Response to Referee # 2

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

There are a few sections which may require some clarification, and it may be helpful to the reader if the authors reordered some of the content.

The order of the subsections in section 2 (Methods) does not follow the order that the four components of the inverse modelling framework are listed in the abstract or discussion section. It may make it easier for the reader to follow from the methods through to the results if there is an explicit methods section for each of the four sensitivity analyses.

The sections are now slightly reordered to better reflect the whole structure of the paper.

Section 2.4 lines 172 to 185: Could the authors explain what the percentages are referring to and provide the overall quantity? Does this come from an inventory analysis?

These are percentages of the total Indianapolis methane emissions depending on a given estimation. For instance, some estimates (Cambaliza et al., 2015) found landfill to contribute only to 22% of the total Indianapolis methane emissions, while other estimates (GHG reporting program) found landfill to be responsible for 63% of total city methane emissions. The total emission value could be the same or different. Some estimates come from inventories; others come from top-down studies such as aircraft mass balance and inversion. Figure 1 in the article summarizes different estimates of methane at Indianapolis. It just does not have the breakdown of the total emissions by sources. We know the breakdown only for some studies. There is now an attempt to summarize this breakdown in section 2.7.

The section following, starting at line 187, is also labelled as Section 2.4.

This is an error. It is now fixed.

Section 2.5 line 218: Should this be a subsection of the previous background section (2.4)? Or should this subsection be called Variability in Background Concentration?

Section 2.5 describes bivariate plots, so it is titled correctly. Due to the reordering of the methodology part of the paper some sections now changed their number.

Equation 3 line 250: This equation and notation are taken from Hanna et al. 1982. It may be useful to the reader to explain the terms more thoroughly. Most of the explanation for terms is taken from Chapter 9 of this text, but some of the terms are explained more fully in earlier chapters. I was particularly distracted by the zi until I realised that it was conventional for the zi to denote the boundary layer height of the box, rather than i as an indicator for height at time step i (or distance i). Do you take zi to be the average boundary layer height during the hour?

Yes, zi is confusing variable name, so we have changed it to H. Yes, we assume H represents boundary layer height for a given hour. We are thinking to leave the rest of the description as is (some minor edits are incorporated), but if there is anything that you would like us to explain specifically we could do that.

In the text the units of Qa are described to be in mass per unit time per unit area, and the units of C are described to be in mole fraction in the first instance. Should the concentration not be converted to moles per volume? Later on from line 263 this does appear to be the case.

You are absolutely correct. It was an error; it should say concentration in the first instance. On line 263 it states that because our data is given in mole fractions we must first convert it to concentration (mass per volume) before we can apply the budget equation. Correct, we are not converting CH_4 to moles per volume, but to complete abovementioned conversion from CH_4 mole fractions to CH_4 concentration we do need to use average molar dry density of air, which has units of moles per volume.

Section 3.1 lines 292 to 307: This information seems to be related to methods, and should perhaps be in Section 2, probably before the methods section on Background concentrations to be consistent with the abstract and discussion sections.

Lines 299 to 307 have been moved to methods as requested. The other lines seem to fit appropriately to the results section as they give the domain issue some context. It would be to jarring to jump into line 308 right away.

Line 350: I think it would be useful to justify why you needed to use the bootstrapping approach. Lines 350 to 363 and Figure 6: I would propose that the *analysis for assessing the number of measurements required to obtain a reliable background concentration estimate should focus on the standard deviation rather than on the standard error.*

Bootstrapping approach allows us to vary sample size of a theoretical experiment and to see how the background uncertainty responds to it. Below I explain why this may not work for the standard deviation.

It is unsurprising that the standard error (standard deviation of the mean) reduces as the sample size is increased, as you state in line 359. This is a property of the standard error. If you're interested in the stability of the background concentration difference estimate, you should rather be looking at the standard deviation of the concentration differences (which you can do in the same way, except instead of looking at the standard deviation of the mean values for the 5000 samples, you look at the mean of the variances for the 5000 samples – which can then be converted to a standard deviation). The standard deviation will provide information about the uncertainty in the background concentration. The plots in Figure 6 are slightly different for each wind direction because the standard deviation of the background concentration from these directions differs and because the bias for each direction differs. At some optimum sample size the standard deviation of the concentration differences will stabilize for each direction. If you assume that under normal circumstances (where you don't have two competing domains or multiple background sites) you would be able to obtain the uncertainty in the background concentration from historical data and this uncertainty remains stable over time, you would want to ensure that the background concentration estimate is obtained from a sufficiently large sample size so that you know what the uncertainty in this estimate is. Therefore, if you have multiple background sites where you can assess at which sample size the standard deviation of differences between background sites stabilizes, you may want to determine for each wind direction what this sample size is and what the stabilized standard deviation is. Basing the background concentration on a sample of this size or larger should provide an estimate with a predictable uncertainty, which is now independent of the sample size. If you know what the standard deviation is, then it follows what sample size is required to obtain a background concentration with the required standard error (precision) (if you can use SE = SD/sqrt(n)).

If you decide to stick with the standard error plots, I think you should show on each plot at which sample size each direction reaches the required precision, say 3 ppb, as this differs for each wind direction.

Thank you for such a thoughtful comment. This really gets me thinking deeper about this background variability concept.

I agree with you that it is unsurprising that standard error is decreasing with increasing sample size. However, I do find Figure 6 useful in that standard errors vary depending on wind direction. So given a sample size of around 20 we can get a general idea which wind direction will provide us with the lowest random error of the difference between backgrounds. It is important to note that this Figure 6 does not say anything at all about bias. This is just random error. The bias is a difference between backgrounds and is shown in Figure 5. In ideal world we would want this difference to be zero and have very little random error on it. Unfortunately that is not the case here. The background is quite complex. Now, you do make a valid point regarding standard deviation and that it also gives us information regarding background variability. Because of this I added to Figure 5 another plot that shows standard deviation times 2 for each of the wind directions. That shows us potential background discrepancy that can occur on a given a single day. This is also useful. Here is the updated Figure 5. On the other hand, standard error shows us that as sample size increases our average difference of backgrounds would approach a known bias. But yes, on any given day things could be really variable or not so variable. Additionally, standard deviation plot indicates that W is the best direction regarding the background. It has the lowest variability of background differences. It does have a bias, but overall error is the smallest. This also is evident in Figure 6, where W standard error is the smallest.

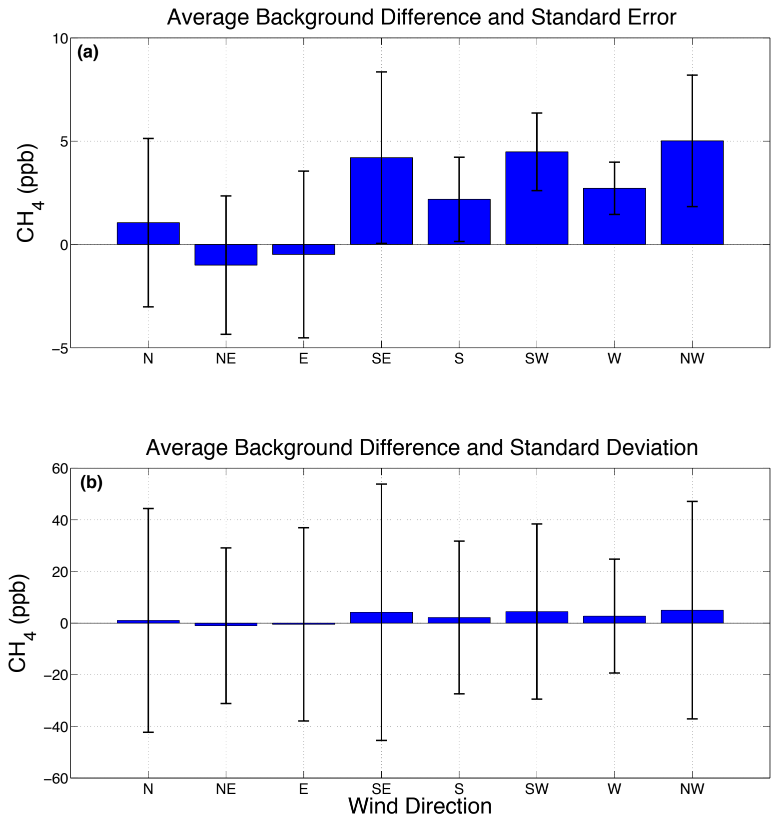

Figure 5. Average of the differences between criteria 2 and 1 CH_4 backgrounds at Indianapolis as a function of wind direction. These averages are generated from the same data as used in Fig. 4 and reflect results shown in Fig. 4g. Error bars indicate in (a) 2 × standard error and in (b) 2 × standard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

To respond to your other request, I tried to perform the bootstrap experiment with standard deviations, but it does not seem to work. The variance stabilizes very fast and does not seem to be a function of sample size (maybe only initially). So I think that it would suffice to add a standard deviation plot to Figure 5 because these standard deviations are basically the same ones you would get with the bootstrap experiment. I think this happens because we are sampling from the pool of the same differences and ultimately there is no way for variances to change much after 5000 iterations. In other words, low sample size with 5000 iterations will be similar to a large sample size with 5000 iterations because both of these cases sample from the same PDF of background differences. Please see Figure 6b.

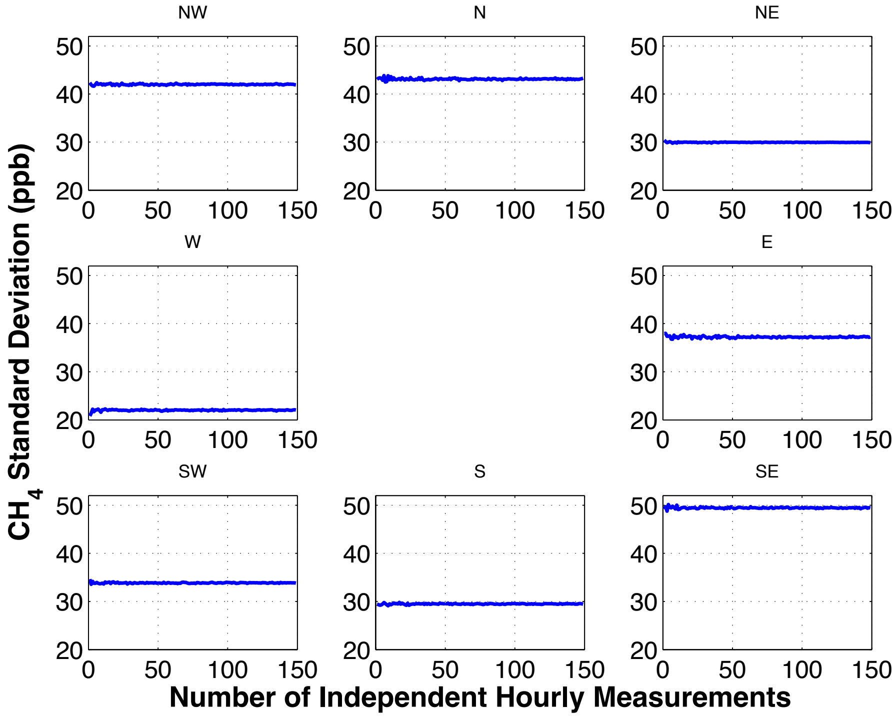

Figure 6b. Bootstrap simulation of the standard deviations multiplied by 2 in Indianapolis CH_4 background mole fraction differences (between criteria 2 and 1) as a function of sample size and wind direction (see text for details).

To answer your last part of this question, I do not think that we have a required precision at this point for background differences. The plot illustrates at approximate what sample size we would approach a reasonable standard error. Perhaps one way to classify 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 3.2) by a factor of 2 when combined with a bias (in this case it is helpful to think in terms of absolute magnitudes, so let say the requirement is 6 ppb). In this case each wind direction would have a different threshold. 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. This is going to be added to this to the paper now.

Here are the absolute values of the biases for each wind direction, their respective required thresholds, and needed sample size:

N: 1 ppb	T: 5 ppb	N: 74
NE: 1 ppb	T: 5 ppb	N: 36
E: 0.5 ppb	T: 5.5 ppb	N: 46
SE: 4 ppb	T: 2 ppb	N: >150
S: 2 ppb	T: 4 ppb	N: 53
SW: 4.5 ppb	T: 1.5 ppb	N: >150
W: 3 ppb	T: 3 ppb	N: 52
NW: 5 ppb	T: 1 ppb	N: >150

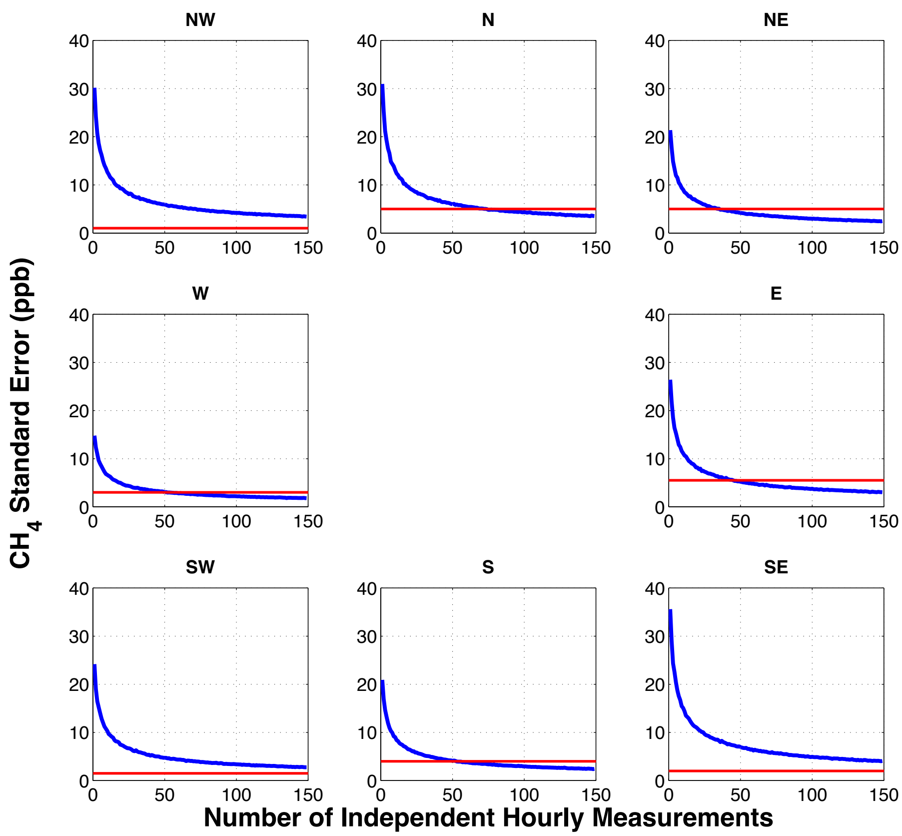

Figure 6a. Bootstrap simulation of the standard errors multiplied by 2 in Indianapolis CH_4 background mole fraction differences (between criteria 2 and 1) as a function of sample size and wind direction (see text for details). Thresholds for each of the wind directions indicate a random error threshold needed for the background uncertainty to be within 50% assuming average CH4 enhancement from Indianapolis is 12 ppb.

A point that should be discussed is that the measurements you obtain for the background site are taken at different times, and as the number of measurements increases, so too does the averaging period, which changes the interpretation of this average. There's a danger that if the averaging period is too long, the background concentration measurements may be representative of different synoptic periods.

There are definitely cases when a front is rolling through the area and the methane gradients are huge causing a background difference of 40 ppb or so. Averaging over a long period of time smooths out these variations. This is why large sample size of estimations is suggested because unavoidably in any type of top-down experiment (aircraft or inversion) there are going to be days that are unsolvable because of complex background. The hope is that there are more days with homogenous background (background variability is less than city enhancement) than days with heterogeneous background (background variability is more than city enhancement). We can improve our chances by eliminating wind directions that are especially problematic.

Line 453-454: "Background random error is a function of sample size and decreases as a number of independent samples increase". As mentioned earlier, I don't think that this is the interesting part of the temporal variability analysis and is already a property of standard error. I think it would be more interesting to discuss how different the standard deviation in the background concentration differences are between wind directions. It would be interesting to know if the sample size at which the standard deviation of the background concentrations stabilizes is similar for all directions. This would be a helpful number if it can be assumed to be generalizable. Basing the minimum sample size on the standard error is less generalizable as the required precision may differ, and the variability in background concentrations would differ between regions.

Part of this question is already addressed above. It would be preferable to carry out an analysis presented in this paper for any other region in question where CH4 emissions are of interest. As you point out, each region is unique and presents its own challenges. But if one is able to understand what issues may arise when beginning their top-down estimation for a particular area, they may be able to avoid large errors simply by better constraining their experiment.

Figure 9: This caption should be expanded in order to make each figure stand-alone.

Done.

Response to Referee # 3

The narrative of the paper would be improved by more directly addressing the relationship between this paper and the previous investigations of methane from Indianapolis (all of the papers cited in Figure 1). The Introduction does a fine job of giving a general review of the previous studies, but the connection to the central problem of different studies/methods yielding different results is weaker in the rest of the sections.

We would like to thank the reviewer for their insightful suggestions. We tried to address noted issues to the best of our ability. Each comment of the reviewer is addressed below individually.

For example, how are the methods for background estimation in this paper different than the methods used in the previous studies?

This is a very good question. The description of methods to determine background used by other studies is now added to Section 2.4 (methodology section has been reordered due to requests from another reviewer).

The point of the article in question is to emphasize the challenge that background estimation may pose for flux estimation. There is no perfect method for background estimation; it is something that must always be addressed differently depending on a situation at hand. For instance, for aircraft mass balance studies there are 3 main methods to determine a background value. First is to pick a smallest edge value of a transect. Second is to linearly interpolate background field of a transect from one edge to another edge. Third is to use an upwind transect as a background field. In the case of an inversion, it is common to pick a tower that is located generally away from the sources and has on average smallest overall enhancement. In the current paper, background is chosen using 2 different criteria. For both of these criteria, each wind direction (using 8 main wind directions) is considered separately. This is possible because we have 9 towers and we can always change our background tower based on a wind direction. This is an advantage as with this strategy we may be able to better reduce contamination of local sources and to better represent upwind sources that are approaching the city.

In the Conclusions, you recommend a strategy for background estimation for the aircraft mass balance method, but you did not describe how it was done in the previous studies.

Here is what we wrote: “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.” The strategy recommended is intended to better help an analyst to understand the background conditions for a given day. It may be possible that the day in question should not be used for flux estimation due to complex and variable background. We have not seen this type of critical discussion in the articles that employed aircraft mass balance methodology. Generally speaking, there are some “standard” methods of background calculation for an aircraft approach. Unfortunately no method is perfect and each one has its disadvantages. First is to pick a smallest edge value of a transect. This method could be wrong when the upwind plume is narrow and is not represented well by the edges of a transect. Second method is to linearly interpolate background field of a transect from one edge to another edge. This method is better, but it may not always account for the complex gradient of background that may occur over the plume. However, this method could help to identify that background is complex and the day should not be analyzed further for a flux value. Third is to use an upwind transect as a background field. This is potentially the best method if a case is in steady state, but realistically the issue here is lag. Plume is always moving, so upwind and downwind transects are not sampled simultaneously. If typical aircraft mass balance approach assumptions are satisfied this should work well, but from our experience that is not always the case and therefore a closer analysis must determine if a given day is acceptable for a flux estimation or not depending on how background is behaving.

So our goal was not to introduce a new background methodology for aircraft studies necessarily, rather to suggest caution when such data is analyzed. However, we did add the background estimation methodology of aircraft studies as you suggested (Section 2.4) as well as some recommendation regarding background for aircraft data analysis (conclusion).

Also, the Lamb paper identifies a major discrepancy between top-down and bottom-up estimation of the non-biological portion of Indianapolis methane emissions, and the current paper is a follow-on to that paper, but it is not clear whether this paper resolves that question or not, or only partly resolves it.

In this paper we are unable to address this question directly as we have no measurements of ethane (C2H6), which is a tracer gas used by Lamb et al., 2016 to separate biological CH4 from non-biological CH4. However, we can answer this question indirectly by estimating total emissions of the city and subtracting “known“ biological sources (such as landfill, see the comment about landfill for more details) from that total. The residual is hypothesized to originate from non-biological sources such as NG. So we think it is likely there is no major discrepancy between top-down and bottom-up solutions. We think that some of the top-down solutions in Lamb et al. 2016 are biased high and should be lower more in line with bottom-up estimations. As we point out later uncertainty remains, but the high top-down estimates could be potentially explained by the erroneous assumptions in analyzes.

Specific Comments:

Line 19 – details about the type of analyzers and the measurement heights in the abstract are unnecessary and irrelevant.

The details are now removed.

Line 103: Please describe briefly how/why the landfill emissions are considered wellknown.

Originally this statement was based on the Greenhouse Reporting Data (GHGRP), which gave very similar emissions values for this landfill over 2010-2015 time frame. This is supported by Lamb et al. 2016 paper that also cited GHGRP as a reputable source for the landfill. However, 2016 and 2017 GHGRP indicates a 70% decrease in landfill emissions. That seems unrealistic given that our towers near the landfill do not show any decrease in average methane mixing ratios over these years in comparison with the previous years (Figure R1). We are currently in the process of investigating this discrepancy. So far we received no response from EPA regarding this.

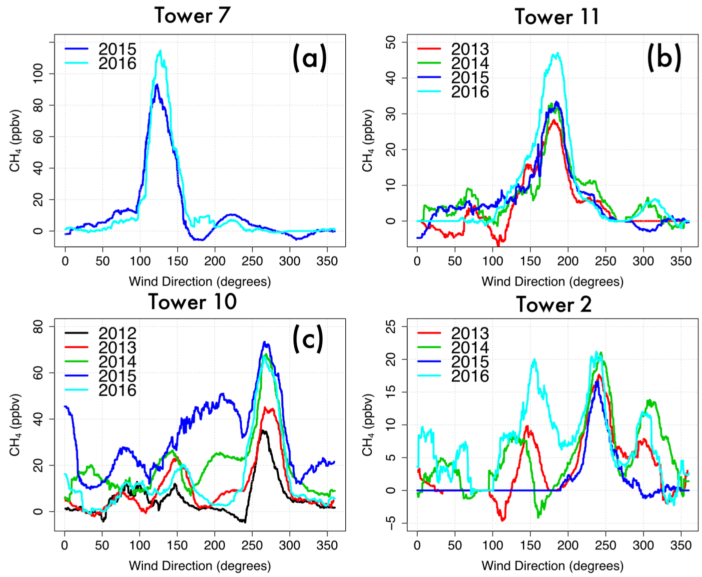

Figure R1. Yearly CH4 enhancement directional profiles for 4 INFLUX towers located in the city of Indianapolis: (a) tower 7, (b) tower 11, (c) tower 10, and (d) tower 2. Note that there is a lot more variability in the towers closer to the landfill (Towers 10 and 11). Often the plume is unable to fully mix in a close proximity to these towers resulting in the higher variability. Landfill peak is apparent in all of the towers. Tower two, which is more representative of a fully mixed plume, shows no dramatic change in the landfill emissions from year 2015 to year 2016.

For more details see (this needs to be copied and pasted into a browser):
https://ghgdata.epa.gov/ghgp/service/facilityDetail/2017?id=1002683&ds=E&et=&popup=true

We do have some potential evidence for variability in landfill emissions from Cambaliza et al. 2015 article that used aircraft mass balance on five different occasions to calculate methane flux from this landfill. Their average is higher than GHGRP, but their estimation has high uncertainty. They were not able to make consistent calculations. Their city totals seem abnormally high on two of the flights, which may indicate there were issues with a background assumption. In addition, Cambliza et al. 2015 used California Landfill Methane Inventory Model v 5.4 or CALMIM to estimate methane emissions from South Side Landfill (SSLF), where results are more consistent with GHGRP.

Overall, we think we know an approximate value of the SSLF methane emissions, but this section in the paper about the landfill is now rewritten to reflect a truer state of knowledge regarding SSLF landfill emissions.

Section 2.2 – There are extra details here that are not very relevant to the current paper and have already been described in other papers. This section could be made more concise.

The section's length has been slightly reduced.

Section 2.4 (Sources) – Although references are given, the source of the numbers in this paragraph is not clear. Are they from direct measurements, an inventory, or something else?

The sources of theses numbers are a combination of bottom up and top down estimates to give a range of possible emission values. This section is now rewritten as another reviewer also asked about this (it is 2.7 now).

Section 2.4 (Background) – As written, I had to read this section many times to try and understand it and I'm still not sure I fully understand the two methods, so it needs to be re-worked for clarity. Why is a viable method not to take the lowest measurement among all towers at a given hour as a background? How do these two approaches compare to those used in the cited aircraft and tower-based top-down studies?

We apologize for the confusion. Also this section should be numbered 2.5, we will correct the numbering in the next version of the paper.

The main point of this section is that we can pick multiple backgrounds for a given situation and it would be impossible to say which one is better. Hence, two arbitrary but acceptable backgrounds are chosen here to estimate methane enhancements. If background is uniform or closely so, as sometimes stated in literature, then we would see no significant difference between the enhancements calculated with different backgrounds. Yet we show here that this difference is significant and choice of background matters.

We edited this section to try to clarify this point.

It is not a viable method to take the lowest measurement among all towers at a given hour as a background because the background we are interested in is not always the lowest value as illustrated in the schematic shown in Figure R2 (although such method may be a good compromise – our first criterion is somewhat similar to this approach). True background lies upwind of the city (or the downwind tower of interest from which the enhancement is calculated) and it is not the lowest value. Because we are trying to identify the enhancement specifically from the city we must subtract exactly what is coming into the city. The methane is heterogeneous as described earlier and therefore it is a challenge to identify exact background even at a not-so-large scale as Indianapolis.

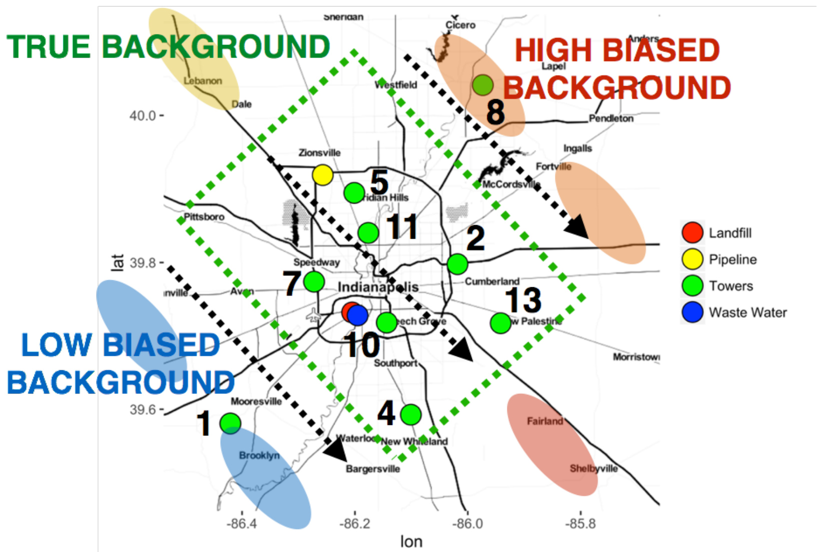

Figure R2. Theoretical representation of boundary layer CH_4 plume heterogeneity across Indianapolis and nearby areas when winds are from the northwestern direction. The colors indicate relative CH_4 concentrations where yellow is neutral, blue is low, and red is high. Green dashed lines indicate the assumed boundaries of Indianapolis. Also shown are INFLUX towers with CH_4 measurements and known sources.

The approaches used in Criteria 1 and 2 are not entirely different from aircraft and inversion studies, but it is exactly the point. All of these approaches are acceptable, it is just important to understand what kind of uncertainty they are causing. In some cases, we recommend not to perform flux calculation since the uncertainty is too large due to complex background. Identifying those complex days is whole another topic, which deserves a separate paper, but we do think this is very important and must be emphasized.

Here are a couple of figures that show the heterogeneity of methane in Indianapolis. Indianapolis CH4 observations indicate highly variable background with day-to-day variations at times reaching 150 ppb (Fig. R3). Similarly, WRF-CHEM simulations show occasional spatial non-uniformity of CH4 (Fig. R4).

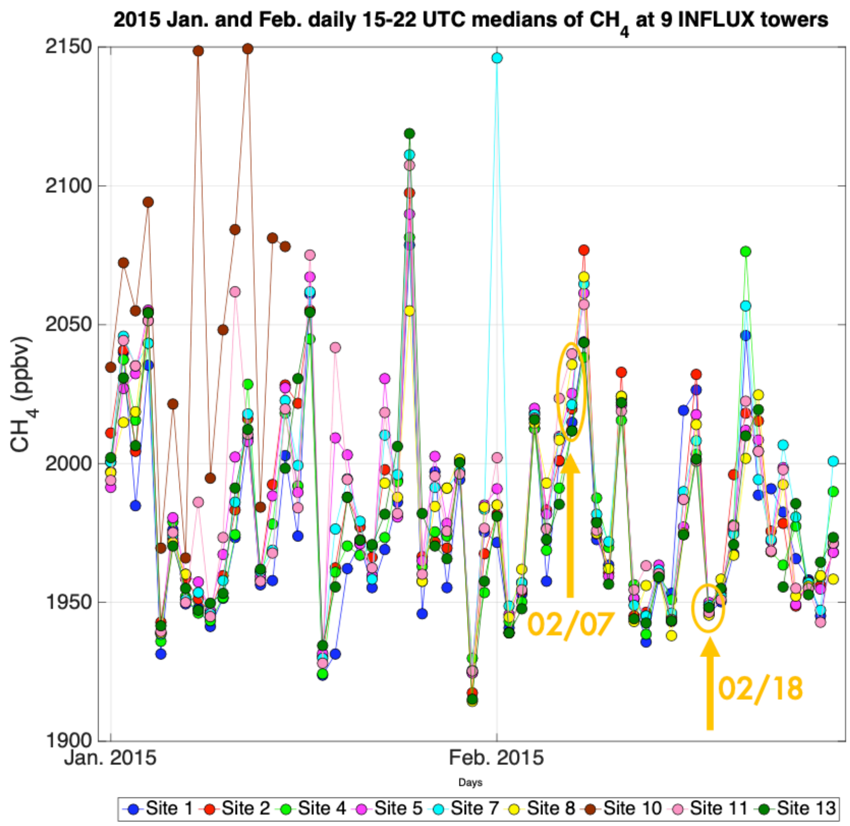

Figure R3. Daily CH_4 medians over 15-22 UTC at 9 INFLUX towers.

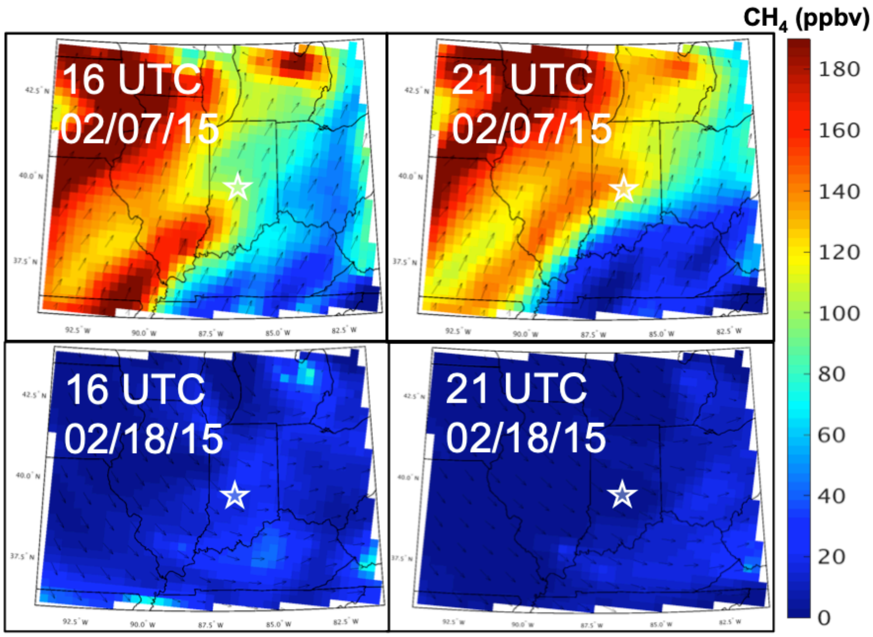

Figure R4. 27 km WRF-CHEM simulations of CH_4 enhancements (ppb) for 2 different days using EPA 2012 CH_4 emissions (Maasakkers et al, 2016): **(top left)** late morning on 02/07/15 **(top right)** late afternoon on 02/07/15 **(bottom left)** late morning on 02/18/15 **(bottom right)** late afternoon on 02/18/15.

Section 2.5 – How far away are your receptors and wind measurement locations since you say that this method requires them to be nearby?

In this case towers are the receptors and they are labeled as green diamonds in the figure below (Fig. R5). The wind data is gathered from the 3 stations identifiable by white colored shapes on the figure as explained in section 2.3. We found that the wind measurements are generally consistent between these 3 stations and therefore their combination is well representative of the city overall. Perhaps occasionally tower 8 may not be represented perfectly by these winds, but we do not think that such situation occurs often.

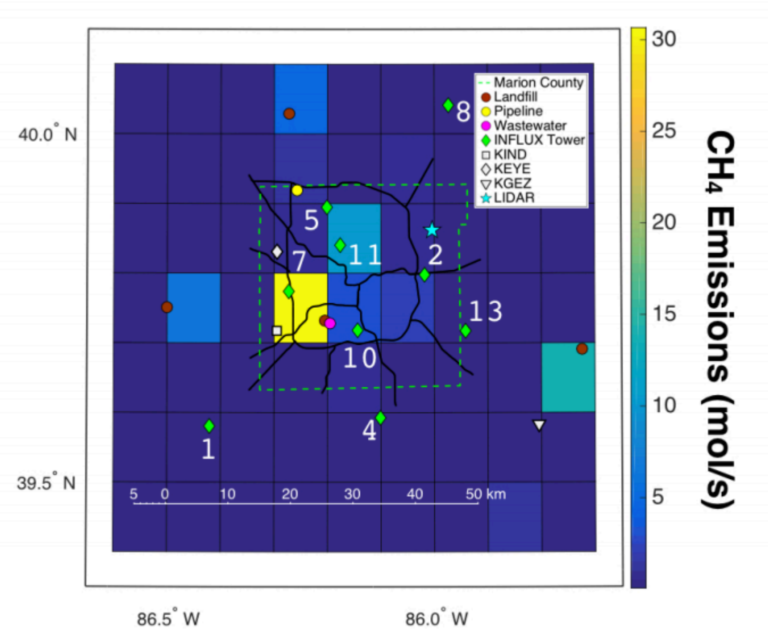

Figure R5. Map of the primary roads in Indianapolis, INFLUX towers, lidar system, weather stations, and a few CH_4 point sources plotted over the gridded CH_4 emissions (mol/s) from the EPA 2012 Inventory (Maasakkers et al., 2016). The gridded map of emissions includes emissions from these point sources; their position is provided to aid in interpretation of the observations. The dashed bright green line denotes Marion County borders.

Section 2.6, Line 261 – Which towers were used for which wind directions?

This question is answered at the end of Section 2.6 (lines 283-287). Here are the relevant sentences: "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."

Section 3.1 - The first three sentences could probably be condensed into one concise sentence without losing any meaning. - I am having trouble squaring your description of the domain differences with my understanding of the Lamb paper. By my reading, the Lamb paper describes developing an inventory for the larger *domain, but you say that the inventory covered mostly only Marion County. I find it hard to believe that Lamb et al. would perform such a detailed analysis and accidentally compare totally different areas for the inventory and tower inversion. What am I misunderstanding? - With the revised inverse emissions estimate, it is not clear whether you've replicated the methods of the inversion in Lamb et al. over a smaller domain, or whether you've used the boundary layer budget method described in the method section.*

Unfortunately there is indeed an inconsistency between domains used by Lamb inventory and inversion. The domain used by the inversion contains 3 landfills that are not part of the Lamb inventory. We are not sure how this happened, but that is what we are seeing when we read Lamb's paper. The Figure R3 shows domain used by the inversion. You can find this domain in the supplemental of Lamb paper. We also have access to the prior used in the inversion of Lamb paper and these 3 landfills are in there. You can see landfills marked by the brown dot in the Figure R5.

The point we are trying to make here is that it is imperative to be very careful when such comparisons are performed. It may seem obvious that boundaries of emission areas need to be the same when they are compared, but it seems that occasionally such detail can get neglected when analysis considers many other complex parameters. In this we work we would like to emphasize the importance of this initial step.

The revised inversion estimate just shows what would happen if the original inversion had solved for Marion County. We asked the person who did the original INFLUX inversion to rerun his code for the whole region and then just for the Marion County. The result is shown in Figure 1 of the submitted article. Boundary layer budget method is not used for this result. It is used later in the paper to try to understand temporal variability of CH_4 flux in Indianapolis. Clarification is now added to section 2.1.

Section 3.2 - How much of the data are filtered using the criteria you give? - Line 372: Suggest: "Because Indianapolis is a relatively small emitter of methane, and because there are relatively large sources outside of the city, uncertainties due to background estimation are comparatively large."

The answer is at the beginning of the Section 3.2. Here are the relevant sentences: "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)."

Because the city of Indianapolis is surrounded be sources that are similar to its CH4 flux magnitude it is not surprising that occasionally there are complex background scenarios that are difficult to address (include modeled map?). If Indianapolis was much larger source than its surrounding sources background would not be a big issue, but in this case it is not so. The goal of our background variability study is to show how variable CH4 background is on average at this location. The data was filtered only to eliminate extreme cases, but other cases, which are likely to be used by inversion studies, or even aircraft methodologies were left in. Another important point of our study is that inversions and mass balances should be carefully filtered to exclude complex background days. However, we did not see an evidence of that filtering in case of studies performed at Indianapolis.

Thank you. Suggestion is incorporated.

Section 3.4 - Isn't the result that the SSLF is the largest and strongest source in the city consistent with your prior understanding, as described in the methods? - Looking at figure 8, I can't tell which enhancements around T10 you think are from SSLF and which are from NG leaks. - How do you square your findings that emissions from NG is not a significant portion of emissions with the findings in Lamb et al. that approximately half the emissions are from NG using ethane as a tracer? Are you saying that you can't see the sources because they are below your detection threshold or that their signals or swamped by that of the SSLF, or are you saying that their existence is entirely not supported by the data? On the one hand, you say there is not much evidence for a diffuse NG source, but on the other hand, even after adjusting the domain, your top-down estimate is still much higher than the inventories. You should at least acknowledge this contradiction or remaining possible existence of unknown sources. - Line 488: The description of *"occasional" on seems incorrect since this apparent signal shows up in Figure 8, which represents a two-year average.*

It is indeed consistent with our prior understanding; however, there are views that natural gas could be larger than SSLF as a source of CH4. We agree that Figure 8 does not provide definitive answer, yet it allows us to see that the landfill is likely the strongest source in the city as no other point or area source is making such a noticeable enhancement at any of the towers. Landfill is located to the west of tower 10 and therefore the plume that arrives from the west is unquestionably belongs to landfill. Even in Lamb et al. 2016 we see similar result by observing low ethane to methane ratios from southwest of tower 11. In general, the ethane to methane ratios provide an insight into the composition of a given plume measured at a certain point, but overall city composition is hard to measure unless the tower is downwind of the whole city. The issue here is that plumes may come outside of the city and we have no way of separating plumes inside the city from the outside using tower 11. Lamb article does say that they ran an inversion of sorts, but unfortunately they poorly explain how they did it and the assumptions that were made in those calculations make the result largely uncertain. We spent sometime trying to understand what they mean by "source footprints" in their supplemental material (S3.4); however, this terminology is not standard and therefore we have no idea how they arrived at their answer. Also their sample size is just 11 days, which is not enough to make a definitive conclusion. And their domain includes sources outside of Marion County (they even mention multiple landfills), which makes this even more confusing.

With regard to adjusted inversion, there is still uncertainty in that result since it only used 3 towers at most (the tower record is sparse in 2012-2013 time frame). In our second attempt of running this inversion it seemed sensitive to the prior, which hinted that the system might need some more experimenting and testing. However, we admit that we cannot say for sure there is no significant diffuse source at Indianapolis comparable to landfill. But the evidence suggests that it is not as large as previously was suggested. The goal of this paper is to show that some of the very large values attained by the top-down estimates in this case appear to be unconvincing since adjustments in background and domain do affect the flux values. We are going to leave a space for potential diffuse source as suggested, but we must stress that its existence is highly uncertain given the data at this point.

Technical Comments

Line 61: Suggest: "...atmospheric methods and inventory assessment have sometimes succeeded..." Are there are cases when these two criteria have been met but reconciliation has not been achieved?

This depends on the definition of reconciliation. However, it may be possible that a study found an agreement between an inventory and a top-down methodology, while another study did not find that for the same region. Then perhaps reconciliation is under question.

Line 70: Suggest: "Recent studies of urban CH4 emissions in California indicate..."

Done.

Line 72: The phrasing "large NG infrastructures" is strange and evokes large individual pieces of equipment, which I don't think is your intent.

Done.

Line 79: Suggest: "in" → "for"

Done

Line 85: Suggest: ". . .comprised of irregular or periodic in situ aircraft measurements, continuous in situ observations. . ."

Done.

Line 91: Suggest: "well-suited" → "designed"

Done.

Line 94: Delete: "Recently"

Done.

Line 103: Suggest: "Uncertainty in total emissions is driven by. . ."

Done.

Line 132: Suggest: "tubes secured" → "air collected"

That is probably okay as it is.

Line 139: Suggest: "inflow" → "sample air"

Done.

Line 152: The given link re-directs to some other website.

Fixed.

Line 154: Suggest: "The anemometers are located at about 10 m AGL."

Done.

Line 200: Suggest: ". . .based on two different sets of criteria. Both approaches identify. . ."

Done.

Line 316: Suggest: "inventory" → "inventories (Fig. 1)"

Done.

Line 352: The meaning of the numbers "2 to 150" is unclear.

These numbers indicate an experiment sample size. Clarification is added. The idea is to see by how much the uncertainty decreases if the sample size is 150 (arbitrary large sample size) values. In theory each value could be used to solve for flux. But with sample size of 2 the uncertainty is large. This is an attempt to try to figure out how much data is optimally needed to solve for the emissions from the city. But due to various assumptions this is just an approximation. Topic related to this has been covered in great depth in the response to another reviewer. The revised article will contain some changes in that section.

Line 385: Suggest: "at least twice as high" → "approximately twice as large"

Done.

Line 396: Suggest: "did not change significantly between 2014 and 2016."

Done

Line 519: "Dennis" – Do you mean Brian?

Yes, sorry.

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

Nikolay V. Balashov[1], Kenneth J. Davis[1], Natasha L. Miles[1], Thomas Lauvaux[1,2], Scott J. Richardson[1], Zachary R. Barkley[1], Timothy A. Bonin[3, 4]

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[2]Laboratory of Climate Sciences and Environment, Gif-sur-Yvette, France
[3]Cooperative Institute for Research in Environmental Sciences, Boulder, Colorado, USA
[4]Chemical Sciences Division, National Oceanic and Atmospheric Administration, Boulder, Colorado, USA

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 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 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 CH_4 mole fractions are shown to be heterogeneous and temporally variable. Variability in a single point background mole fractions

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observed at any given moment 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 from 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). No significant unknown CH_4 sources are found. The data from the towers suggest that the strongest CH_4 source in Indianapolis is South Side Landfill. 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 CH_4 emissions in Indianapolis. However, some uncertainty regarding occasional significant CH_4 leaks from NG distribution exists. 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 are 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

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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 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} (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 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 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, studies indicate that ~60-100% of Boston CH_4 emissions are attributable to the NG distribution system (McKain et al., 2015; Hendrick et al., 2016). 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 that result from the 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 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.

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

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Lamb et al. (2016) compared Indianapolis CH_4 emissions estimates from a variety of approaches, which are 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) study that 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). 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

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(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). 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 has been 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

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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.

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, which were reported on the WMO X2004A scale.

2.3 Meteorological 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

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stations is available at https://www.weather.gov/media/asos/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 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.

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 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

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flux estimate (Cambaliza et al., 2014; Lamb et al., 2016). The CH_4 mole fraction enhancement is defined as,

$$C_{enhancement} = C_{downwind} - C_{bg} \tag{1}$$

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 at Indianapolis mentioned in this article 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 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

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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

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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 (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,

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

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).

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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 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.

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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 upwind 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.

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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,

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

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 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 is taken from KIND weather station. The difference between concentrations

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$C - C_b$ 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 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

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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 and GHG reporting program) and 45 mol/s (aircraft; Cambaliza et al., 2015) depending on an emission estimation methodology. 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 contigent on how much of the total city emissions are coming from NG. 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 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 21-67% (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. There could also be a possibility of temporal variability in either of the sources as described in the section above.

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3 Results and discussion

3.1 Inversion and city boundaries

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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.

It is difficult for us to critically assess this result without performing complex sensitivity analysis with this inversion system (which is not the goal of this article), but it is important to note that inverse modeling is prone to errors in prior, background, and meteorological transport. Also, Figure 3 indicates that tower data were sparse in 2012-2013 perhaps contributing to potential error in the inversion.

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

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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 enveloping 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.

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

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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

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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

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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 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 higher error bar of Lamb et al. (2016) calculation. Another possible scenario is that SSLF emissions are

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higher than what is currently assumed. Given these complexities, uncertainty regarding the exact emissions from NG distribution at Indianapolis still remains.

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 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),

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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 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 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, it is imperative to be careful and acknowledge the limitations of the current analysis. 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.

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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; 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). Although it is difficult to generalize with certainty regarding this particular inversion, significant errors are possible in an inversion system due to lack of data, false assumptions regarding prior, biased background, and erroneous modeled meteorological transport.

To better understand background variability at Indianapolis two different but acceptable background towers are selected 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 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

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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. Low sample volumes, 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, noticeably smaller than 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 inventory estimates for Indianapolis than their individual components as 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

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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.

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

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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 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 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

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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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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

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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

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Figures

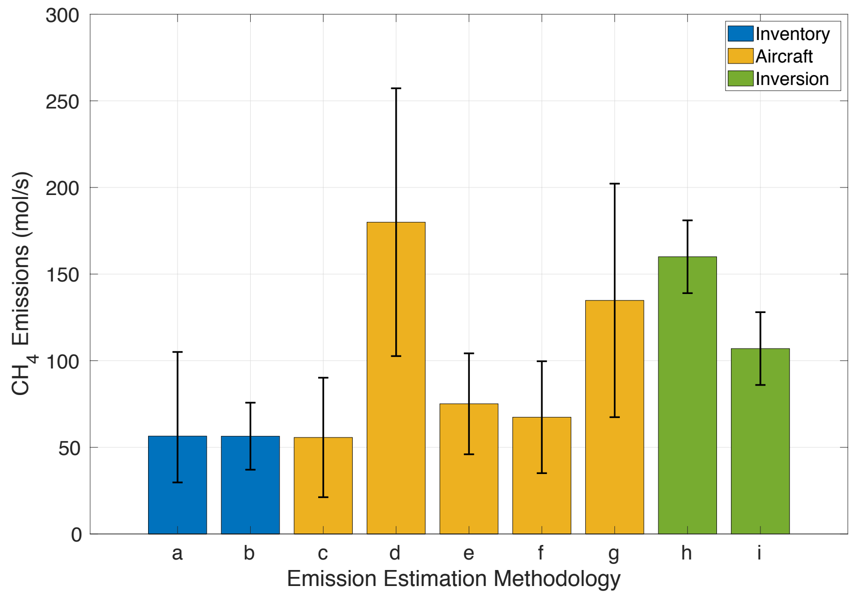

Figure 1. Various estimates of CH_4 emissions at Indianapolis. **(a, b)** Bottom-up estimates of CH_4 emissions conducted by Lamb et al. (2016) in 2013 and Maasakkers et al. (2016) based on the EPA 2012 inventory respectively. Error bars show 95% confidence intervals (for more details see above-mentioned 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 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).

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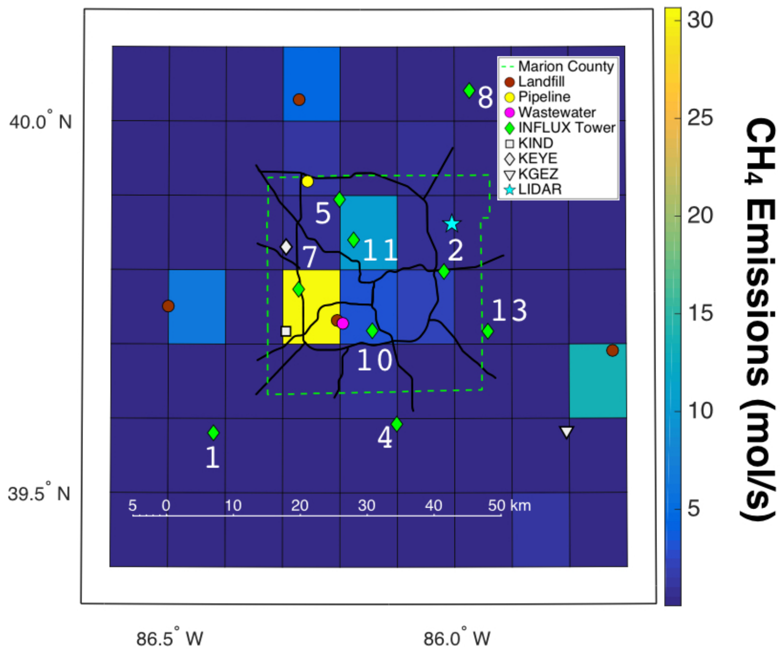

Figure 2. Map of the primary roads in Indianapolis, INFLUX towers, lidar system, weather stations, and a few CH_4 point sources plotted over the gridded CH_4 emissions (mol/s) from the EPA 2012 Inventory (Maasakkers et al., 2016). The gridded map of emissions includes emissions from the mentioned point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

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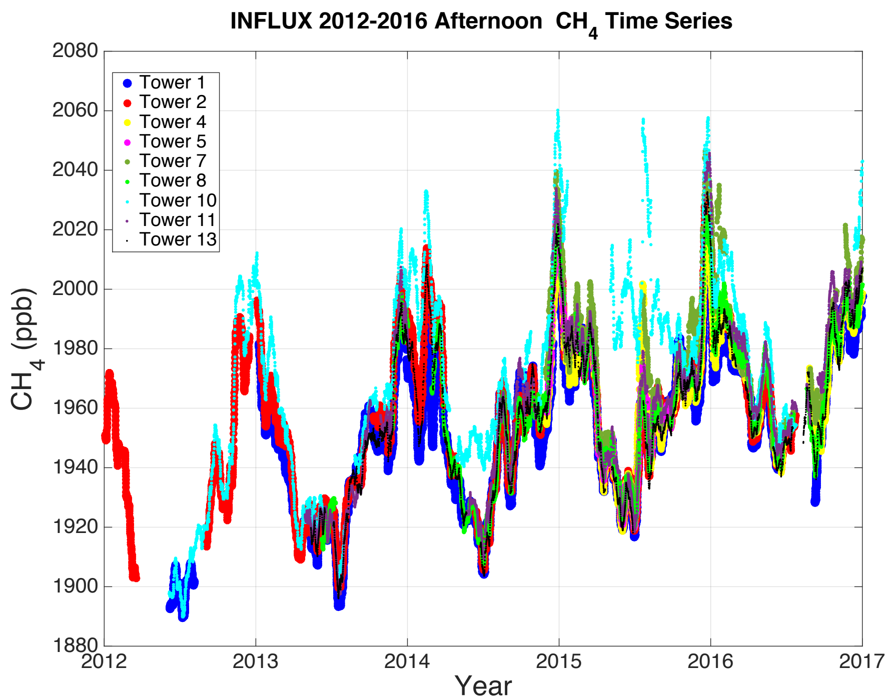

Figure 3. 20-day running average of afternoon (12-16 LST; the hours are inclusive) CH_4 mole fractions as measured by the INFLUX tower network (highest available height is used) from 2012 through 2016.

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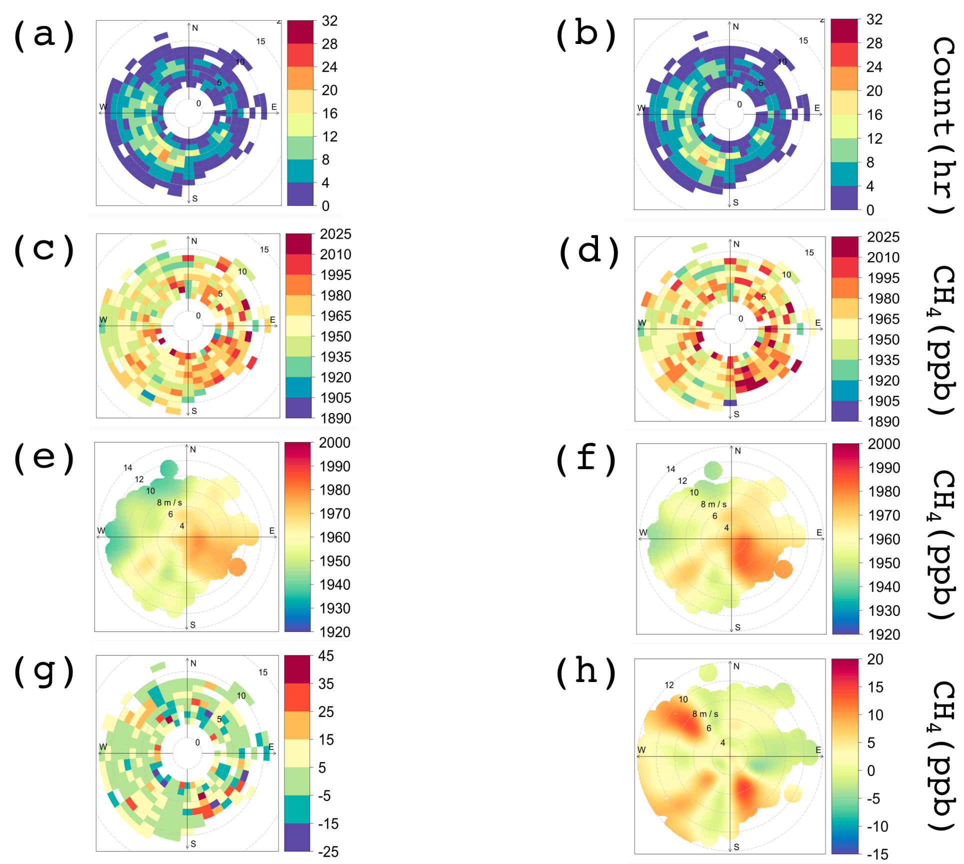

Figure 4. Frequency and bivariate polar plots of CH_4 background for Indianapolis using data from 12-16 LST, November 2014 through December 2016 given 2 different criteria (Table 1). **(a)** Polar histogram indicating a number of hourly measurements available using criterion 1. **(b)** Same as (a) only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

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Deleted: 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 ... [8]

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Moved up [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.

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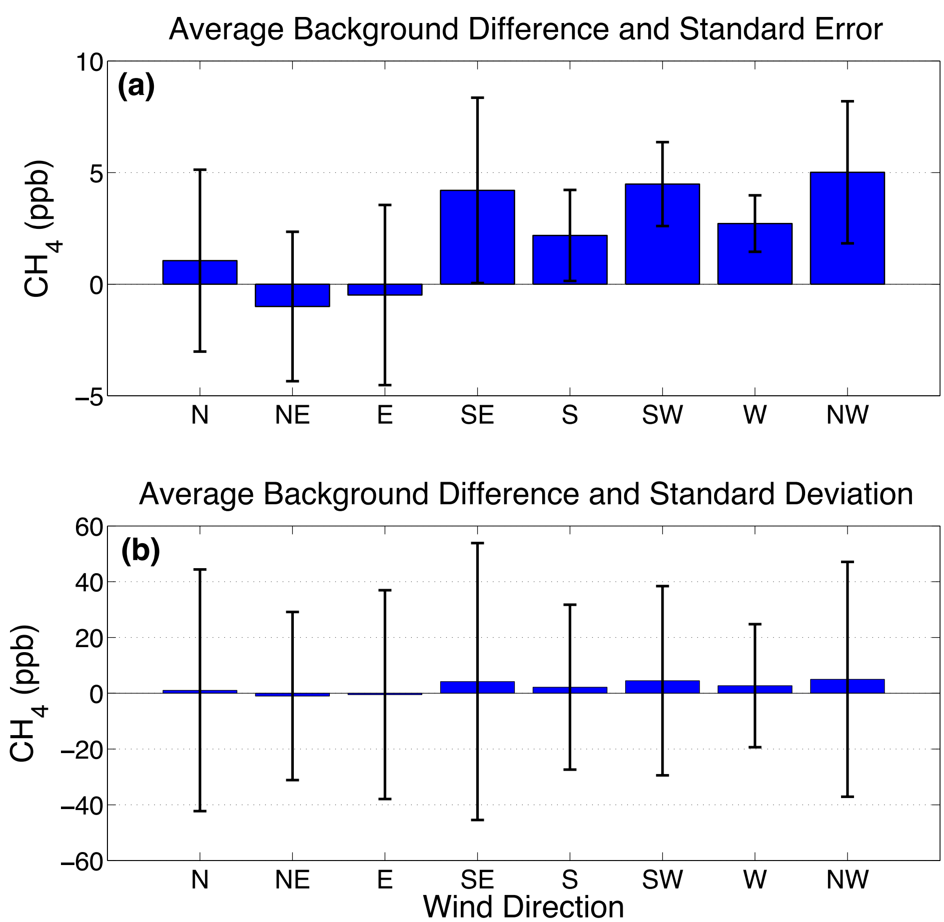

Figure 5. Average of the differences between criteria 2 and 1 CH_4 backgrounds at Indianapolis as a function of wind direction. These averages are generated from the same data that is used in Figure 4 and reflect results shown in Figure 4g. Error bars indicate in (a) 2 × standard error and in (b) 2 ×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

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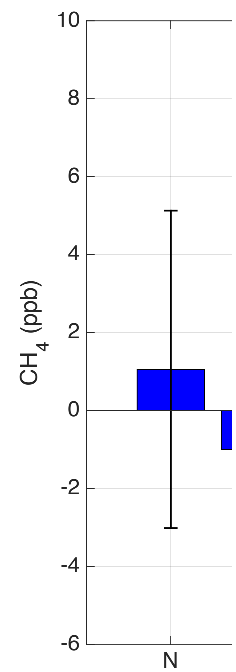

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Figure 5. Average of the differences between criteria 2 and 1 CH_4 backgrounds at Indianapolis as a function of wind direction. These averages are generated from the same data as used in Fig. 4 and reflect results shown in Fig. 4g. Error bars indicate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

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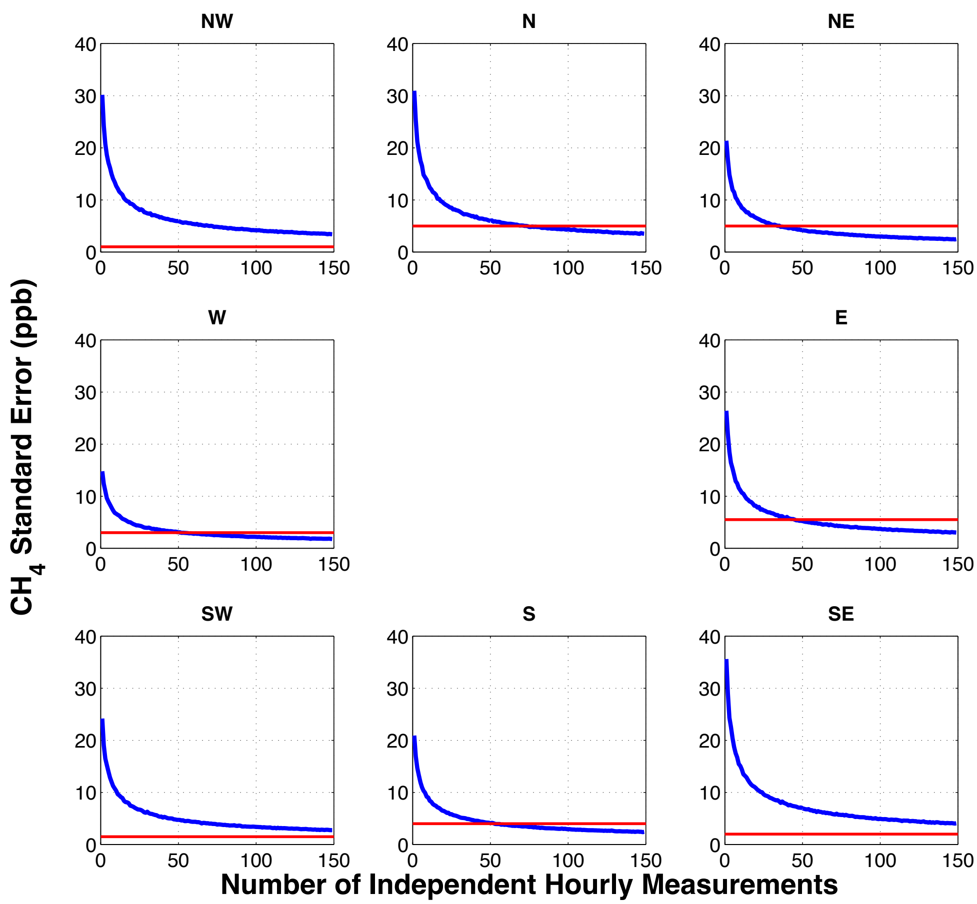

Figure 6. Bootstrap simulation of the standard errors × 2 in Indianapolis CH_4 background mole fraction differences (between criteria 2 and 1) as a function of sample size and wind direction (see text for details). Thresholds for each of the wind directions indicate a random error threshold needed for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

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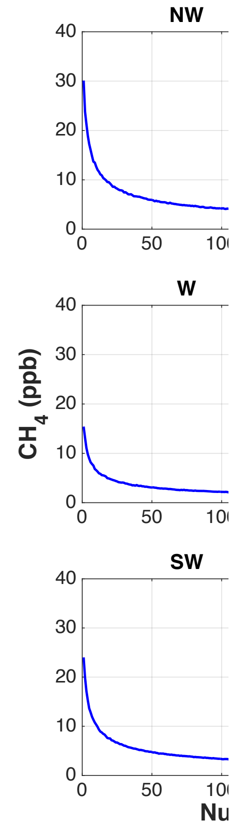

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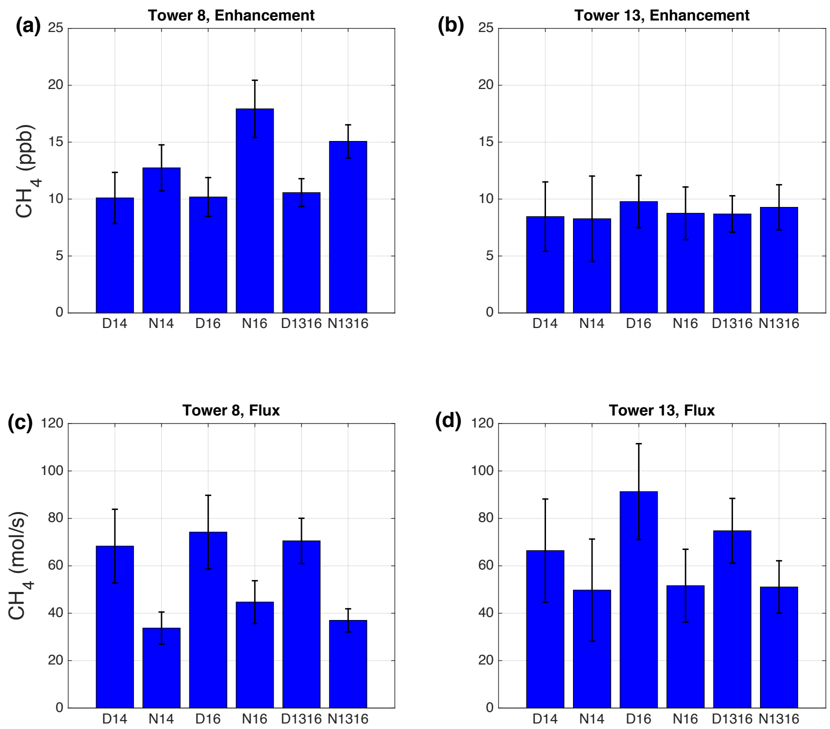

Figure 5. Averages of the daytime (D) and nighttime (N) CH_4 enhancements and fluxes at INFLUX towers 8 and 13 for years 2014 (14), 2016 (16), and 2013-2016 (1316). The error bars represent 95% confidence interval of each mean value. **(a)** Estimates of CH_4 enhancements from tower 8. **(b)** Estimates 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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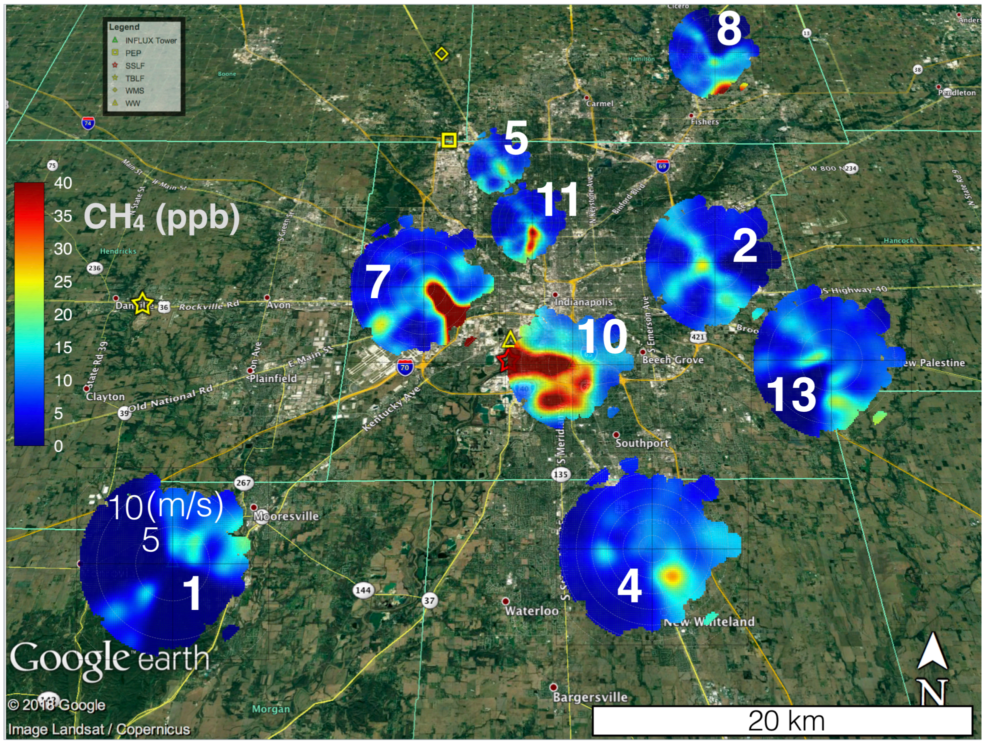

Figure 6. Google Earth image overlaid with bivariate polar plots (section 2.5) of the CH_4 enhancements at 9 INFLUX towers in Indianapolis using the criterion 1 background (Table 1) for full years of 2014 and 2015 over the afternoon (12-16 LST). The wind speed scale is only labeled at site 1; other sites 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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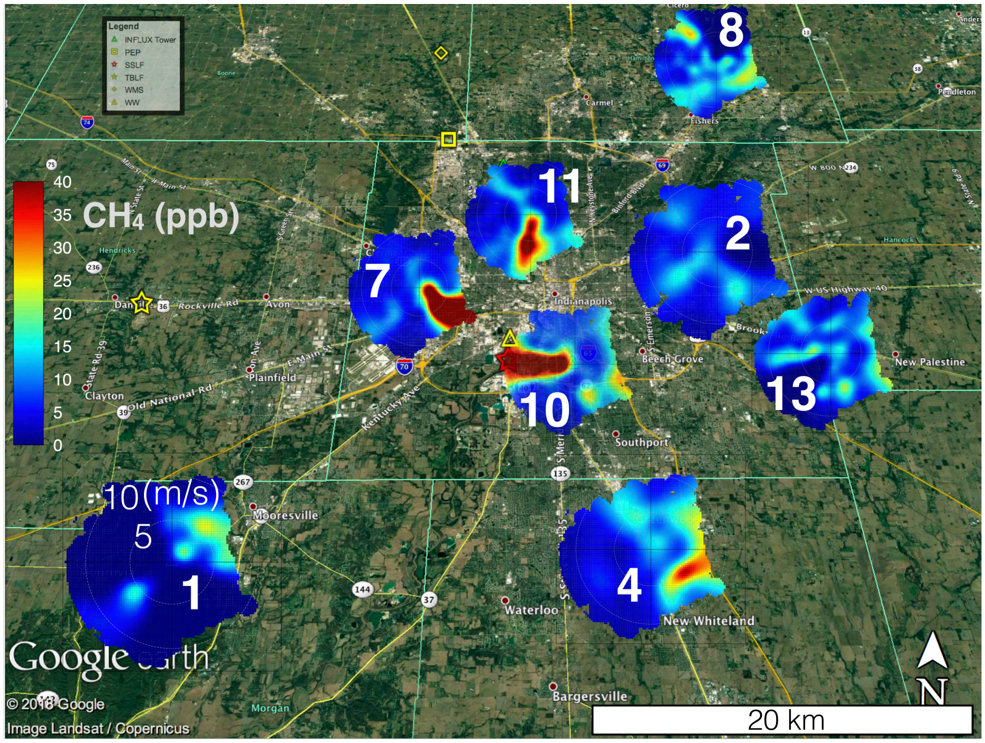

Figure 7. Google Earth image overlaid with bivariate polar plots (section 2.5) of the CH_4 enhancements at 9 INFLUX towers in Indianapolis using the criterion 1 background (Table 1) for year 2016 over the afternoon (12-16 LST). The wind speed scale is only labeled at site 1; other sites follow the same 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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