Response to Referee # 3

We would like to thank the reviewer for the helpful suggestions. Below we address all of the comments presented to us by the reviewer.

Reviewer Comments

When people talk about "missing" or "unknown" NG sources in urban environments they are usually thinking of leaks that occur downstream of the customer meter. If many small leaks in homes and businesses throughout the city were each leaking a tiny amount, how would that appear on your Figures 6 and 7? Would you be able to see it? The way you describe "unknown" sources you seem to be looking for point sources rather than a broad diffusive source.

First we would like to apologize regarding our error in figure numbering. We noticed that Figures 8 and 9 somehow got renumbered to 6 and 7 in the main paper. This is now fixed. We assume that this question refers to Figures 8 and 9.

If we are talking about small leaks (what do we mean by small is also an important matter, but we will ignore this for now) such as homes and businesses throughout the city then their signal depends to a large extent on their proximity to towers as well as on the actual amount of these leaks. On Figure 8 it is possible to see some very large enhancements that are occurring near tower 10 (the color scale is saturated, but if we were to increase the color range we would see enhancements near 70 ppb). No other tower sees these enhancements at comparable magnitudes; therefore, it is possible to conclude that these leaks are located near tower 10 and unable to fully mix before they reach our sensor (that is at tower 10). Here immediately we can note an interesting point – these leaks do not appear to be common, at least according to our towers. We do not see anything similar at other towers, and in 2016 these leaks near tower 10 are gone. Of course it is possible that these leaks are unique to some specific infrastructure and perhaps there are still copious small leaks from other places as noted above. In that case, for these leaks to matter they must mix at some point and form a noticeable plume that would emanate from the city. That plume should be at least partially comparable to the landfill plume to really matter. Because if there are small leaks and they are barely detectable it doesn't matter much for the overall city budget. So to address this question a little deeper, let us perform a thought experiment. We can pose the following question: could SSLF explain the SW plume at towers 2 and 8 (these are the towers where the city plume should be generally well mixed)? The reason this question is important is because if landfill can explain this plume, then there is not much space for a significant gas source out there. Sure, there could be a noticeable gas source overall – something like 20-30% of the landfill, but nothing shocking as sometimes implied. In order to address this question it is useful to think of an approximation of a Gaussian plume equation.

The equation could be simplified to the following form:

$$C(x,0,0) = \frac{Q}{U}\frac{1}{\pi\sigma_y\sigma_z} \qquad (1)$$

This equation approximates the change in concentration (or mole fraction) of the plume center (of a given gas) as a function of distance away from a source (x), where Q is the source strength in mol/s, U is wind speed in m/s, σ_y is lateral spread of plume in m, and σ_z is vertical spread of plume in m. Both sigmas depend on atmospheric stability and additionally σ_z on a height of a mixed layer. Equation indicates that plume strength will rapidly decline as it moves away from the source mixing in its surrounding environment. Here we will use this equation to illustrate how the mole fraction of SSLF declines with distance and how our towers reflect this decline. Of course what we are about to show is a rough approximation since we are assuming unstable conditions, winds of 9 m/s, boundary layer of about 700 m, and SSLF strength of 32 mol/s. It is imperative to understand that these chosen parameters are not so important and general principle by which plumes spreads remains the same. Occasionally you get higher mole fractions and occasionally you get lower mole fractions, but over a long period of time these mole fractions will average out.

Before we return to our thought experiment with Gaussian plume, let us take note of some interesting aspects of the observations that are shown in Figure 9. Figure 1 (in this response) shows binned averaged CH_4 enhancements by wind direction at towers 10, 11, 2, and 8 for 2016. In some ways this picture is similar to Figure 9 of the article, but allows for a different perspective. Right away we notice a relatively large enhancements at towers 10 and 11, where for tower 10 the enhancement is observed when winds are from west (at about 270 degrees) and for tower 11 the enhancement is observed when winds are from south (at about 180 degrees). These enhancements occur at wind directions consistent with the location of SSLF. There is a little doubt that SSLF is the main culprit here. Now if we look at towers 2 and 8, we notice that tower 2 also exhibits 10-20 ppb enhancement at SW wind direction, which is again consistent with SSLF. Finally, at tower 8 wind directions of S-SW show a smaller but still noticeable enhancement of about 10-15 ppb. And again, the direction is consistent with SSLF. Green circles in Figure 1 emphasize the plumes that are suspected to originate from SSLF. Could these plumes really be attributed to SSLF?

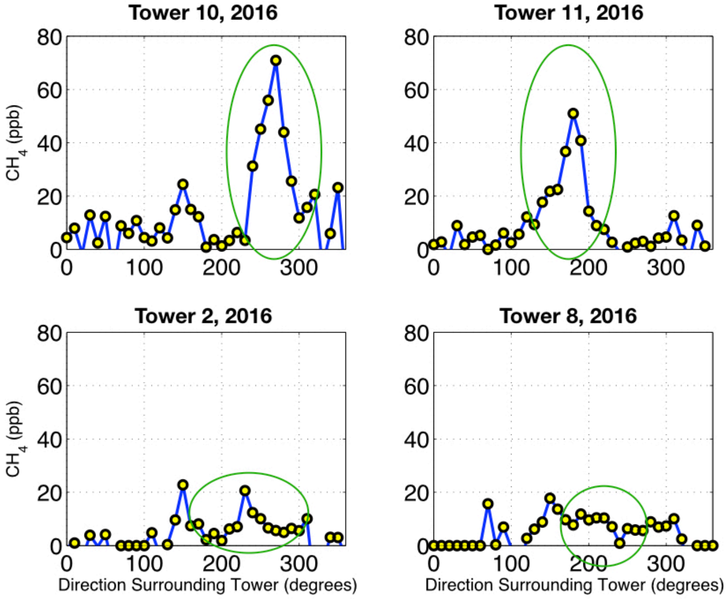

Figure 1. Directionally (meaning wind direction) averaged CH$_4$ enhancements binned by 10 degrees for towers 10, 11, 2, and 8 for 2016. Criterion 1 is used for background as described in the article. Green circles highlight CH$_4$ enhancement that is associated with SSLF.

Let us come back to our thought experiment with Gaussian plume. Figure 2 shows how a mole fraction of CH$_4$ would vary as we move away from SSLF. The locations of towers 10, 11, 2, and 8 are indicated by the colorful circles. Once again, the equation 1 is a rough estimation of how plume spreads and not a high-resolution modeling. But on average, plume will generally follow these rules of dispersion. We are assuming unstable conditions, wind speed of 9 m/s, source strength of 32 mol/s, and a boundary layer of 700 meters. So once the plume is mixed to 700 meters, it will no longer expand vertically as the layer is capped. At this point the plume expands only laterally, but that expansion is rather slow as could be noted in the Figure 2. If we compare mole fractions that are expected by our simple simulation at each of the towers we notice that they are generally consistent with what is observed in Figure 1. More importantly, the ratios by which the mole fractions decrease as we move away from SSLF are consistent. In Figure 1, the plumes peak near 70, 50, 20, and 10-15 ppb at towers 10, 11, 2, and 8 respectively. This is similar to what Figure 2 shows. The only discrepancy that is noticeable is at tower 11, which is about 20 ppb higher in Figure 1 than in Figure 2, but that could be explained by an error of our approximation. There are many factors to which real plume is slightly sensitive as we described above. The main story however is not changed. The plumes observed in Figure 1 are closely consistent with what we would expect to originate from a SSLF. Therefore, it appears that SSLF is indeed the most important source in the city and is responsible for most of the CH$_4$ emitted. It could be noted that there are other smaller peaks in Figure 1, which indeed are likely to originate from natural gas leaks. But from this analysis it is clear that these leaks, even if combined, are smaller than the emissions from Landfill. If these leaks were to emit more gas than a landfill, we would see much larger enhancements at towers 2 and 8, which would be inconsistent with the scenario where the main source of CH_4 in Indianapolis is SSLF. But we do not observe that. SSLF is able to explain the majority of CH_4 that is observed at our towers. Of course, this Gaussian plume experiment is a crude estimation and we cannot completely rule out significant source from natural gas leaks, but at this point it is very difficult to make a logical argument that even if such source does exits that it is bigger than SSLF.

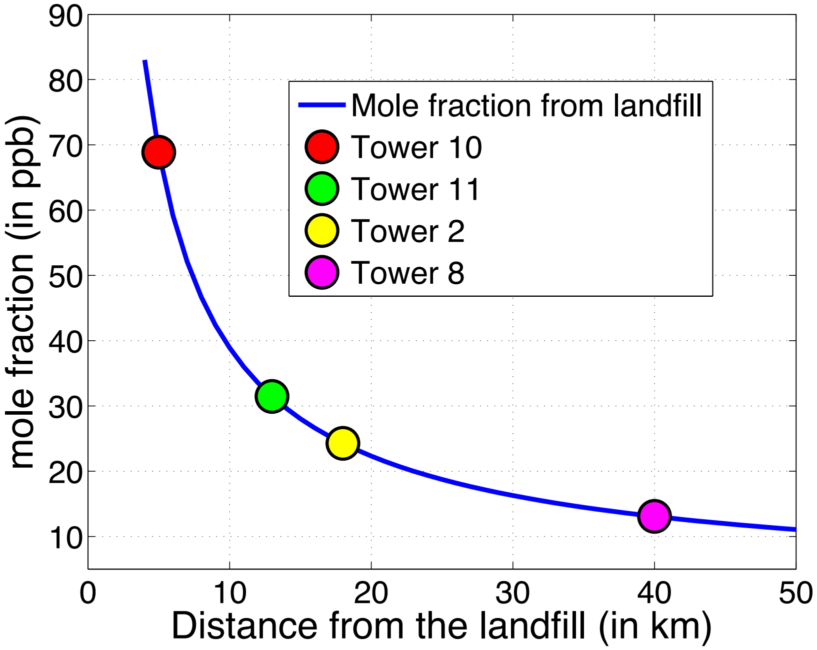

Figure 2. Simulation of plume dispersion using equation 1. The plume originates from SSLF and the source strength is assumed to be 32 mol/s, which is consistent with EPA estimates. Color circles indicate towers based on the distance away from SSLF. Y-axis shows mole fractions that are expected at these towers given a rough approximation resulting from the equation 1.

So to answer your question, the small leaks should appear relatively large on Figures 8 and 9 of the article when they are in close proximity to towers and smaller when they are detected by the away towers such as 2 and 8. At towers 2 and 8 we could expect leaks to combine into a single combined plume and if this plume would compare in magnitude to SSLF we would unquestionably detect it. In fact, we are currently working on another INFLUX paper where we analyze various flights that measured CH_4 downwind of Indianapolis and simulate these downwind plumes with 3-km WRF-Chem. Preliminary results show that downwind plume is indeed sensitive to even a slight increases in small gas leaks. Many flights we analyzed do appear to indicate that SSLF is the main source consistent with the observations, but we will not get into these results here. It is pointed out here just to support the point that mixed plumes are well detected by the receptors downwind of the city (such as tower 8 for instance). And yes, when we describe “unknown” sources, we are talking about both diffuse sources and point sources. For example, if there was another point source comparable to SSLF in the vicinity of Indianapolis, our tower analysis would immediately pick it up. So just because there is not a large unknown source in the area this time doesn't mean it is always true for a given city. And if such source would suddenly appear, we would see it in our towers because of their superb temporal resolution. Diffuse source will also be detected as already was pointed out in this note earlier. We recommend our type of analysis to anyone starting to analyze either CH_4 or CO_2 for a particular city. It is helpful to understand the observations well before carrying out any kind of modeling.

Making a small aside here, it is important to point out that a good inversion study should always carefully examine available data (such as our paper has done, which is a very thorough analysis of almost every single measurement made at towers) and see if their prior assumptions make sense. And after inversion is carried out, it is imperative to assess the results and see if they make actual sense given the observed data. Unfortunately, many studies do not do that. Many presentations do not even show the raw data used for inversions (such as time series, etc.), let alone examine the observations with great scrutiny. Now, we are not saying that Lamb et al. (2016) was not a careful study, but there are certainly aspects in that work that are questionable.

Section 2.6 in part describes how data are filtered for this analysis but it does not say how many data are remaining. I'm wondering if enough data are available to do seasonal-level analysis?

We do not actually perform seasonal-level analysis. We do try to understand annual variability of CH_4 emissions by comparing three different time periods. For tower 8 if we counted every single hour used for daytime analysis, we get 259 values (60 days) for 2014, 346 values (78 days) for 2016, and 756 values (172 days) for 2013-2016. For nighttime analysis, we get 334 values (42 days) for 2014, 306 values (44 days) for 2016, and 789 values (107 days) for 2013-2016. Note that more hours are allowed per *24-hour-day* for nighttime than daytime (see section 2.6).

For tower 13 if we counted every single hour used for daytime analysis, we get 120 values (31 days) for 2014, 159 values (38 days) for 2016, and 399 values (98 days) for 2013-2016. For nighttime analysis, we get 94 values (13 days) for 2014, 228 values (31 days) for 2016, and 445 values (64 days) for 2013-2016.

Overall, the number of samples is robust according to the central limit theorem.

Section 2.7 – Lamb et al. 2016 gives an estimate for the SSLF based on plume dispersion modeling that would seem worth including since it an independent metric based on direct measurements. That paper says the estimates for SSLF from GHGRP, aircraft mass balance and plume dispersion modeling all yield similar estimates. Whereas in this section you give a wider range for the SSLF, citing essential the same sources as Lamb et al. 2016.

You are correct. Lamb et al. 2016 does perform an inversion to estimate emissions from SSLF and the result is similar to GHGRP. We will include that estimate. However, estimates of GHGRP and aircraft mass balances (the ones performed in Cambaliza et al. (2015)) do not yield similar results hence in our article we point that out. Lamb et al. (2016) does say that new aircraft estimates of landfill are consistent with GHGRP, but that actual "new" experiment is nowhere to be found. It is likely that Cambaliza et al. (2015) background estimates are wrong, but Lamb et al. (2016) did not show how they modified the methodology and therefore it seems not appropriate to use their value over Cambaliza et al. (2015) value. So in our paper we now mention both values: from Cambaliza et al. (2015) and recalculation from Lamb et al. (2016).

The paragraph starting at line 361 is confusing. Presumably errors were thoroughly assessed in the original work. Are you saying that you are not sure if doing the inversion for the smaller domain is valid? What kind of sensitivity analysis do you think is needed? Same comment for the sentence in the conclusions section starting at line 518. It makes it sound like you have reason to doubt this result, but you haven't said what the reasons are other than "inversions can have large errors."

You make a good point that this paragraph is not well supported by the data, so we will delete it. Due to personal communications with some of the authors of Lamb et al. (2016), there is a reason to think the inversion errors were not fully assessed. Unfortunately, it is unclear how it could be relayed here, so these passages will have to be deleted. The point was to emphasize that inversion errors are bigger than the error bars suggest in Lamb et al. (2016).

Inversion for smaller domains could be valid if proper meteorological model is used. For one, it is unclear whether the meteorological model used in Lamb et al. (2016) inversion was able to handle boundary layer heights properly. Another issue was prior, which was based on 5 flights from Cambaliza et al. (2015). Cambaliza et al. (2015) should not be used as an estimate for prior due to its poor background strategies and extremely low sample size.

Multiple steps must be performed to insure that inversion is valid. First, footprints derived from meteorological model must be double-checked. To do so derived boundary layer heights must be compared to observations and the corresponding error needs to be known. The analysis of footprints showed that realistic time series at certain towers could not be reproduced. Second issue is prior. How does the inversion vary as prior varies? This analysis was partially performed with the inversion model used in Lamb et al. (2016) and the results were unsatisfactory.

So here it is, yes, there are reasons to doubt inversion. But because it is not possible to put unofficial results into the paper, we will edit the current article to reflect what is published so far.

Reviewer Suggestions

We would like to thank the reviewer for carefully reading our manuscript. The suggestions presented here are well-thought-out and noticeably improve this article.

Lines 25-26: Suggest: "…within the error range of one of the two inventories."

Done.

Line 27: Suggest: "…higher than inventory estimates."

Done.

Line 28: Suggest: "…spatially heterogenous and temporally variable. Variability in background mole fractions observed at any given moment and a single location could be…"

Done.

Line 31: Suggest: "from most wind directions" to "for most wind directions"

Done.

Line 35: Suggest: "No significant unknown CH4 sources are found." to "We found no evidence for significant CH4 point sources that are otherwise missing from the inventories."

Done.

Line 37: Suggest: "Other sources, such as leaks from the natural gas (NG) distribution system, are localized and transient, and do not appear to be a consistently large source of CH4 emissions in Indianapolis." to "Leaks from the NG distribution system that were detected with the tower network appeared localized on non-permanent and do not appear to constitute as large of a source of CH4 as previously hypothesized by some top-down studies"

Done.

Line 39: Suggest deleting the sentence: "However some uncertainty…"

Done.

Line 51: Delete ", however, "

Done.

Line 59: Miller et al. is not the correct citation for the EPA and EDGAR inventories.

Corrected.

Line 72: Suggest: "For example, two studies (McKain, Hendrick) indicate that…"

Done.

Line 76: Suggest: "due to fugitive emissions from NG infrastructure in urban environments."

Done.

Line 96: Suggest: "estimates" to "quantification"

Done.

Line 97: Delete "estimates"

Done.

Line 99: “which are” to “specifically”

Done.

Line 109: Suggest: “from Cambaliza et al. (2015) which used”

Done.

Line 113: Delete: “This uncertainty has not yet been resolved.”

Done.

Line 118-119: Suggest for clarity: “(1) inconsistent geographic boundaries between top-down and bottom-up studies, (2) heterogeneity in the urban scale CH4 background and (3) temporal variability in urban emissions, both of which were not accounted for in top-down studies, and (4)…”

Done.

Line 138: Suggest: “has been” to “was”

Done.

Line 160: CH4 scale information is irrelevant.
Deleted.

Line 163: Suggest: “was” to “were”

Done.

Line 168: Suggest: The accuracy of the wind speed measurements are…”

Done.

Line 181: Suggest: “…when the determined BLH is unreliable…”

Done.

Line 197: Suggest: “Aircraft mass balance studies of Indianapolis mentioned used two main…”

Done.

Line 244: Suggest: “…slow transport across the city”

Done.

Line 275: “upwind” to “downwind”?

Yep, should be downwind.

Line 326: Suggest deleting “contigent on how much of the total city emissions are coming from NG.”

Deleted.

Line 333: NG distribution leaks are not mentioned as potential sources.

It is now added into the sentence.

Line 338: Suggest: “An important question remains of whether SSLF or NG is the dominant CH4 source in Indianapolis, or whether they are approximately equal.”

Done.

Line 340: “…as described above” – not clear where you are referring to. Could just delete this last sentence.

Deleted.

Line 379: “enveloping” to “flowing into”

Done.

Lines 434, 439: Figure 7 to 5

This is actually correct; the Figure number should be 7 here. We apologize for our error in figure numberings. We fixed it now.

Line 458: consistent with the larger error bar of Lamb et al. (2016) inventory calculation.

Done.

Line 475: Suggest: “Based on these observations it can be concluded that there are no other point source in Marion County comparable in size to the SSLF.

Done.

486: Suggest: “an urban” to “a specific”

Done.

Line 501: Suggest deleting: "it is imperative…", so it reads, "However, our flux estimates…"

Done.

Line 508: Suggest: "We have examined four potential contributions to discrepancies between urban top-down and bottom-up estimates of CH4 emissions from Indianapolis: domain definition, heterogeneous background mole fractions, temporal variability in emissions, and sources missing from inventories."

Done.

Line 523: "towers are selected" to "estimates"

Done.

Line 532: "Low sample volumes" to "small sample sizes"

Done.

Line 542: "noticeable smaller than" to "which is half the magnitude of"

Done.

Line 549: "than their individual components as" to "than several of the individual estimates"

Done.

Line 580: "since its CH4 emissions are comparable in magnitude to" to "since the enhancement signal from its CH4 emissions is comparable in magnitude to variability in"

Done.

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 (CH_4), a powerful greenhouse gas, has grown. Large discrepancies in Indianapolis CH_4 emissions have been observed when comparing inventory, aircraft mass-balance, and tower inverse modeling estimates. Four years of continuous CH_4 mole fraction observations from a network of nine towers as a 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 CH_4 background, (3) temporal variability in CH_4 emissions, and (4) the presence of unknown CH_4

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sources. Reducing the Indianapolis urban domain size to be consistent with the inventory domain size decreases the CH_4 emission estimation of the inverse modeling methodology by about 35% and thereby lessens the discrepancy by bringing total city flux within the error range of one of the two inventories. Nevertheless, the inverse modeling estimate still remains about 40% higher than inventory estimates. Hourly urban background CH_4 mole fractions are shown to be spatially heterogeneous and temporally variable. Variability in background mole fractions observed at any given moment and a single location could be up to about 50 ppb depending on a wind direction, but decreases substantially when averaged over multiple days. Statistically significant, long-term biases in background mole fractions of 2-5 ppb are found from single point observations for most wind directions. 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). We found no evidence for significant CH_4 point sources that are otherwise missing from the inventories. The data from the towers suggest that the strongest CH_4 source in Indianapolis is South Side Landfill. Leaks from the natural gas distribution system that were detected with the tower network appeared localized and non-permanent and do not appear to constitute as large of a source of CH_4 as previously hypothesized by some top-down studies. 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 real representation of emission estimates are recommended for precise and accurate quantification of urban CH_4 emissions.

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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 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).

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In particular, the estimates of continental U.S. anthropogenic CH_4 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} (U.S. EPA, 2013; European Commission Joint Research Centre and Netherlands Environmental Assessment Agency, 2010). 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 sometimes 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

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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 NG supply chain (Alvarez et al., 2018).

Due to the uncertainties in CH_4 emissions from NG distribution it is natural that urban emissions are of interest as well. For example, two studies (McKain et al., 2015; Hendrick et al., 2016) indicate that ~60-100% of Boston CH_4 emissions are attributable to the NG distribution system. Recent studies of urban CH_4 emissions in California indicate that the California Air Resources Board (CARB) inventory tends to underestimate the actual CH_4 urban fluxes possibly due to fugitive emissions from NG infrastructures in urban environments (Wunch et al., 2009; Jeong et al., 2016; Jeong et al., 2017). The accuracy and precision of atmospheric estimates of urban CH_4 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; Karion et al., 2015; Heimburger et al., 2017). In this work, detailed analysis of urban CH_4 mole fractions is performed for 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 irregular insitu aircraft measurements (Heimburger et al., 2017; Cambaliza et al., 2014), continuous in situ observations from communications towers using

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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 designed for emissions quantification using top-down methods such as tower-based inverse modeling (Lauvaux et al., 2016) and aircraft mass balance estimates (Cambaliza et al., 2015).

Lamb et al. (2016) compared Indianapolis CH_4 emissions estimates from a variety of approaches, specifically 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 (GHG reporting program). Although evidence of possible variability in landfill emissions exists from Cambaliza et al. (2015), which used aircraft mass balance on five different occasions to calculate CH_4 flux from this landfill. Uncertainty in total city emissions is mainly 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). In this study, we explore potential explanations for

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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 between top-down and bottom-up studies, (2) heterogeneity in the urban scale CH_4 background and (3) temporal variability in urban emissions, both of which were not accounted for in top-down studies, 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.

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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). In 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) does 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).

Figure 2 shows two domains that have been used for the evaluation of Indianapolis CH_4 emissions (Lamb et al., 2016; Lauvaux et al., 2016). The first domain is the whole area shown in the figure enclosing both Indianapolis and places that lie outside of its boundaries. This domain was used for the inversion performed in Lamb et al. (2016). 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 Indianapolis CH_4 emissions by about 50% when compared to the smaller domain. The inversion explained in Lamb et al. (2016) has been rerun for two of the domains mentioned above and the results (Fig. 1) have been reexamined.

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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 CH_4 measurements from 2013 – 2016. From June through December 2012, there were two or three towers with operational CH_4 measurements. By July 2013, five towers included measurements of CH_4, and throughout the majority of the years 2015 – 2016 there were eight INFLUX towers with CH_4 measurements (Fig. 3). 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). In this study we use hourly means of CH_4

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2.3 Meteorological data

Wind data were 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 https://www.weather.gov/media/asos/aum-toc.pdf. The accuracy of the wind speed measurements are ±1 m/s or 5% (whichever is greater) and the accuracy of the wind direction is 5 degrees when the wind speed is $\geq$ 2.6 m/s. The anemometers are located at 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.

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The planetary boundary layer height (BLH) was determined from a Doppler lidar deployed in Lawrence, Indiana 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 determined 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 Urban methane background

Both aircraft mass balance and inverse modeling methodologies rely on an accurate estimation of the urban CH_4 enhancement relative to the urban CH_4 background in order to produce a reliable flux estimate (Cambaliza et al., 2014; Lamb et al., 2016). The CH_4 mole fraction enhancement is defined as,

$$C_{enhancement} = C_{downwind} - C_{bg} \qquad (1)$$

where $C_{downwind}$ is the CH_4 mole fraction measured downwind of a source and C_{bg} is the CH_4 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 assumed to accurately represent mole fractions that are upwind of the source of interest and measured simultaneously with the downwind mole fractions.

Aircraft mass balance studies of Indianapolis mentioned used two main methods to determine a background value. The first method calculates an average of the aircraft transect edges that lie outside of the city domain (Cambaliza et al., 2014). In the second approach, a horizontally varying background is introduced by linearly interpolating median background values of each of the transect edges (Heimburger et al., 2017). In theory there is also a third method that uses an upwind transect as a background field, but in the studies above it was

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assumed that the edges are representative of an upwind flow. In the case of an inversion, it is common to pick a tower that is located generally away from urban sources and has on average the smallest overall enhancement (Lavaux et al., 2016). Because choosing the background involves a degree of subjectivity (Heimburger et al., 2017) we consider how these choices 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 CH_4 background fields are generated for the city of Indianapolis based on two different sets of criteria. The notion is based on the fact that a choice of background is currently rather arbitrary in the literature (Heimburger et al., 2017) and at every point in time it is possible to choose multiple background values that are equally acceptable for the flux estimation. In our case both approaches 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°). Estimating background for different wind directions is implemented to more accurately represent upwind flow that is hopefully not contaminated by local sources.

Criterion 1 corresponds to a typical choice of a background in a case of tower inversion and is based on the concept that the lowest CH_4 mole fraction measured at any given time is not affected by the city sources and therefore is a viable approximation of the background CH_4 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 CH_4 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 CH_4 source (Fig. 2). 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 atmospheric CH_4 background at Indianapolis.

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 across the city (the transit time from tower 1 to tower 8 is about 7 hours at a wind speed of 2 m/s).

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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 mole fraction values, which are fitted to a surface using a Generalized Additive Model (GAM) framework in the following way,

$$\sqrt{C} = \beta + s(u, v) + \epsilon \tag{2}$$

where C is the CH_4 mole fraction transformed by a square root to improve model diagnostics such as a distribution of residuals, β is mean of the response, s is the isotropic smoothing function of the wind components u and v, and ϵ is the residual. For more details on the model see Carslaw and Beevers (2013).

2.6 Temporal variability and approximate flux estimation

Temporal variability may play an important role in the quantification of urban CH_4 emissions. Lamb et al. (2016) suggested that temporal variability might partially explain the differences among CH_4 flux estimates shown in Figure 1. If temporal variability of CH_4 emissions exists within the city, disagreements in the CH_4 flux between studies could be attributed to differences in their sampling period. Because the INFLUX tower data at Indianapolis contain measurements

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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 CH_4 emissions Q_a (mass per unit time per unit area) are not chemically active and are constant over a distance Δx spanning a significant portion of the city. The next assumption is that a CH_4 plume measured downwind of the city is well mixed within a layer of depth H (which is the same as BLH). 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 CH_4 concentration C within a box in the following fashion,

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$$\Delta x H \frac{\partial C}{\partial t} = \Delta x Q_a + uH(C_b - C) + \Delta x \frac{\partial H}{\partial t}(C_a - C) \tag{3}$$

where C_b is the CH_4 concentration upwind of the city (or background), and C_a is the CH_4 concentration above the mixed layer (Hanna et al., 1982; Arya, 1999; Hiller et al., 2014). The left hand side of the equation represents the change in CH_4 concentration with time, $\Delta x Q_a$ denotes a constant CH_4 source over the distance Δx, $uH(C_b - C)$ indicates a change of CH_4 concentration due to horizontal advection, and finally $\Delta x \frac{\partial H}{\partial t}(C_a - C)$ term accounts for the vertical advection and encroachment processes that result from changing BLH. By assuming steady state conditions ($\frac{\partial C}{\partial t} = 0$ and $\frac{\partial H}{\partial t} = 0$), the equation can be simplified to

$$Q_a = \frac{uH(C - C_b)}{\Delta x} \tag{4}$$

We use equation 4 to estimate hourly CH_4 emissions (Q_a) from Indianapolis (see assumptions in the paragraph below) given hourly averaged data of H from the lidar positioned in the city, wind speed (u) from the local weather stations, and upwind (C_b) and downwind (C)

CH_4 mole fractions measured (and then converted to concentrations) at towers 1, 8, and 13 (depending on a wind direction) using data from heights of 40 m, 41 m, and 87 m respectively (see Fig. 2).

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 are 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 (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 H 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 H 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

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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.

2.7 Indianapolis CH_4 sources

Only a few known CH_4 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 thought to be the largest point source in the city with emissions ranging between about 28 mol/s (inventory from Maasakkers et al. (2016), GHG reporting program, and inverse estimates from ground-based mobile sampling employed in Lamb et al. (2016)) and 45 mol/s (aircraft; Cambaliza et al. (2015)) depending on an emission estimation methodology. However, using Cambaliza et al. (2015) aircraft data and applying a different background formulation Lamb et al. (2016) found emission values of SSLF closely agreeing with 28 mol/s estimate. SSLF could account for as little as 33% (top-down from Cambaliza et al., 2015) or as much as 63% (invetnory from Maasakkers et al., 2016) of total Marion County CH_4 emissions. Other city point sources are comparatively small; the wastewater treatment facility located near SSLF contributes about 3-7 mol/s (inventory from Lamb et al. 2016), 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 1 mol/s (inventory from Lamb et al. 2016). The remaining CH_4 sources, mainly from NG infrastructure leaks and livestock, are considered to be diffuse sources

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and are not well known. Potential sources of emissions related to NG activities include gas regulation meters, transmission and storage, distribution leaks, and Compressed Natural Gas (CNG) fleets. These diffuse NG sources account for 21-67% (this value varies due to the uncertainty in SSLF emissions) of the city emissions or 20 mol/s (inventory from Maasakkers et al., 2016) to 64 mol/s (top down from Cambaliza et al., 2015). Livestock emissions for Marion County are estimated to be around 1.5 mol/s (inventory from Maasakkers et al., 2016). An important question remains of whether SSLF or NG is the dominant CH_4 source in Indianapolis, or whether they are approximately equal.

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3 Results and discussion

3.1 Inversion and 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 decrease in the domain area could add or remove a large point source and significantly impact the total emissions defined within the domain.

In the case of Indianapolis, 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 (Fig. 2), 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 Figure

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 inventories.

Additionally, the subject of the domain is 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 CH_4 background

Comparisons between criterion 1 and criterion 2 CH_4 background mole fractions as a function of wind speed and direction are visualized using frequency and bivariate polar plots (Fig. 4). 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 background difference plots (Figs. 4g-h) indicate, there is a noticeable variability between the magnitudes of the CH_4 backgrounds, where criterion 2, by design, typically has higher background mole fractions. The background differences, at a given hour, suggest that the CH_4 field flowing into the city is heterogeneous with differences between towers ranging from 0 to over 45 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.

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To further understand the nature of background variability we calculate the mean, standard deviation, and standard error of background hourly differences between criterion 2 and criterion 1 from November 2014 to December 2016 for each of the eight wind directions mentioned in Table 1. The results are shown in Figure 5. Systematic bias is evident for the SE, S, SW, W, and NW wind sectors, whereas random error dominates N, NE, and E wind directions. Wind directions showing statistically significant bias have mean biases ranging from 2 to 5 ppb, with values as large as 8 ppb falling within the range of 2 × standard error. Standard deviation plot indicates potential background discrepancy that can occur on any given day, where W wind direction is the least variable with 2 × standard deviation close to 20 ppb, while SE wind direction is the most variable with 2 × standard deviation falling at about 50 ppb.

Random errors in the mole fractions of background differences (biases) are also important and are a function of the length of the data record. We quantify the random error in the CH_4 background mole fraction differences using the bootstrap method by randomly sampling 2 to 150 hours (small and large sample size) of the background CH_4 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 CH_4 background differences, multiplied by 2 to represent the 95% confidence intervals, are shown as a function of the length of the data record in Figure 6. Because random error falls as sample size grows it makes sense to assign a threshold indicating a minimum number of samples needed to achieve a theoretical precision for each wind direction.

One way to assign a required precision would be to make sure that the standard error (random error) reaches a point where it is less than Indianapolis enhancement of about 12 ppb (a higher estimate of the Indianapolis enhancement from section 3.3) by a factor of 2 when combined with a bias (Table 2). Meaning that the sum of bias and standard error must be at most 6 ppb. In this approach each wind direction would have a different threshold because of the differences in biases. For instance, given this requirement NW direction would need a random error of 1 since its bias is 5. For NW direction, this threshold would require more than 150 samples. For N direction on the other hand, where the bias is 1, the requirement is fulfilled when random error crosses 5 ppb at 74 samples. Now we consider these random and systematic errors in CH_4 background differences in the context of Indianapolis urban CH_4 emissions.

For Indianapolis, using INFLUX tower network, we estimated that depending on sample size (number of hours sampled) and wind direction, background gradient across the city over 12-16 LST could vary from 0 to about 50 ppb (Fig. 5b). Given that the average afternoon CH_4 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 easily be over 100% if the analysis does not approach the issue of background with enough sampling. A sample size of about 50 independent hours significantly decreases background uncertainty for N, NE, E, S, and W wind directions and allows for a more accurate assessment of the CH_4 emissions at Indianapolis. For CH_4 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, and because there are relatively large sources outside of the city, uncertainties due to background estimation are comparatively large. Our uncertainty assessment suggests that the highly variable CH_4 emission values of Indianapolis from aircraft mass balance calculations shown in Figure 1 are at least partially due to the variability in the urban CH_4 background of Indianapolis.

3.3. Temporal variability of methane enhancements and fluxes in Indianapolis

Figure 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 section 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.

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One of the more interesting features in the Figure 7 is a day/night variability of CH_4 emissions at Indianapolis. The most prominent example of this feature is found in Figure 7c, where the estimates for both years suggest that daytime emissions are approximately twice as large 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 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 overall CH_4 emissions did not change significantly between 2014 and 2016.

Although it is important to be cautious about interpreting actual flux estimations given the assumptions mentioned in section 2.6, it is interesting to note that the flux values from both towers average at about 70 mol/s, which puts our value right in between inventory and inversion estimates shown in Figure 1. If we assume that SSLF emissions are generally known (GHG reporting program) that would indicate that emissions from NG distribution are likely to be somewhat higher than both of the inventories currently estimate and consistent with the larger error bar of Lamb et al. (2016) inventory calculation. Another possible scenario is that SSLF emissions are higher than what is currently assumed. Given these complexities, uncertainty regarding the exact emissions from NG distribution at Indianapolis still remains.

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3.4 Methane Sources in Indianapolis

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. Figure 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. Figure 9 shows the same plot, but for the year 2016. Here we have separated 2016 from 2014-2015 because of different results noted during these times.

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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

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that there are no other point sources in Marion County comparable in size 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 a specific source.

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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 and localized NG distribution leaks. It is difficult to ascertain the exact combined magnitude of these leaks since they mix together with SSLF into an aggregated city plume when observed from downwind towers such as 8 and 13. Yet, none of these leaks appear to be even remotely close in magnitude to the emissions that originate from SSLF. 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 assertion questions 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

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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 making it prone to large random errors (see section 3.2). However, our flux estimations at towers 8 and 13 discussed in the previous section do imply that emissions from NG distribution may be higher than estimated by the inventories indicating that an overall NG contribution may be comparable in strength to SSLF. This discrepancy requires further investigation.

4 Conclusions

We have examined four potential contributions to discrepancies between urban top-down and bottom-up estimates of CH$_4$ emissions from Indianapolis: domain definition, heterogeneous background mole fractions, temporal variability in emissions, and sources missing from inventories. 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; Gurney et al., 2012). 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).

To better understand background variability at Indianapolis two different but acceptable background estimates, based on specific criteria for each wind direction, and their differences are used to assess heterogeneity of CH$_4$ background at Indianapolis. Background criterion 1 looks for a tower that is consistently lower than other towers, while background criterion 2 picks a

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tower that is outside of Marion County domain and is not downwind of any nearby sources as determined by EPA 2012 inventory. We focus on midday atmospheric conditions to avoid the complexities of vertical stratification in the stable boundary layer. 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. Random errors of background differences are a function of sample size and decrease as a number of independent samples increase. Small sample sizes, such as a few hours of data from a single point, are prone to random errors on the order of 10-30 ppb in the CH_4 background, similar to the magnitude of the total enhancement from the city of Indianapolis, which is estimated to be on average around 10-12 ppb. Longer-term sampling and/or more extensive background sampling are necessary to reduce the random errors. Sample size required to reduce random errors of background differences to an acceptable value for flux calculation is largely dependent on a wind direction. Both bias (long-term average of background differences) and its random error are important when estimating total background uncertainty. The results indicate that N, NE, E, S, and W wind directions are more favorable for flux estimation and would require multiple days of measurements (e.g. about 50 independent hours of measurements) to reduce background uncertainty to about 6 ppb, which is half the magnitude of the typical CH_4 enhancement from Indianapolis. The remaining wind directions would require over 150 independent hourly measurements to achieve similar precision. We also estimate that depending on a wind direction for any given hour the spatial variability in background can be anywhere from 0 to 50 ppb. This uncertainty in the CH_4 background may partially 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 noticeably closer to the

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inventory estimates for Indianapolis than several of the individual estimates presented in Figure 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 to 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 rates 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.

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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 no clear 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 an evidence for occasional NG leaks, but they appear localized and limited in their strength. However, we cannot completely rule out occasional significant leaks of CH_4 from NG at Indianapolis due to the nature of our assumptions.

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 the enhancement signal from its CH_4 emissions is comparable in magnitude to variability in 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.

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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 Fig. 1 and section 3.1 concerning interpretation of the inversion modeling results from Lamb et al. (2016). Thomas Lauvaux also helped with repeating the inversion experiment for two different Indianapolis domains (Fig. 1). 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.

Acknowledgements

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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 outside of Marion County and not downwind of large sources (including the city as a whole).

Wind Direction	CH_4 Background Towers	
	Criterion 1	Criterion 2
North (N)	8	**13**, 8
Northeast (NE)	8	**13**, 8, 2
East (E)	**2**, 8	**8**, 4, 1, 2
Southeast (SE)	1	**8**, 13, 4, 1
South (S)	1	**4**, 13, 1
Southwest (SW)	13	**1**, 4
West (W)	1	**4**, 1

Northwest (NW)	1	**8**, 1

Table 2. A number of independent samples needed (column 4) to satisfy combined requirement of 6 ppb background error based on the sum of bias and random error (explained in section 3.2) as a function of wind direction.

Wind Direction	Bias (ppb)	Threshold (ppb)	Samples Needed
N	1	5	74
NE	1	5	36
E	0.5	5.5	46
SE	4	2	>150
S	2	4	53
SW	4.5	1.5	>150
W	3	3	52
NW	5	1	>150

Figures

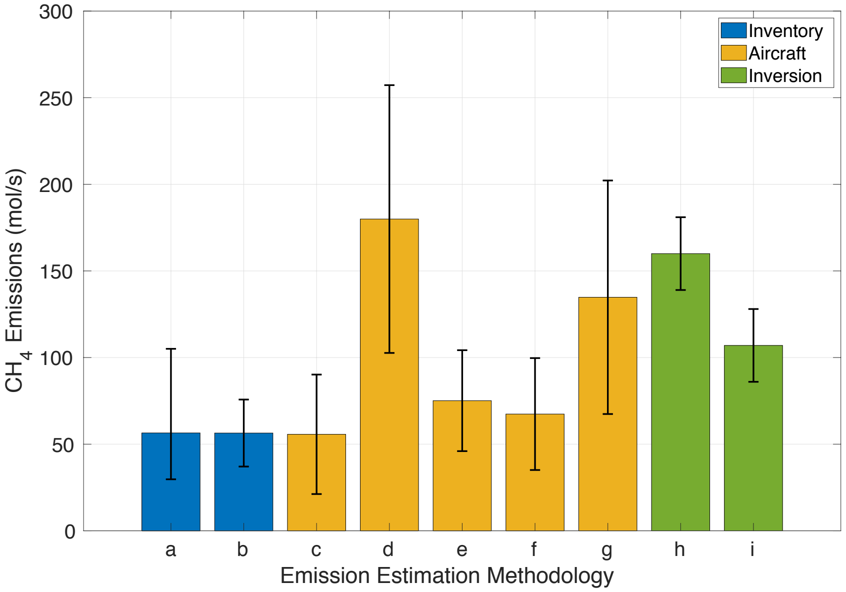

Figure 1. Various estimates of CH_4 emissions at Indianapolis. **(a, b)** Bottom-up estimates of CH_4 emissions conducted by Lamb et al. (2016) in 2013 and Maasakkers et al. (2016) based on the EPA 2012 inventory respectively. Error bars show 95% confidence intervals (for more details see above-mentioned 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 as in (e) 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 Figure 2 and **(i)** uses only the Marion County domain of Figure 2. The inversion methodology and 95% confidence intervals are described in detail in Lamb et al. (2016).

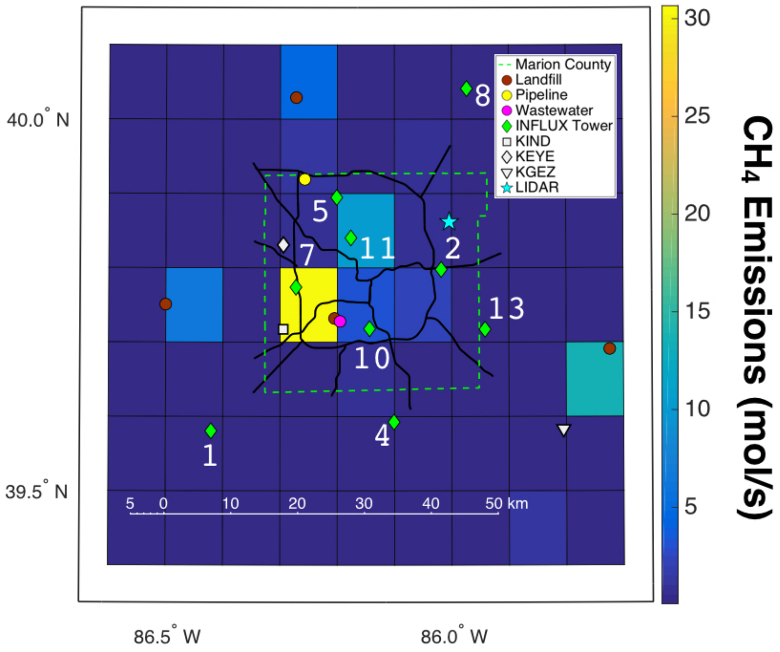

Figure 2. Map of the primary roads in Indianapolis, INFLUX towers, lidar system, weather stations, and a few CH_4 point sources plotted over the gridded CH_4 emissions (mol/s) from the EPA 2012 Inventory (Maasakkers et al., 2016). The gridded map of emissions includes emissions from the mentioned point sources; their position is provided to aid in interpretation of the observations. The dashed bright green line denotes Marion County borders.

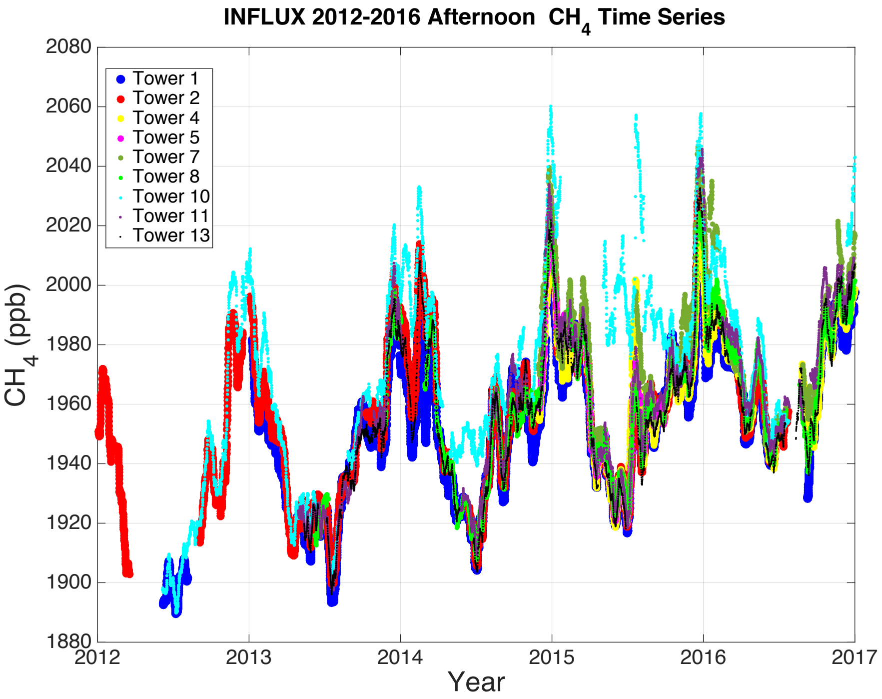

Figure 3. 20-day running average of afternoon (12-16 LST; the hours are inclusive) CH_4 mole fractions as measured by the INFLUX tower network (highest available height is used) from 2012 through 2016.

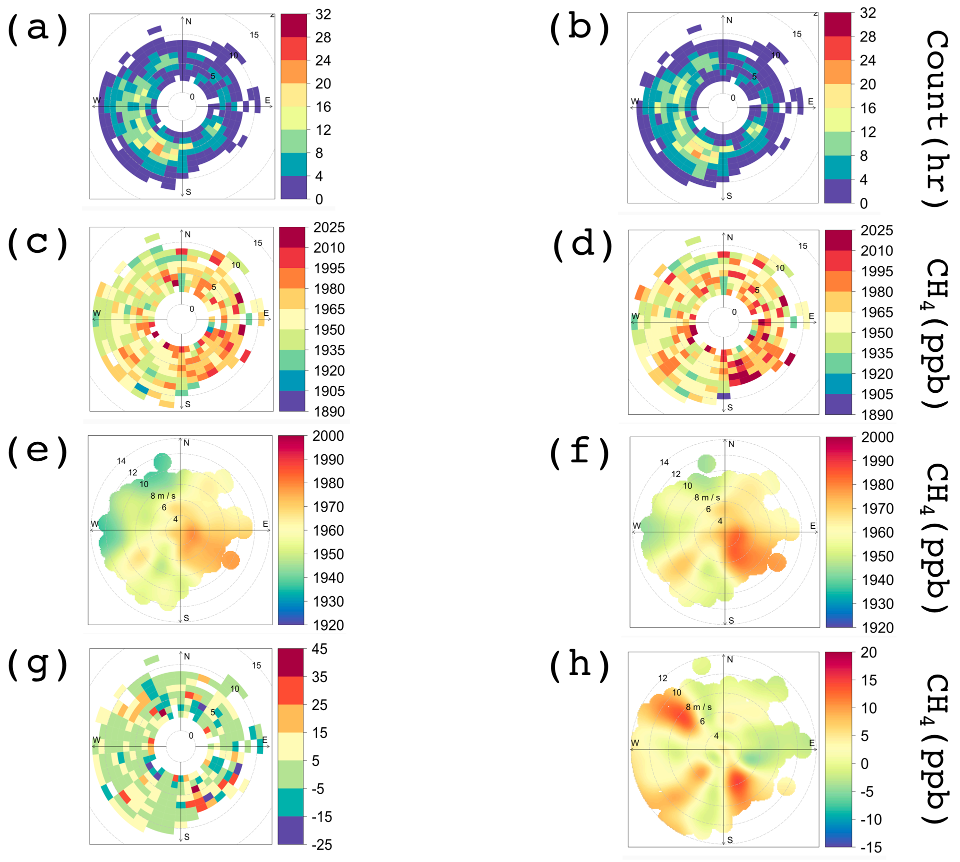

Figure 4. Frequency and bivariate polar plots of CH_4 background for Indianapolis using data from 12-16 LST, November 2014 through December 2016 given 2 different criteria (Table 1). **(a)** Polar histogram indicating a number of hourly measurements available using criterion 1. **(b)** Same as (a) only 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 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.

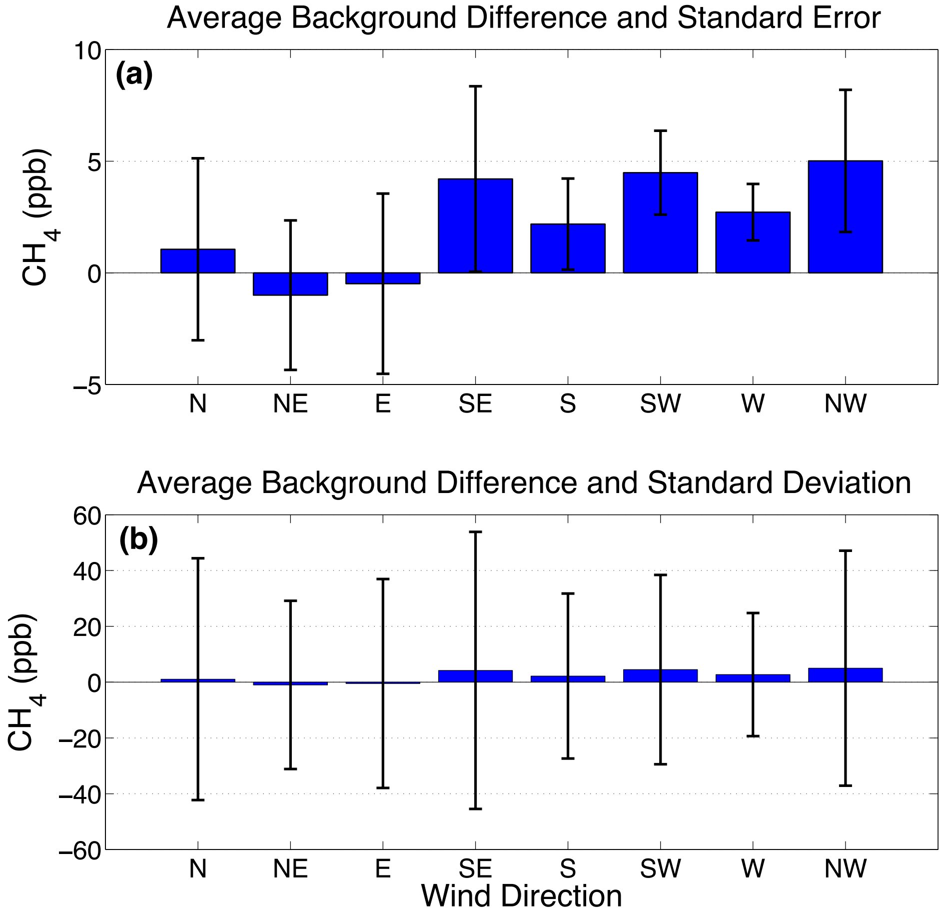

Figure 5. Average of the differences between criteria 2 and 1 CH_4 backgrounds at Indianapolis as a function of wind direction. These averages are generated from the same data that is used in Figure 4 and reflect results shown in Figure 4g. Error bars indicate in (a) 2 × standard error and in (b) 2 × standard deviation.

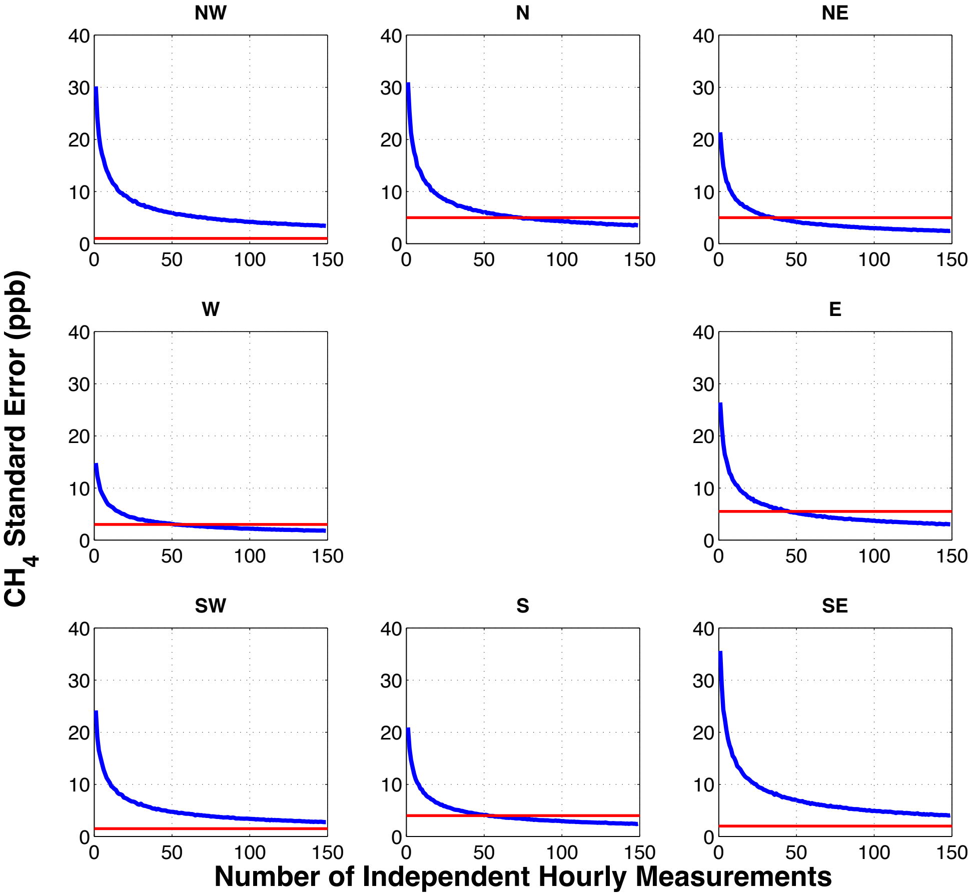

Figure 6. Bootstrap simulation of the standard errors × 2 in Indianapolis CH_4 background mole fraction differences (between criteria 2 and 1) as a function of sample size and wind direction (see text for details). Thresholds for each of the wind directions indicate a random error threshold needed for the background uncertainty to be within 50% of Indianapolis CH_4 enhancement of 12 ppb.

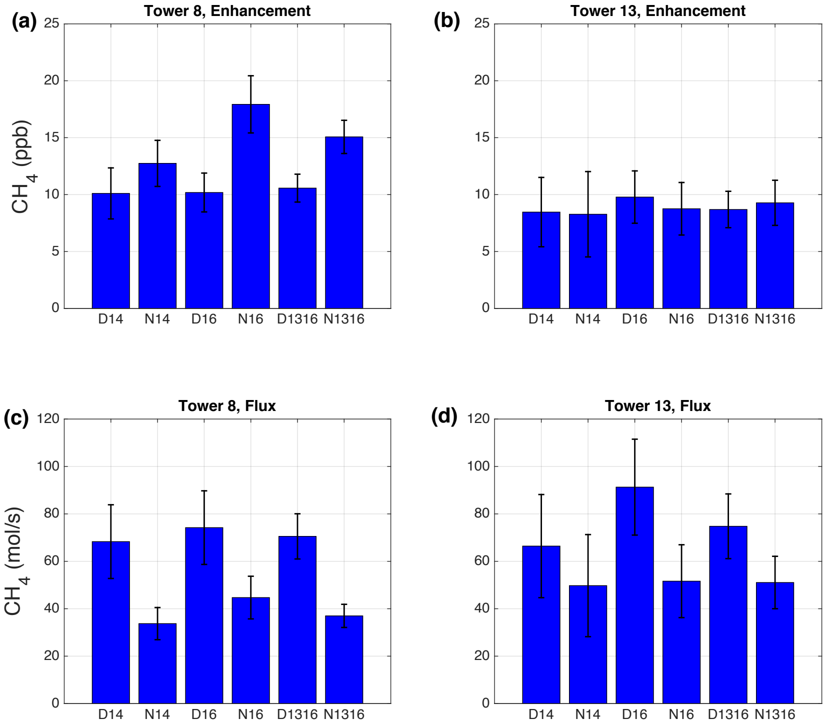

Figure 7. Averages of the daytime (D) and nighttime (N) CH$_4$ enhancements and fluxes at INFLUX towers 8 and 13 for years 2014 (14), 2016 (16), and 2013-2016 (1316). The error bars represent 95% confidence interval of each mean value. **(a)** Estimates of CH$_4$ enhancements from tower 8. **(b)** Estimates 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.

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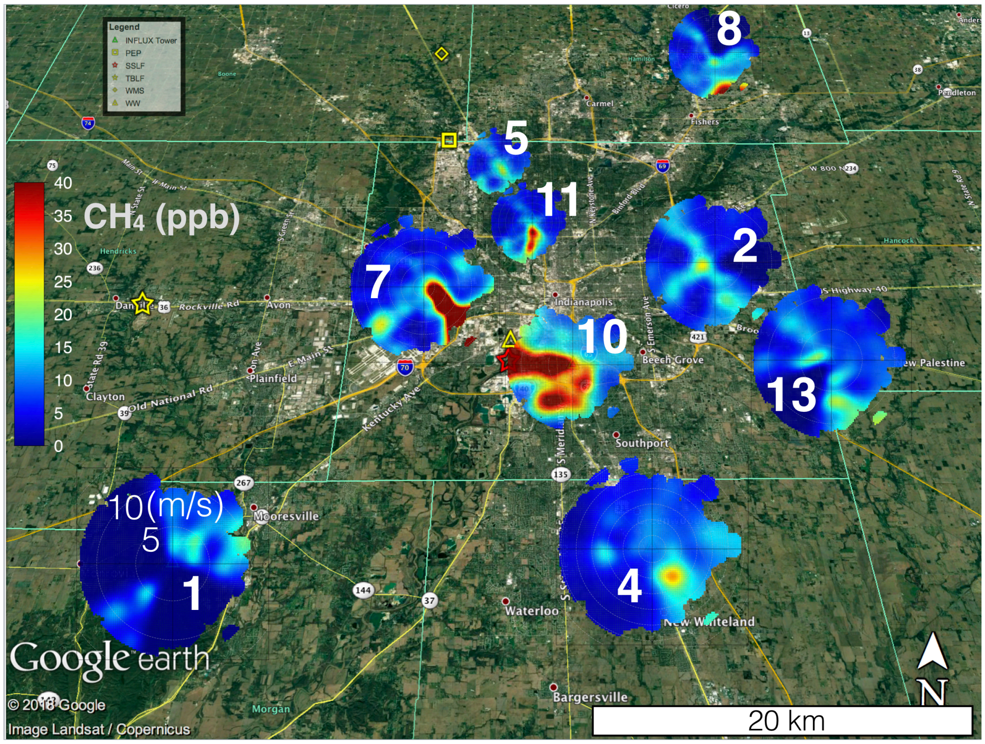

Figure 8. Google Earth image overlaid with bivariate polar plots (section 2.5) of the CH_4 enhancements at 9 INFLUX towers in Indianapolis using the criterion 1 background (Table 1) for full years of 2014 and 2015 over the afternoon (12-16 LST). The wind speed scale is only labeled at site 1; other sites 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), and Waste Water treatment facility (WW). The known magnitudes of sources that are in Marion County (PEP, SSLF, and WW) are reported in section 2.7. Magnitudes of TBLF and WMS according to EPA are approximately 5 mol/s. The largest known source on the map is SSLF.

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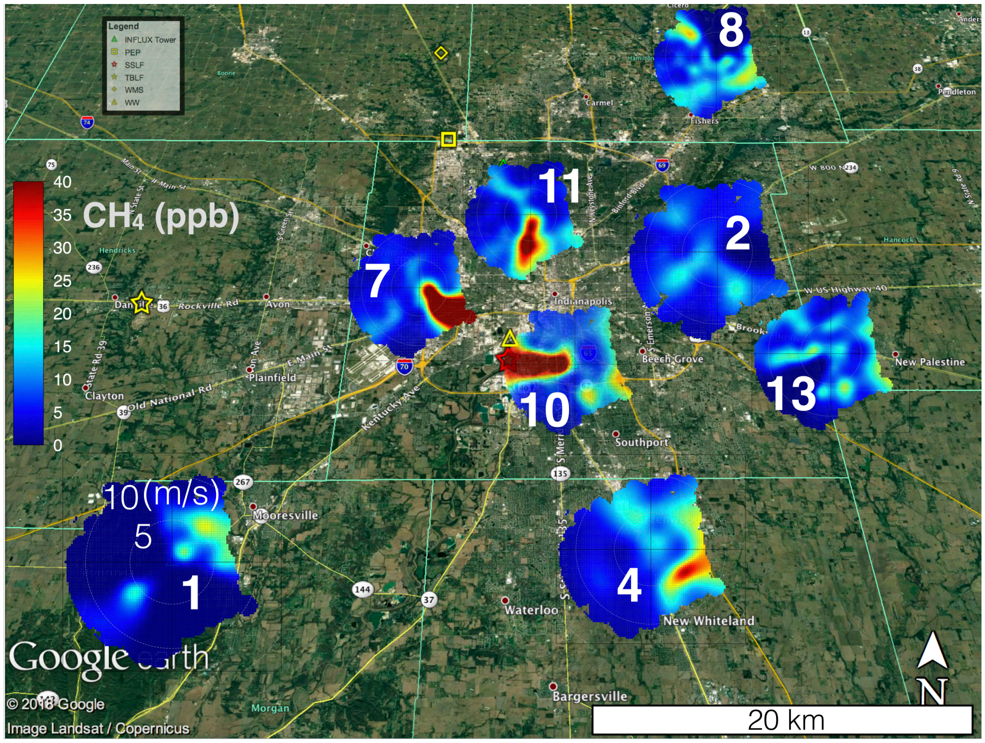

Figure 9. Google Earth image overlaid with bivariate polar plots (section 2.5) of the CH$_4$ enhancements at 9 INFLUX towers in Indianapolis using the criterion 1 background (Table 1) for year 2016 over the afternoon (12-16 LST). The wind speed scale is only labeled at site 1; other sites follow the 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), and Waste Water treatment facility (WW). The known magnitudes of sources that are in Marion County (PEP, SSLF, and WW) are reported in section 2.7. Magnitudes of TBLF and WMS according to EPA are approximately 5 mol/s. The largest known source on the map is SSLF.

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