower is outside of Marion County (outside of the city boundaries) and is not downwind of any known regional CH₄ source (Fig. 3). For some wind directions, there are multiple towers that could qualify as a background;
we pick towers in such a manner that they are different for each criterion given a wind direction in order to calculate the error associated with the use of different but acceptable backgrounds. The towers used for both criteria and for each of the eight wind directions are displayed in Table 1. Quantifying differences between these two backgrounds allows for an opportunity to better understand the degree of uncertainty that exists in the Indianapolis background atmosphere.

2.5 Frequency and bivariate polar plots

Frequency and bivariate polar plots are used in this work to gain more knowledge regarding CH$_4$ background variability based on criteria 1 and 2, and to identify sources located within the city. To generate these polar plots, we use the openair package (from R programming language) created specifically for air quality data analysis (Carslaw and Ropkins, 2012). Bivariate and frequency polar plots indicate the variability of a pollutant concentration at a receptor (such as an observational tower) as a function of wind speed and wind direction, preferably measured at the location of the receptor or within several kilometers of the receptor. The frequency polar plot is generated by partitioning the CH$_4$ hourly data into the wind speed and direction bins of 1 m s$^{-1}$ and 10° respectively. To generate bivariate polar plots, wind components $u$ and $v$ are calculated for hourly CH$_4$ concentration values, which are fitted to a surface using a Generalized Additive Model (GAM) framework in the following way,

$$\sqrt{C} = \beta + s(u, v) + \epsilon$$

where $C$ is the CH$_4$ mole fraction transformed by a square root to improve model diagnostics such as a distribution of residuals, $\beta$ is mean of the response, $s$ is the isotropic smoothing function of the wind components $u$ and $v$, and $\epsilon$ is the residual. For more details on the model see Carslaw and Beevers (2013).
2.6 Temporal Variability

Temporal variability may play an important role in the quantification of urban methane emissions. Lamb et al. (2016) suggested that temporal variability may partially explain the differences among CH₄ flux estimates shown in Fig. 1. If temporal variability of CH₄ emissions exists within the city, disagreements in the CH₄ flux between studies could be attributed to differences in their sampling period. Because the INFLUX tower data at Indianapolis contain measurements at all hours of the day over multiple years, we can utilize this dataset to better understand the temporal variability in methane emissions in the city.

We apply a simplified atmospheric boundary layer budget, not to estimate precisely the actual city emissions, but rather to evaluate temporal variability of the emissions. We begin by assuming CH₄ emissions \( Q_a \) (mass per unit time per unit area) are not chemically active and are constant over a distance \( \Delta x \) spanning a significant portion of the city. The next assumption is that a CH₄ plume measured upwind of the city is well mixed within a layer of depth \( z_l \). We treat wind speed \( u \) as constant within the layer for every hour considered. Given the above-mentioned assumptions we can write a continuity equation describing mass conservation of CH₄ concentration \( C \) within a box in the following fashion,

\[
\Delta x z_l \frac{\partial C}{\partial t} = \Delta x Q_a + uz_l(C_b - C) + \Delta x \frac{\partial z_l}{\partial t}(C_a - C)
\]

where \( C_b \) is the CH₄ mole fraction upwind of the city (or background), and \( C_a \) is the CH₄ concentration above the mixed layer (Hanna et al., 1982; Arya, 1999; Hiller et al., 2014). The left hand of the equation represents the change in CH₄ concentration with time, \( \Delta x Q_a \) denotes a constant CH₄ source over the distance \( \Delta x \), \( uz_l(C_b - C) \) indicates a change of CH₄ concentration due to horizontal advection, and finally \( \Delta x \frac{\partial z_l}{\partial t}(C_a - C) \) term accounts for the vertical advection.
and encroachment processes that result from changing mixed layer height. By assuming steady state conditions ($\frac{\partial C}{\partial t} = 0$ and $\frac{\partial z_t}{\partial t} = 0$), the equation can be simplified to

$$Q_a = \frac{u z_t (C - C_b)}{\Delta x}$$

We use equation 4 to estimate hourly CH$_4$ emissions ($Q_a$) from Indianapolis (see assumptions in the paragraph below), given hourly data of $z_t$ from the lidar positioned in the city, wind speed from the local weather stations, and upwind ($C_b$) and downwind ($C$) CH$_4$ concentrations measured at towers 1, 8, and 13 (depending on wind direction) using data from heights of 40 m, 41 m, and 87 m respectively (see Fig. 3).

The CH$_4$ concentrations are derived from CH$_4$ mole fractions by approximating average molar density of dry air (in mol m$^{-3}$) within the boundary layer for every hour of the day, where variability of pressure with altitude is calculated using barometric formula and it is assumed that temperature decreases with altitude by 6.5 K per kilometer. The hourly surface data for pressure and temperature is taken from KIND weather station. The difference between concentrations ($C - C_b$) is instantaneous and not lagged, where $C_b$ represents air parcel entering the city and $C$ represents the same air parcel exiting the city (for more details see Turnbull et al., 2015). The CH$_4$ enhancements ($C - C_b$) are estimated for daytime by averaging observations spanning 12-16 LST and for nighttime by averaging observations spanning 20-5 LST. These time periods are based on lidar estimations of when on average $z_t$ varies the least. The day and night were required to contain at least 3 and 9 hourly CH$_4$ values respectively for averaging to occur, otherwise the day/night is eliminated. Observations when $z_t$ is below 100 m are not used to avoid the cases when measurements from towers may be above the boundary layer. In order to better achieve the assumption that the boundary layer is fully mixed (especially at night), all hours with wind speeds below 4 m/s are eliminated (Van De Wiel., 2012). To approximate the emissions of the whole city
we need to know the approximate area of the city and the distance over which the plume is affected by the city CH$_4$ sources. The area of the city is about 1024 km$^2$ (the area of Marion County) and the length that plume traverses when it is over the city ranges from 32 to 35 km depending on which downwind tower is used. We assume that CH$_4$ measurements at towers 8 and 13 are representative of a vertically well-mixed city plume as the towers are located outside of the city boundaries and allow for sufficient vertical mixing to occur. For S and SW wind directions tower 8 observations are used to represent downwind conditions with background observations coming from towers 1 and 13, respectively (based on Criterion 1 shown in Table 1). For W wind direction, tower 13 observations represent the downwind with background obtained from tower 1. The wind direction is required to be sustained for at least 2 hours, otherwise the data point is eliminated.

3 Results and discussion

3.1 City Boundaries

A significant portion of CH$_4$ emissions across the U.S. can be characterized by numerous large point sources scattered throughout the country rather than by broad areas of smaller enhancements (Maasakkers et al., 2016). Because of this, the total emissions for a given domain can be very sensitive to how that domain is defined. A small increase or a decrease in the domain area could add or remove a large point source and significantly impact the total emissions defined within the domain. This issue can be observed in prior studies of CH$_4$ emissions in Indianapolis, described below.

In Fig. 3, two possible domains are identified (Lamb et al., 2016; Lauvaux et al., 2016) that could be used for the evaluation of Indianapolis CH$_4$ emissions. The first domain is the whole area shown in the figure enclosing both Indianapolis and places that lie outside of its boundaries. The second domain is Marion County outlined with a green dashed line. It is assumed here that this
domain is much more representative of the actual Indianapolis municipal boundaries as this area encompasses the majority of the urban development associated with the city of Indianapolis (Gurney et al., 2012). The larger domain has three additional landfills that, based on the EPA gridded inventory (Maasakkers et al., 2016), increase CH$_4$ emissions by about 50% when compared to the smaller domain.

This issue became apparent when the emissions were calculated using an atmospheric inversion model (Lamb et al., 2016; Lauvaux et al., 2016). The atmospheric inversion solved for fluxes in domain 1, which significantly increased the estimated emissions in comparison with the inventory values that were gathered mainly within Marion County (domain 2). When reduced to domain 2, inverse modeling emission estimates decrease to 107 mol/s, which falls within an error bar of Lamb et al. (2016) inventory estimate. This difference is significant and could at least partially explain the discrepancy shown in Fig. 1 between the emission values from the inventories and emission results from the inverse modeling. However, even the decreased inverse modeling estimate is about 40% higher than the inventory.

The subject of the domain is also relevant for airborne mass balance flights because a priori the magnitude and variability of background plume is unknown and could be easily influenced by upwind sources. The issue of background is discussed further in the next section.

3.2 Variability in Background Tower Mole Fraction

Comparisons between Criterion 1 and Criterion 2 CH$_4$ mole fraction enhancements as a function of wind direction are visualized using frequency and bivariate polar plots (Fig. 4). To make the comparison as uniform as possible, only data from 12-16 LST are utilized (all hours are inclusive), when the boundary layer is typically well-mixed (Bakwin et al., 1998). A lag 1
autocorrelation is found between 12-16 LST hours, i.e., the hourly afternoon data are correlated to
the next hour, but the correlation is not significant for samples separated by two hours or more.
Therefore, hours 13 and 15 LST are eliminated to satisfy the independence assumption for hourly
samples. Furthermore, we make an assumption that the data satisfy steady state conditions. If the
difference between consecutive hourly wind directions exceeds 30 degrees or the difference
between hours 16 and 12 LST exceeds 40 degrees, the day is eliminated. Days with average wind
speeds below 2 m/s are also eliminated due to slow transport (the transit time from tower 1 to
tower 8 is about 7 hours at a wind speed of 2 m/s).

Both backgrounds generally agree on the higher CH$_4$ originating from the SW, SE, and E
wind directions (Figs. 4c-f); however, the values themselves differ especially when winds are from
NW, SW, and SE. As the background difference plots indicate, there is noticeable variability in
the magnitudes of the CH$_4$ mole fraction background, where criterion 2, by design, typically has
higher background mole fractions. The background differences, at a given hour, suggest that the
CH$_4$ field enveloping the city is heterogeneous with differences between towers ranging from 0 to
over 20 ppb (Fig. 4g). Because large gradients in CH$_4$ background over the city could pose
challenges for flux estimations using top down methods such as inverse modeling and aircraft mass
balance, it is imperative to establish whether the background differences vary randomly or
systematically and how to choose a background to minimize these errors.

To further understand the nature of background variability we calculate the mean and
standard error of background hourly differences over November 2014 to December 2016 for each
of the eight wind directions mentioned in Table 1. The results are shown in Fig. 5. Systematic
bias is evident for the SE, S, SW, W, and NW wind sectors, whereas random error dominates N,
Wind directions showing bias have mean biases ranging from 2 to 5 ppb, with values as large as 8 ppb falling within the range of two times the standard error.

Random errors in the mole fractions of background differences are also important and are a function of the length of the data record. We quantify the random error in the CH₄ background mole fraction differences using the bootstrap method by randomly sampling 2 to 150 hours of the background CH₄ differences for each of the wind directions with replacement (we make the assumption that our differences are independent since we eliminated lag 1 autocorrelation from the data). This sub-sampling experiment is repeated 5000 times (Efron and Tibshirani, 1986). The standard deviations of the mean (standard error) of the 5000 simulated differences are calculated for each wind direction. The resulting standard errors of the city CH₄ background, multiplied by 2 to represent the 95% confidence intervals, are shown as a function of the length of the data record in Fig. 6. All wind directions demonstrate that, as expected, the random error falls as the sample size grows. In general, 25 hourly samples of data reduce random errors by about 70%. Using less than 25 hourly samples to estimate emissions may result in random errors of CH₄ mole fractions of 5 to 35 ppb. Now we consider these random and systematic errors in the CH₄ background in context of Indianapolis urban CH₄ emissions.

For Indianapolis, using INFLUX tower network, we estimated that depending on sample size (number of hours sampled) and wind direction systematic and random errors of CH₄ background gradient across the city over 12-16 LST could vary from 0 to 5 ppb and from 5 to 35 ppb respectively. Given that the average afternoon CH₄ enhancement of the city is around 8-12 ppb (section 3.3; Fig. 7; Cambaliza et al., 2015; Miles et al., 2017), the error on the estimated emissions could be over 100% if the analysis does not approach the issue of background with enough sampling. For CH₄ sources with a significantly larger signal than their regional...
background, the mentioned background variability becomes less impactful on results, but because Indianapolis is a relatively small emitter of CH$_4$, the uncertainties due to background are comparatively large. Our random error assessment suggests that the highly variable CH$_4$ emission values of Indianapolis from the aircraft mass balance calculations (Fig. 1) are at least partially due to the variability in the urban CH$_4$ background of Indianapolis.

3.3. Temporal Variability

Fig. 7 presents average CH$_4$ mole fraction enhancements and flux calculations (equation 4) at towers 8 and 13 for years 2014, 2016, and 2013-2016 (for the detailed methodology see sections 2.6). The years of 2014 and 2016 are chosen for temporal comparison because they do not contain major BLH data gaps. The error bars in the figure show the standard error multiplied by 2 indicating 95% confidence interval of each average.

One of the more interesting features in the Fig. 7 is a day/night variability of CH$_4$ emissions at Indianapolis. The most prominent example of this feature is found in Fig. 7c, where the estimates for both years suggest that daytime emissions are at least twice as high as the emissions at night. The decrease of the CH$_4$ emissions at night also appears in tower 13, but the errors are too high in those estimates to make any definitive conclusions. A similar urban CH$_4$ emissions diurnal variability is reported by Helfter et al. (2016) in their study of GHGs for London, UK, where they attribute diurnal variation of the CH$_4$ emissions to the NG distribution network activities, fugitive emissions from NG appliances, and to temperature-sensitive CH$_4$ emission sources of biogenic origin (such as a landfill). Taylor et al. (2018) suggest that CH$_4$ emissions from landfills exhibit a diurnal cycle with higher emissions in early afternoon and 30-40% lower emissions at night.
With regard to yearly temporal variability we are only able to compare years 2014 and 2016 due to limited BLH data for other years. Results from both towers suggest that Indianapolis CH$_4$ emissions did not change significantly.

3.4 Sources

Bottom-up emission inventories have difficulty tracking changes in sources over time. Our continuous tower network observations can monitor temporal and spatial variability in sources of CH$_4$ in Indianapolis. To do so we employ the aforementioned bivariate polar plots to verify known sources and potentially identify unknown sources across the city. We compare two time periods, 2014-2015 (two full years) and 2016. Fig. 8 displays bivariate polar plots of CH$_4$ enhancements using criterion 1 background at 9 INFLUX towers in Indianapolis over the two years of 2014 and 2015. Fig. 9 shows the same plot, but for the year 2016. Here we have separated 2016 from 2014-2015 because of different results noted during this time.

The images reveal that the most consistent and strongest source in the city is the SSLF. This is most evident from the 40+ ppb CH$_4$ enhancements detected at towers 7, 10 and 11 coming from the location of the SSLF (by triangulation). Enhancements from the landfill appear to also be detectable at towers 2, 4, 5, and 13. Based on these observations it can be concluded that there is no other source in Marion County comparable in strength to the SSLF. A small fraction of the SSLF plume is likely due to the co-located wastewater facility, but the inventory estimates suggest that the wastewater treatment facility is responsible for no more than 7% of this plume (Cambaliza et al., 2015; Massakkers et al., 2016). The PEP, located in the northwestern section of the city, may be partially responsible for a plume of 5-10 ppb at towers 5 and 11. However, the plume is less detectable using the criterion 2 background value that has higher background (using tower 8...
as a background) from NW wind direction (not shown), adding uncertainty to the true magnitude of the enhancement from this source. The same is true for towers 2 and 13, which have pronounced plumes when winds are from the NW with the criterion 1 background, but when background 2 is used these plumes vanish (not shown). Such inconsistency makes it difficult to attribute these plumes to an urban source.

Another important point is the cluster of large enhancements surrounding tower 10 in 2014-2015. Because no other tower sees these enhancements (at least at comparable magnitudes), we believe that these plumes are the result of local NG leaks likely from residential sector of Indianapolis. These plumes are not consistent temporally or spatially as they mostly disappear in 2016, potentially indicating that they are transient NG distribution leaks. It is reasonable to hypothesize that NG related CH$_4$ is being emitted by diffuse, small leaks all across the city. However, towers downwind of the city do not see a large or distinct enhancement from the city, especially when compared to the SSLF source. Thus, the diffuse NG source suspected to be twice as large as the SSLF source (Lamb et al., 2016) does not appear to be supported by these data. This finding contradicts conclusions made by Cambaliza et al., (2015), who attributed most of the CH$_4$ emitted by Indianapolis to NG related activities. We hypothesize that the relatively high Indianapolis CH$_4$ emissions (see Fig. 1) reported by Cambaliza et al., (2015) are the result of the low sample size of airborne flux estimates, which is prone to large random errors (see section 3.2). Our results indicate that the main CH$_4$ source in the city is SSLF and that other sources potentially associated with NG distribution are difficult to identify with clarity. This conclusion is in agreement with EPA 2012 inventory (section 2.3).

4 Conclusions
We have examined four specific contributions to discrepancies between urban top-down and bottom-up CH$_4$ emission estimates from Indianapolis: domain definition, heterogeneous background mole fractions, temporal variability in emissions, and source knowledge. Results indicate that the urban domain definition is crucial for the comparison of the emission estimates among various methods. Atmospheric inverse flux estimates for Marion County, which is similar to the domain that is analyzed by inventory and airborne mass balance methodologies (Mays et al., 2009, Cambaliza et al., 2014, Lamb et al., 2016), is 107 mol/s compared to 160 mol/s that is estimated for the larger domain (Hestia inventory domain). This partially explains higher emissions in inverse modeling estimates shown by Lamb et al., (2016); however, 107 mol/s is still about 40-50% higher than what EPA and Lamb et al., (2016) find in their inventories (Fig. 1).

The midday Indianapolis atmospheric CH$_4$ mole fraction background is shown to be heterogeneous with 2-5 ppb, statistically significant biases for NW, W, SW, S and SE wind directions. We focus on midday atmospheric conditions to avoid the complexities of vertical stratification in the stable boundary layer. Background random error is a function of sample size and decreases as a number of independent samples increase. Low sample volumes, such as a few hours of data from a single location, are prone to random errors on the order of 10-20 ppb in the CH$_4$ enhancement, similar to the magnitude of the total enhancement from the city of Indianapolis. Longer-term sampling and/or more extensive background sampling is necessary to reduce the random errors. Several days of measurements (e.g. 25 total hours of measurement) would reduce random errors to 3-5 ppb, noticeably smaller than the typical enhancement from Indianapolis emissions. This large random error in the CH$_4$ background may explain Heimburger et al. (2017) finding of large variability in airborne estimates of Indianapolis CH$_4$ emissions. Given many
samples, the airborne studies converge to an average value of CH$_4$ flux that is close to inventory estimates for Indianapolis (see Fig. 1).

Measurement and analysis strategies can minimize the impacts of these sources of error. Spatially extensive measurement of upwind CH$_4$ mole fractions are recommended. For towers or other point-based measurements, multiple upwind measurement locations are clearly beneficial. For the aircraft mass balance approach, we recommend an upwind transect be measured, lagged in time if possible, to provide a more complete understanding of the urban background conditions. Complex background conditions might suggest that data from certain days or wind directions should not be used for flux calculation. Finally, a mesoscale atmospheric modeling system informed with the locations of important upwind CH$_4$ sources can serve as a powerful complement to the atmospheric data (Barkley et al., 2017). Such simulations can guide sampling strategies, and aid in interpretation of data collected with moderately complex background conditions.

With regard to temporal variability, no statistically detectable changes in the emission rate were observed when comparing 2014 and 2016 CH$_4$ emissions. However, a large difference between day and night CH$_4$ emissions was implied from a simple budget estimate. Night (20-5 LST) emissions may be 2 times lower than the emissions during the afternoon (12-16 LST) hours. Because prior estimates of top-down citywide emissions are derived using afternoon-only measurements, overall emissions of Indianapolis may be lower than these studies suggest. This bias may be present in studies performed in other cities as well. Our study suggests that day/night differences in CH$_4$ emissions must be understood if regional emission estimates are to be calculated correctly. Long-term, tower-based observations are an effective tool for understanding and quantifying multi-year variability in urban emissions.
One final point addressed in this study is the location of major CH$_4$ sources in Indianapolis. Analysis of the INFLUX observation data suggests that inventories for Indianapolis are mostly accurate and that there is likely no evidence of a large, diffuse NG source of CH$_4$ as implied by Lamb et al., (2016). The only major source in the city is SSLF and it is observed at multiple towers. There is evidence for occasional NG leaks, but they appear localized and limited in their strength.

Overall, assessment of the CH$_4$ emissions at Indianapolis highlights a number of uncertainties that need to be considered in any serious evaluation of urban CH$_4$ emissions. These uncertainties amplify for Indianapolis since its CH$_4$ emissions are comparable in magnitude to the regional background flow and as our results show it may be difficult at times to distinguish noise in the background from the actual city emissions signal. The evaluation of larger CH$_4$ sources may be easier with respect to separating signal from background. However, all of the points raised in this work will be nonetheless relevant and need to be addressed for our understanding of urban CH$_4$ emissions to significantly improve.

**Author Contribution**

Nikolay Balashov, Kenneth Davis, and Natasha Miles developed the study and worked together on generating the main hypothesis of this work. They also wrote most of the manuscript. Nikolay Balashov wrote all of the codes and performed the analyses presented in this work as well as generated all of the figures. Natasha Miles and Scott Richardson helped with maintenance and gathering of the INFLUX tower data. They also wrote section 2.2 of the paper. Thomas Lauvaux helped with the analysis presented in Figure 1 and section 3.1 concerning interpretation of the inversion modeling results from Lamb et al. (2016). Zachary Barkley significantly contributed to
discussions regarding the hypothesis and careful presentation of sections 2.6 and 3.3. Timothy Bonin provided all of the lidar data and wrote the second part of section 2.3 regarding the lidar and the methodology used to determine planetary boundary layer heights. He also contributed to sections 2.6 and 3.3.

Competing Interests

The authors declare that they have no conflict of interest.

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References


Tables

Table 1. INFLUX towers used to estimate CH$_4$ background based on two different criteria. Numbers in bold indicate towers chosen to generate a background field when multiple options are possible (for more details see discussion). In short, Criterion 1 uses towers with the lowest mean CH$_4$ for a specific wind direction, and Criterion 2 uses towers not downwind of large sources (including the city as a whole).

<table>
<thead>
<tr>
<th>Wind Direction</th>
<th>CH$_4$ Background Towers</th>
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<tbody>
<tr>
<td></td>
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<tr>
<td>North (N)</td>
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</tr>
<tr>
<td>Northeast (NE)</td>
<td>8</td>
</tr>
<tr>
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<tr>
<td>West (W)</td>
<td>1</td>
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<tr>
<td>Northwest (NW)</td>
<td>1</td>
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Figures

Figure 1. Various estimates of CH$_4$ emissions at Indianapolis. (a, b) Bottom-up estimates of CH$_4$ emissions conducted by Lamb et al. (2016) in 2013 and Maasakkers et al. (2016) based on the EPA 2012 inventory respectively. Error bars show 95% confidence intervals (for more details see above-mentioned articles). (c-g) Top-down evaluations of CH$_4$ emissions with aircraft from various flight campaigns where (c) contains 5 flights over March-April of 2008, (d) contains 3 flights over November-January of 2008-09, (e) contains 5 flights over April-July of 2011, (f) contains 9 flights from November-December, 2014, and (g) contains the same 5 flights over April-July of 2011 but uses different methodology. Methodologies for (c-f) are described in Lamb et al. (2016) and methodology for (g) is described in Cambaliza et al. (2015). Error bars show 95% confidence intervals (for more details see above-mentioned articles). (h, i) Top-down evaluations of CH$_4$ emissions for 2012-2013 using tower inversion modeling methodology with two different domains, where (h) uses the full domain of Fig. 3 and (i) uses only the Marion County domain of Fig. 3. The inversion methodology and 95% confidence intervals are described in detail in Lamb et al. (2016).
Figure 2. 20-day running average of afternoon (12-16 LST; the hours are inclusive) CH$_4$ mole fractions as measured by the INFLUX tower network (highest available height is used) from 2012 through 2016.
Figure 3. Map of the primary roads in Indianapolis, INFLUX towers, lidar system, weather stations, and a few CH₄ point sources plotted over the gridded CH₄ emissions (mol/s) from the EPA 2012 Inventory (Maasakkers et al., 2016). The gridded map of emissions includes emissions from these point sources; their position is provided to aid in interpretation of the observations. The dashed bright green line denotes Marion County borders.
Figure 4. Frequency and bivariate polar plots of CH$_4$ background for Indianapolis using data from 12-16 LST, and November, 2014 through December, 2016 given 2 different criteria (Table 1). (a) Polar histogram of number of daily measurements available using criterion 1. (b) Same as (a) only for criterion 2. Differences between (a) and (b) are due to slight differences in data availability at the considered towers. (c) Polar frequency plot of the mean CH$_4$ background using criterion 1. (d) Same as (c) only for criterion 2. (e) Polar bivariate plot of CH$_4$ background using criterion 1. (f) Same as (e) only for criterion 2. (g) Polar frequency plot of difference between the backgrounds: criterion 2 – criterion 1. (h) Same as (g) but shown with a bivariate polar plot.
Figure 5. Average of the differences between criteria 2 and 1 CH₄ backgrounds at Indianapolis as a function of wind direction. These averages are generated from the same data as used in Fig. 4 and reflect results shown in Fig. 4g. Error bars indicate standard error × 2.
Figure 6. Bootstrap simulation of the standard errors multiplied by 2 in Indianapolis CH$_4$ background mole fraction differences (between criteria 2 and 1) as a function of sample size and wind direction (see text for details).
Figure 7. Averages of the daytime (D) and nighttime (N) CH$_4$ enhancements and fluxes at INFLUX towers 8 and 13 for years 2014 (14), 2016 (16), and 2013-2016 (1316). The error bars represent 95% confidence interval of each mean value. (a) Estimates of CH$_4$ enhancements from tower 8. (b) Estimates of CH$_4$ enhancements from tower 13. (c) Estimates of CH$_4$ flux from tower 8. (d) Estimates of CH$_4$ flux from tower 13.
Figure 8. Google Earth image overlaid with bivariate polar plots (section 2.5) of the CH$_4$ enhancements at 9 INFLUX towers in Indianapolis using the criterion 1 background (Table 1) for full years of 2014 and 2015 over the afternoon (12-16 LST). The wind speed scale is only labeled at site 1; other sites follow the same convention. Legend indicates known sources of CH$_4$: Panhandle Eastern Pipeline (PEP), Southern Side Landfill (SSLF), Twin Bridges Landfill (TBLF), Waste Management Solutions (WMS), Waste Water treatment facility (WW). The known magnitudes of sources that are in Marion County (PEP, SSLF, and WW) are reported in section 2.3. Magnitudes of TBLF and WMS according to EPA are approximately 5 mol/s. The largest known source on the map is SSLF.
Figure 9. Same as Fig. 8 only for the year of 2016.