

Ms. Ref. No.: AMT-2018-254

Title: Quantification of CO₂ and CH₄ emissions over Sacramento, California based on divergence theorem using aircraft measurements

Dear Dr. Christoph Kiemle,

*Thank you very much for your efforts on behalf of our paper titled “**Quantification of CO₂ and CH₄ emissions over Sacramento, California based on divergence theorem using aircraft measurements**” submitted by Ju-Mee Ryoo, Laura T. Iraci, Tomoaki Tanaka, Josette E. Marrero, Emma L. Yates, Inez Fung, Anna M. Michalak, Jovan Tadic, Warren Gore, T. Paul Bui, Jonathan M. Dean-Day, Cecilia S. Chang.*

We found the reviewers’ comments helpful, and we have addressed the reviewers’ comments and suggestions and incorporated them in the revised manuscript. Enclosed is a point-by-point response to the review comments.

Thank you very much again for your support, and we look forward to hearing a positive decision from you soon.

Sincerely,

Ju-Mee Ryoo and Coauthors

Response letter to Reviewer #1

Responses from the Authors are given in blue italicized text throughout: Thank you for your encouragement and helpful comments. We addressed them below.

I would like to thank the authors for making substantial revisions to their manuscript and improving it significantly. I will accept the publication of this article after a couple of minor revisions.

Paragraph starting on line 430: Please discuss uncertainty due to background in some detail. From my experience it could contribute significantly to the overall error. Please mention this part somewhere in this paragraph or section.

We discussed uncertainty due to the background in section 3.3.

Figure 4: please add wind direction as you did in Figure 5.

Thank you for the suggestion. We added wind direction in Fig. 4 too as you suggested.

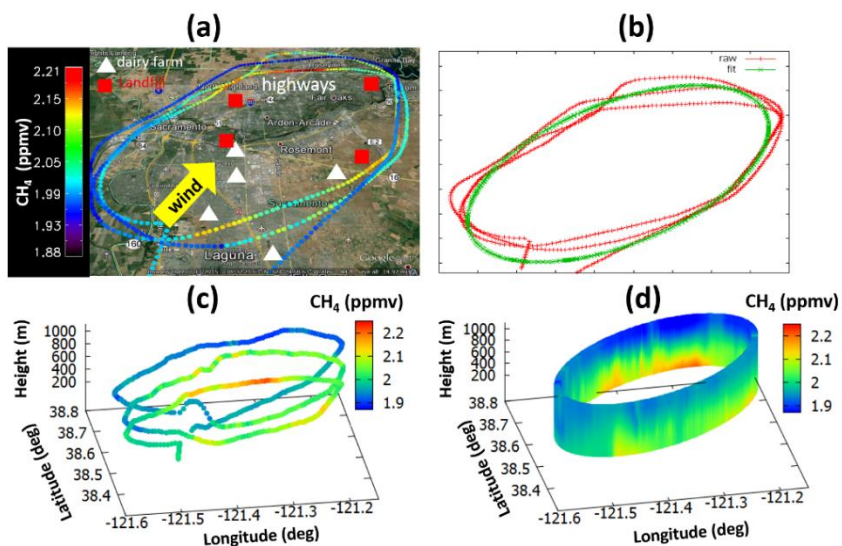


Figure 4: (a) Map of AJAX flight tracks colored by CH₄ mixing ratio for November 18, 2013, plotted in Google™ Earth. (b) The data (red) fitted to an oval (green). The observed CH₄ mixing ratios (c) are kriged to generate the cylindrical surface (d). The axes of the oval are approximately 25 and 40 km. The yellow arrow represents the dominant wind direction.

Response letter to Reviewer #2

Responses from the Authors are given in blue italicized text throughout: Thank you for your encouragement and helpful comments. We addressed them to the best of our understanding and appreciated your guidance on areas where you found our discussion and plots unclear.

Review of Revised Submission:

Quantification of CO₂ and CH₄ emissions over Sacramento, California based on divergence theorem using aircraft measurements: Ju-Mee Ryoo et al.

General Comments:

After the revision this manuscript has improved tremendously. The structure is much clearer and results are presented more uniformly. Figures and tables are well presented and include all the necessary information for the conclusions. Furthermore, a lot of issues due to language use have been resolved. Still, I have two minor revision points I would like to be addressed clearer before publication:

First, I believe that your treatment of interpolation/extrapolation methods is not complete. You show different methods and compare them, but never show how they influence the final emission estimate. A small section on "Sensitivity of the calculated flux to interpolation/extrapolation method" would be great at the beginning of chapter 3.2.

As suggested, we added the section. Please see it in Section 3.2.1 in the revised manuscript.

Second, the chapter on uncertainties needs restructuring and clarification. Several factors of uncertainty are named at several locations in the text, e.g., wind (l. 415, l. 423, ll. 441-446) and kriging (ll.411-414 and ll. 425-429). Please consolidate these sections for better readability. Your method (ll. 430-432) is mentioned after some factors of some uncertainty, but afterwards (l. 432 ff) you start listing additional factors (PBLH, vertical fluxes) again. Please list all factors first, or present your method first, but don't mix. How do you get to the overall estimate of 10% uncertainty? What is the variance and standard deviation of the kriged mixing ratios? Grid resolution (4%), variogram model (5%), wind measurement (? %), mixing ratios (? %), PBLH (10 %), vertical fluxes (1%), does not sum up to 10%. Please explain.

Thank you for the suggestion. We rewrote the paragraph for the uncertainty section in the revised manuscript and believe this better identifies the terms that are summed in quadrature to determine the overall uncertainty. The estimated kriging variance (its square root is the standard deviation) is a natural byproduct of the kriging. It gives the estimation error, along with the actual estimate of the variable itself.

Additionally, how do your sensitivity studies influence your uncertainty estimate? You do all these sensitivity studies, receiving differences in the emission estimates of up to 80 %. Is that no uncertainty?

*This is a good point. An uncertainty analysis is not the same as a sensitivity analysis. As with a more traditional "error analysis" for the measurement of a physical property, we view **uncertainty** as a*

*quantitative understanding of the impact on the final result of the numerical assignments we made for quantities such as PBLH and the kriged values for CO₂, CH₄, and wind. Beyond this "calculation uncertainty," we are fully aware that fundamental choices in our method (such as how to select the background or whether to use raw or averaged winds) will influence the resulting fluxes. Because these choices do not lend themselves to standard deviations or even "replicate measurements" (as one could consider the analysis we used for uncertainty in PBLH), we consider studies of the influence of these fundamental choices to be tests of the **sensitivity** of the outcome to the choice made in each category.*

Specific Comments and Technical Corrections:

I. 49: "The uncertainty is also impacted by meteorological conditions and distance from the emission sources." These are two factors that you don't discuss in Chapter 3.3.

Thank you for pointing them out. I have incorporated them into the revised manuscript.

I. 50: "The largest CH₄ mixing ratio was found over a local landfill." Is this a key finding of your study? Should I be in the abstract at this position?

This can be one of important findings of our study, but we don't think this finding necessarily needs to be in the abstract. So we removed it from the abstract. We think the impact of the difference in shapes of flight, wind treatment, vertical mass transfer, and background to the flux estimate is most important key messages of this study.

I. 58: "modeling" might confuse here. You did not do any real modeling.

This is a good point. We removed this in the revised manuscript.

I. 61: "identifying emission sources". Is that what you focus on in this paper?

This is not a focus of this paper, but we think this study showed that a closed-shape flight pattern could be a useful strategy for identifying local emission sources (e.g., landfill) and estimating local-scale greenhouse gas emission fluxes too. In that sense, we think we can write this in the abstract.

I. 66: Is air quality the main concern of this paper? GHG are generally not considered as air pollutants.

Good point. Air quality is not the main concern of this paper, but GHG can indirectly affect air quality too because GHG such as CO₂ and CH₄ can result in climate change. Reversely, atmospheric warming associated with climate change has also the potential to increase ground-level ozone and other air pollutants, such as particulate matter. That is why we mentioned it in the introduction.

I.143: Here some sentences are different from the marked-up version of your manuscript.

We accidentally didn't change that part. We updated in the revised manuscript.

I. 149ff: Here a whole section is not in your marked-up version. Which will be the final version?

We update the manuscript many times, so this has not been reflected in the marked-up version. Sorry about that. The order of sentences has been changed while we edit the manuscript (e.g., Sampling

occurred..... in the marked-up version went into the last of the first paragraph of section 2.1). The revised manuscript is the correct one.

I. 315: You should investigate this huge difference in your emission estimates due to wind treatment a little more. Could the cause be a certain meteorological condition? Maybe an accumulation of methane and CO₂ in the outflow region due to low wind speeds. If you then average the wind over the entire loop, it might be extremely overestimated in the outflow region and the flux is too high. What is the difference between raw wind and mass-balanced wind in relation to GHG mixing ratio along the lowest and only flight track within the boundary layer?

The differences in our flux estimates can be explained with different factors as you suggested. First, the huge difference in our emission estimates can be due to certain meteorological condition. The raw wind, in that case, may not represent the wind that carries pollutant downwind region. We can tell that wind speeds get lower when it is near the surface. In that case, the flux in the outflow region can be small because of low wind speed. Note that when we average the entire loop, we already only consider the data below the planetary boundary layer height (PBLH). And when we average the wind over the entire loop below the PBLH, the wind coming into the cylinder (inflow) is negative and the wind out of the cylinder (outflow) is positive.

Second, the difference in our emission estimates can be due to the size of scales and distance to the source regions (e.g. urban or local scale). As seen in Table 2, the difference in emission estimate using different methods becomes smaller at the local scale.

The difference in our emission estimates can vary among flights too with those reasons above. However, the key point that we want to deliver here is that using mass balance wind along with closed-shaped flight can reduce the uncertainty of choice of wind and background, further to the emission estimate.

331: As visible from Fig. S5 only your lowest ellipse lies within the PBL. Clearly the wind is quite different within the PBL from above. Does whole-column-averaged wind (as in Fig. 6c) make sense in this case?

We thought we mentioned it clearly in the manuscript, but it seems that it wasn't clearly stated. The whole-column-averaged wind here means vertically averaged wind up to the estimated "PBLH". We wanted to show all the measured data (from the lowest flight level up to roughly 1000 meter for urban scale and 2000 meter for local scale vertically), but we used data up to the estimated PBLH for actual flux calculation. We have revised Figures 6 and 7 to reduce the confusion.

I. 339: "Flux estimates using raw wind are more sensitive to the choice of background..." Do you have any idea why it is this way?

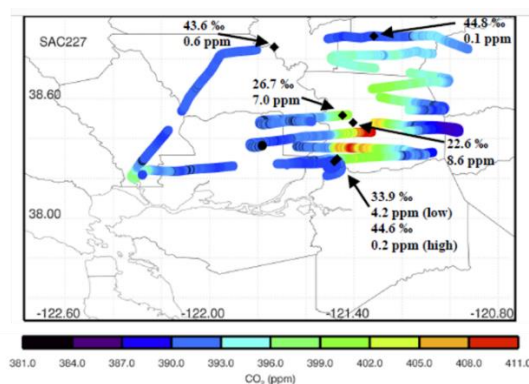
One of the advantages of applying the mass balance approach with an oval (enclosed shape) flight path is that an assignment of the background concentration is not required. Because we calculated the mass of the GHG that entered and exited the cylinder, we are subtracting all GHG upwind from all GHG downwind. So, the background should cancel out. This is also why we used the mass-balanced mean wind, so that influx mass and outflux mass are the same and the total flux estimate is not dependent on having an understanding of background mixing ratios.

However, we do need to know the background value for estimating flux when adopting the curtain pattern of flight. Therefore, we stated that flux estimate using raw wind is sensitive to the choice of the background value (when we do not use mass-balanced wind for cylindrical flights). Sometimes, actually in most cases, the background is not homogeneous, so there can be large uncertainty associated with calculating the background when the non-mass balanced, raw wind is used. There are also challenges in isolating the plume. In a mass-balanced wind approach, such problems do not exist.

I. 388: Table 3 states 13.4 Mt CO₂ for Turnbull.

Thank you for pointing this out. That was a typo, and we corrected it in the revised manuscript.

I. 395ff: Which areas do the Turnbull et al. (2011), Vulcan, and CEPAM inventory values given here cover?



The figure above shows the area where Turnbull et al. (2011) measured. The two bottom-up inventory estimates Vulcan and CARB CEPAM database of the annual total emissions are obtained from Sacramento County.

I. 396: Please give a reference for the 1.1% annual increase in CO₂ fluxes.

Turnbull et al. (2011) use this rate for their study, so we used that to make a consistent comparison with their results. We have included the citation in the revised manuscript.

I. 401: Are there any CH₄ emission inventories you could compare your emission estimates with? How about EDGAR or the U.S. Environmental Protection Agency's inventory?

As you suggested, we looked at the 0.1° x 0.1° gridded 2012 national U.S. Environmental Protection Agency (EPA) CH₄ emission data reported by Maasackers et al. (2016). The November CH₄ emission for our study region around Sacramento is about 89.7 (58.8 – 120.5) Gg yr⁻¹ by increasing from 2012 estimate to 2013 by applying a change of 0.6% per year for CH₄. We added it in Table 3.

		CO2 (Mt yr ⁻¹)	CH4 (Gg yr ⁻¹)
Whole cylinder-AJAX	(bg = min, 100m layer avg)	25.6 ± 2.6	87.1 ± 8.7
	(bg = avg, 100m layer avg)	25.6 ± 2.6	87.4 ± 8.7
Curtain -AJAX	(bg = min)	17.3 ± 1.7	64.4 ± 6.4
	(bg = avg)	8.9 ± 0.9	24.1 ± 2.4
Turnbull et al. (2011)		13.4 (with uncertainty of ~ 100%)	
Vulcan estimates for Sacramento		11.5	
CEPAM estimate for Sacramento		10.0	
Maasakkers et al. (2016) Gridded EPA data (November)			89.3 (58.5 – 120.0)

II. 467-479: Here you emphasize how the ellipses flight pattern reduces the sensitivity towards the choice of background. But, using a curtain flight the choice of background only induces 50 % difference, while for an ellipses flight the difference between wind-treatment techniques is 80%. How does this relate? How sensitive are curtain flights to wind treatment? Please add in Table 3. Why would you still recommend ellipses flights?

For curtain flights, we need to be more careful about wind treatment and background choice. As seen in Table 3 (we added in the revised manuscript), the calculated flux gets more variable depending on the choice of background. The wind treatment will also add challenge to flux calculation. The closed-shape flight using mass-balanced wind will prevent concern.

I. 493: “Second, the seasonality ...” This sentence does not make sense.

We changed it into “Second, the variability of emission estimates with season needs to be examined. “

Figures:

Figure 3: What does the gray dashed line in (c)-(f) depict?

They depict the rough lines of the lowest flight altitude. We added it in the revised manuscript.

Figure 6: It is hard to believe that between the case (a) and (b) the difference is 86 %, but between case (b) and (c) only 3.9 %. Case (c) looks as if the flux estimate should be much larger than (b). Do you have an explanation?

We apologize for the confusion by Figures 6 & 7. In the plots, we wanted to show all the data we measured during the flight, even above the boundary layer height (typically less than 1km above the surface). However, we used the data only up to the top of the estimated boundary layer height when we calculated the final fluxes shown in the Tables. In the old version, we had plotted the fluxes calculated using all the data, which is different than how we calculated the fluxes in the Tables. This was the source of your question about Figs. 6(a, b, c) (as well as Figs. 7(d, e, f)) looking different from their percentages. We updated Fig. 6 and Fig. 7 in the revised manuscript by showing the plots only up to the top of the

estimated PBLH to prevent this confusion. This has the added benefit of allowing a visual comparison of the figures to the tables.

Figure 7: Again it is hard to understand from the figures how (d) and (e) differ by 25 % but (e) and (f) only 2.8 %. The flux in (d) and (e) look almost the same, while (f) looks much different.

Please see above.

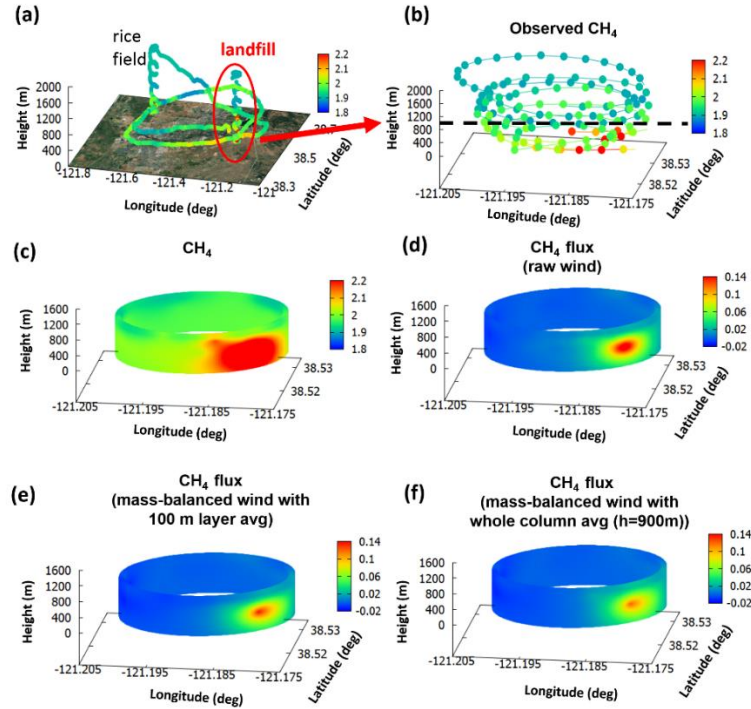


Figure: (a) The map of AJAX flight track with the observed CH₄ mixing ratio. (b) The observed CH₄ mixing ratio, (c) kriged CH₄ mixing ratio, (d) CH₄ flux using raw wind, (e) CH₄ flux using the mass-balanced wind with 100 m vertically averaged wind, and (f) CH₄ flux using mass-balanced wind (averaged through the entire PBLH) over the landfill location on July 29, 2015. The fluxes are computed based on equation (2). The background value was chosen as the minimum value at each vertical layer. The approximate diameter of the cylinder is 3 km, and the color scale is capped at 2.2 ppmv in panels (b) and (c). The black dashed line represents the top of the PBLH for this flight. Figs. (c-f) are only shown below the PBLH.

Tables:

Table 1 and 2: Please indicate negative differences from the base case with a minus in front of the given percentage.

Good points. We added negative differences from the base case with a minus in the revised manuscript.

Table 3: Please add a row of results for curtain flights using raw wind.

As suggested, we also computed the flux estimate over the downwind part (assuming this is for curtain flight) using raw wind. However, note that the fluxes are significantly small ($9.1 \pm 0.9 \text{ Mt yr}^{-1}$ for CO_2 and $37.2 \pm 3.7 \text{ Gg yr}^{-1}$ for CH_4) compared to those using the mass-balanced wind.

In the traditional mass balance analysis, the incoming mass should be the same as the outgoing mass passing the X-Y plane of the measurements. When we used the raw wind measurement, however, we found that the mass of air mass was not conserved, which means we may not fully apply the mass balance approach. The raw wind, in this case, may not represent the wind that carries pollutant downwind region. That is why we used the mass-balanced mean wind at each grid level, and the mean wind will distribute the greenhouse gas inside the cylinder. We assume that the well-mixed condition applies.

Therefore, we don't think using the raw wind at the measurement point is appropriate at this point especially for the urban case so that we don't think we ought to include that in the comparison table.

Reference

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- Stocker, T., Qin, D., Plattner, G. K., Tignor, M. M. B., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P. M.: Climate change 2013 : the physical science basis : Working Group I contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. New York, ISBN 978-1-10741-532-4. OCLC 881236891, Cambridge University Press, 2014.*

Quantification of CO₂ and CH₄ emissions over Sacramento, California based on divergence theorem using aircraft measurements

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In revision for

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Atmospheric Measurement Techniques

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Abstract

45 Emission estimates of carbon dioxide (CO₂) and methane (CH₄) and the meteorological factors affecting them are investigated over Sacramento, California, using an aircraft equipped with a cavity ring-down greenhouse gas sensor as part of the Alpha Jet Atmospheric eXperiment (AJAX) project. To better constrain the emissions fluxes, we designed flights in a cylindrical pattern and computed the emission fluxes from two flights using a kriging method and Gauss's divergence theorem.

50 Differences in wind treatment and assumptions about background concentrations affect the emissions estimates by a factor of 1.5 to 7. The uncertainty is also impacted by meteorological conditions and distance from the emissions sources. ~~The largest CH₄ mixing ratio was found over a local landfill.~~ The vertical layer averaging affects the flux estimate, but the choice of raw wind or mass-balanced wind is more important than the thickness of the vertical averaging for mass-balanced wind for both urban- and local-scale.

55 The importance of vertical mass transfer for flux estimates is examined, and the difference in the total emission estimate with and without vertical mass transfer is found to be small, especially at the local scale. The total flux estimates accounting for the entire circumference are larger than those based solely on measurements made in the downwind region. This indicates that a closed-shape flight profile can better contain total emissions relative to a one-sided curtain flight because most cities have more than one point source and wind direction can change with time and altitude. To reduce the uncertainty of the emissions estimate, it is important that the sampling ~~and modeling~~ strategy account not only for known source locations but also possible unidentified sources around the city. Our
60 results highlight that aircraft-based measurements using a closed-shape flight pattern are an efficient and useful strategy for identifying emission sources and estimating local and city-scale greenhouse gas emission fluxes.

1. Introduction

65 The ability to obtain accurate emission estimates of greenhouse gases (GHG) has been highlighted as an important issue for many decades, not only for regulating local air quality but also for assessing national-scale air quality and climate concerns. In particular, urban emissions need to be well-understood because approximately 70 % of anthropogenic greenhouse gas emissions originate from urban areas (International Energy Agency, 2008; Gurney et al., 2009, 2015). This often causes *urban domes* with higher GHG mixing ratios than surrounding areas (Oke, 70 1982; Idso et al., 1998, 2002; Koerner and Klopatek, 2002; Grimmond et al., 2004; Pataki et al., 2007; Andrews, 2008; Kennedy et al., 2009; Strong et al., 2011). Therefore, estimating greenhouse gas emissions at a regional scale requires an improved understanding of urban GHG emissions (Rosenzweig et al., 2010; Wofsy et al., 2010a, b).

The commonly used *bottom-up* inventories derive estimates of direct and indirect emissions of greenhouse gases based on an understanding of emission factors from the constituent sectors (Andres et al., 1999; Marland et al., 75 1985; Boden et al., 2010; California Air Resources Board, 2015; US EPA, 2016). These estimates rely on monthly or quarterly statistical averages of emission activities and often time-invariant emission factors, which mask behavioral patterns. Recent *bottom-up* inventory data have improved from coarse estimates by using proxy data to produce fine spatial resolution estimates using specific activity data and emission factors corresponding to each emission source. In contrast, *top-down* methods (or inverse modeling), in which observed mixing ratios are 80 partitioned into their sources, have also been used for constraining or cross-checking bottom-up emissions (Huo et al., 2009; Zhang et al., 2009; Cohen and Wang, 2014; Fischer et al., 2016; Miller and Michalak, 2017).

Efforts to understand urban-scale emissions using direct observation have been undertaken in several large urban areas including the Northeastern U.S. (Boston, Baltimore/Washington D.C., He et al., 2013; Dickerson et al., 2016), the U.S. Mountain West (Salt Lake City, Strong et al., 2011), Indianapolis (Mays et al. 2009; Turnbull et al., 85 2015; Lamb et al., 2016; Lauvaux et al., 2016), the southwestern U.S., especially the Los Angeles basin (Duren et al., 2011; Kort et al., 2012) and European cities (Peylin et al., 2005; Kountouris et al., 2018). There are several methods to quantify emissions: in-situ measurements and flask collection through surface tower systems, space-based satellite retrievals, airborne in-situ measurements, mesoscale models, and Large Eddy Simulation (LES) modeling. As part of the Indianapolis Flux Experiment (INFLUX) project, airborne and tower measurements have been 90 collected throughout Indianapolis to generate an extensive database. Over the western U.S., a legacy network over Salt Lake City has collected measurements of CO₂ using surface tower systems for more than a decade (Pataki et al., 2005, 2007; Strong et al., 2011). Results from this extensive dataset have included seasonal variability over years and source apportionment into anthropogenic and biogenic sources. Since current emission inventories do not consider individual characteristics of each city, they have limitations due to their geographical differences in 95 topography, climatology, different source attributions (such as types of industry and agriculture), as well as differences in the measurement and analysis methods.

One approach for estimating CO₂ and CH₄ fluxes over cities is the use of an aircraft-based mass balance method. Several studies have demonstrated the utility of this approach (Kalthoff et al., 2002; Mays et al., 2009; Turnbull et al., 2011; Karion et al., 2013, 2015; Cambaliza et al. 2014; Gordon et al., 2015; Tadić et al., 2017). Mass

100 balance methods utilize many length scales and patterns. The flights target mostly local scales (< 3 km) and areas
around point sources (Nathan et al., 2015; Conley et al., 2017), but they also characterize urban-scales (e.g. 25 x 10
105 km for Gordon et al. (2015), 4 x 9 km for Tadić et al. (2017)) and larger scales (40 km up to 175 km, especially for a
downwind curtain flight (Mays et al., 2009; Turnbull et al., 2011; Karion et al., 2015)).

The flight patterns can be classified into three different categories: 1) single-height transect flight, 2) single
105 screen ("curtain") flight with multiple transects, and 3) enclosed shapes (box, cylinder) (see Fig. S1 in
Supplementary Material). Commonly, there are assumptions made in these airborne sampling approaches. First, the
single-height transect approach assumes a well-mixed boundary layer. Karion et al. (2013) measured CO₂ and CH₄
along a single-height transect with an assumption of uniform distribution of trace gases with altitude within the PBL
and with time. Turnbull et al. (2011) performed a flux estimate by incorporating detailed meteorological information
110 and transecting an emission plume with an aircraft. These studies also assumed that emissions originate from point
sources such as pipes and smokestacks, and travel downwind so that all pollution is reflected on the downwind
"curtain" with constant wind speed. Second, the single-screen multi-transect method does not assume a uniformly
mixed boundary layer condition but is dependent upon constant wind speed. Without a well-mixed boundary layer
assumption, Cambaliza et al. (2014) measured CH₄ along multiple height transects downwind of the city of
115 Indianapolis (See Fig. S1a in Supplementary Material). However, they assumed that winds at the time of
measurement were the same as at the time of emission (i.e., winds after the methane release were time-invariant).
Third, the enclosed 3-D shape flights do not presuppose any of the assumptions described above. Gordon et al.
(2015) measured various GHG with a stacked box flight pattern, to capture the vertical variation in mixing ratio both
upwind and downwind. Tadić et al. (2017) and Conley et al. (2017) accomplished emission estimates by flying a
120 cylinder pattern around an emission source to measure GHG both upwind and downwind for analysis based on the
divergence theorem. More recently, Baray et al. (2017) used both a screen flight and box flight approach around oil
sands facilities and showed that each flight pattern could be preferred, depending on the types of emissions and
spatial characteristics.

The method of extrapolation to unsampled areas can also be a large source of uncertainty. For example,
125 Gordon et al. (2015) demonstrated the significant impact of extrapolation methods over the unsampled, near-surface
region on the final emission estimate, unlike Cambaliza et al. (2014) who assumed that the city plume is rarely
observed in a transect between the surface and the lowest altitude flight measurement. Assumptions can break down
when wind direction and speed vary with time and three dimensional space (see Fig. S1); incorrect use of wind data
can result in increased uncertainty and reduction of accuracy. Flux estimates also require an estimate of the
130 planetary boundary layer height (PBLH), an important physical parameter. State-of-the-art atmospheric models and
reanalysis products often estimate the PBLH, but substantial differences exist in both models and reanalysis data
(Wang et al. 2014). In addition, entrainment from the free troposphere into the planetary boundary layer (PBL) and
fluxes from the surface have been ignored in most previous studies. Thus, more careful consideration and
understanding of these factors are required for determining emission estimates using any of the three mass balance
135 flight patterns.

The primary goals of this study are: i) to assess the impact of different interpolation and extrapolation methods on the emission estimate, ii) to test the sensitivity of emission estimates to a variety of factors such as wind treatment, background mixing ratios, and different flux estimation methods, and finally iii) to examine the importance of vertical mass transfer on the flux estimates. To address these goals, we present here CO₂ and CH₄ data collected during two research flights over Sacramento (See Fig. 1a, Fig. S2 in Supplementary Material) for urban (25-40 km)

and local scales (< 3 km), and determine emission fluxes using various treatments of wind conditions, background mixing ratios, and vertical mass transfer. The data and methodology are presented in section 2. Calculated CO₂ and CH₄ fluxes for all flights are shown in section 3. The sensitivities of flux estimates to different treatment of the wind, background, and vertical mass transfer are also investigated. The conclusions of this study are presented in section 4.

2. Data and Methods

2.1 Data collection

In situ measurements of CO₂ and CH₄ were performed as part of the Alpha Jet Atmospheric eXperiment (AJAX) project. As can be seen in Fig. 1, flight were generally performed in large, nominally oval circuits around the Sacramento urban area. The shape and size of the circuits depended on air traffic control considerations on an individual flight day, with a goal of circumscribing an area of approximately 25 x 40 km. Level circuits at multiple altitudes at and below the top of the boundary layer were performed. In addition to the regular urban-scale flight pattern, this paper also presents data from a flight designed to study two local scale features enclosed by the small circles in the cyan flight track shown in Fig. 1a. Sampling occurred 21:10 – 22:00 UTC on November 18, 2013 (local standard time is UTC minus 8 h, 13:10 – 14:00 PST) and 20:55 – 21:45 UTC on July 29, 2015.

The CO₂ and CH₄ instrument (Picarro Inc., model 2301-m) is calibrated before each flight using two whole-air standards from the National Oceanic and Atmospheric Administration's Earth System Research Laboratory (NOAA/ESRL; CO₂ = 416.267 and 393.319 ppmv; CH₄ = 1.98569 and 1.84362 ppmv). In addition, a set of secondary, synthetic standards was used to verify the linearity of the instrument across a wider range of concentrations. Water vapor corrections using Chen et al. (2010) were applied to calculate the dry mixing ratios of CO₂ used during this study. The overall uncertainty was determined to be 0.16 ppm for CO₂ and 2.2 ppb for CH₄ (Tanaka et al., 2016; Tadić et al., 2014). The Meteorological Measurement System (MMS) measures high-resolution pressure, temperature, and 3-D (*u*, *v*, and *w*) winds (Hamill et al., 2016). The CO₂ and CH₄ mixing ratios and horizontal wind speed are plotted in Fig. 2.

2.2 Data gridding

2.2.1 Extrapolation to the surface

Because the lowest flight level was typically between 250 and 380 m above the surface and there were no ground-based measurements along the flight tracks, there is always a gap in measurement data between the surface and the lowest flight altitude. Many studies adopt a well-mixed layer assumption below the lowest flight altitude

170 (Karion et al., 2013), but the unmeasured values can lead to a significant bias and large uncertainties in estimating
GHG mixing ratios and fluxes, depending on interpolation and extrapolation schemes, especially at lower altitudes
where there are no aircraft data available (Gordon et al., 2015). Thus, here we investigated four methods to
extrapolate mixing ratio values to the surface, which are termed 1) constant, 2) exponential, 3) gaussian, and 4)
kriged (see Fig. 3). The constant method assumes an elevated plume with a constant mixing ratio. The constant
175 mixing ratio here is derived from the lowest flight measurement: $X(t, z) = X(t, z_L)$ for $z_0 < z < z_L$, where z_L is the
lowest flight level. The exponential-fit method assumes an exponential increase of $X(t, z)$ from z_L to z_0 . The
gaussian fit method is similar to the exponential fit method, except that the surface-sourced plume dispersion
follows a gaussian distribution function. The detailed calculation method is based on Gordon et al. (2015). The
kriged fit was applied down to the surface level, extended from the sampled area above.

180 Figures 3a and 3b show observed and estimated CO₂ mixing ratios at several locations over Sacramento on
November 18, 2013. These results demonstrate that a large source of uncertainty and difference comes from not only
the interpolation between flight levels but also the extrapolation of the data between the lowest flight level and the
surface. For example, uncertainty in estimated GHG mixing ratios below the lowest flight level (indicated by the
yellow diamond) can be large (up to ~ 20 %). In the worst cases, CO₂ mixing ratios span more than 80 ppm at the
185 surface among the methods (Fig. 3b); CH₄ ranges > 0.15 ppm. Note that the differences between interpolation
schemes where data exists (above ~ 250–380 m) are smaller than the differences between various methods below the
lowest flight data. Without ground-based data, a proper choice of extrapolation schemes requires knowledge or
presumption of the mixing ratio behavior in this region. Gordon et al. (2015) proposed that the case of elevated
sources beneath the lowest flight level is best suited to constant extrapolation of mixing ratio to the surface (blue
190 curve), while a ground-source should be represented with an exponential-fit extrapolation (red).

The various fits rely on different assumptions; the ordinary kriging method (magenta trace in Figs. 3b and 3f)
also requires some assumptions (e.g., constant mean, constant variance, second-order stationarity and isotropy, and
validity of the theoretical model), but the method leverages spatial and statistical properties of the observations to
derive estimates, and seems to be less arbitrary than alternative interpolation/extrapolation methods. We note the
195 similarity between the kriged values and the constant extrapolation method for both CO₂ and CH₄ (not shown). The
gaussian and the exponential extrapolations produce large values below the measurement level, increasing the
uncertainty. However, the values above the lowest measurement level are very similar among the different fit
methods. This indicates how sensitive the final flux estimate can be depending on the given interpolation and
extrapolation method and how much care should be taken when selecting the extrapolation methods when no data is
200 available.

2.2.2 Elliptical fit and measurement interpolation (Kriging method)

Because the aircraft flew in a cylindrical pattern around the city, the flight paths were transformed into a polar
coordinate system. The path was projected to the surface first and fit into an ellipse using the least squares method to
minimize the difference between the measured data and the fitted data. Then, we computed each point using the

205 major and minor axis of the ellipse and parameter t . Each point on the ellipse was represented by a single parameter (t , eccentric anomaly), according to the equations:

$$\begin{aligned} X(t) &= X_0 + a \cos t \cos \phi - b \sin t \sin \phi \\ Y(t) &= Y_0 + a \cos t \sin \phi + b \sin t \cos \phi \end{aligned} \quad (1)$$

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210 where a and b are the radius of the major and minor axes of the ellipse, ϕ is the angle between the X -axis and the major axis of the ellipse, and the parameter t is obtained from Eqn. (1), varying from 0 to 2π . Then, the data was gridded into a two-dimensional plane [t , height].

215 In order to assess the strengths of a kriging approach to quantifying emissions, two interpolation methods were assessed: interpolation using kriging and interpolation using an exponential weighting function (see Fig. S3). The exponential weighting function at a given point (P) was defined as the weighted average of all the other points where the weights decrease exponentially with distance to P . Both approaches captured the general plume pattern (regions with high and low concentrations of CO_2), but the kriging approach did better at capturing individual plume features such as the range and magnitude, while interpolation with the exponential weighting function could not resolve such details. Another benefit of kriging is that it can estimate values at unsampled locations using a weighted average of neighboring samples, thus reproducing the characteristics of the observed values.

220 Interpolation was performed by the ordinary kriging method (Chilés and Delfiner, 2012), modified from the IDL v8.1 kriging tool to fit an elliptical pattern. We chose ordinary kriging because there is no obvious trend in the data we use. Before kriging, we modeled the variograms for all relevant variables. A variogram (or semivariogram) is a function describing the degree to which the data are correlated as a function of the separation distance between observations. The empirical semivariogram of the data was fit using an exponential variogram model, based upon visual inspection of the experimental variograms. Three parameters were used to fit the theoretical variogram, namely the sill (the expected value of the semivariance between two observations as the lag distance goes to infinity), the range (the distance at which the variogram reaches approximately 95% of the sill), and nugget (representative of measurement error and amount of microscale variability in the data). Variogram modeling was first performed to derive parameters required to obtain ordinary kriged estimates. Various other types of kriging exist in the literature on quantifying greenhouse fluxes (Tadić et al., 2017), but examining their differences is beyond the scope of this study.

235 We kriged the CO_2 , CH_4 , wind, temperature, and pressure observations to obtain both the estimate and the uncertainty for each variable at each grid point. The individual semivariograms of the variables for each flight were produced, and we present them for one flight in Fig. S4 in Supplementary Material. For each flight, the sampled data were kriged to a grid of maximum height divided by 150 in the vertical dimension; the horizontal dimension was kriged from end to end of the flight transect, enclosing the circumference of the entire city, divided by 360 in the horizontal direction. The vertical dimension was interpolated from the ground to the top of the flight measurement, but only data up to the estimated planetary boundary layer height (PBLH) were used for computing the flux estimates.

240

3. Flux calculations

Figure 1 shows a map of the AJAX flight tracks on November 18, 2013 and July 29, 2015 over Sacramento and the vertical structure of the CO₂ mixing ratio on November 18, 2013. A simple illustration of the air flow demonstrates the basic idea of this study, Gauss's divergence theorem, which relates the flow through the surface to the volume of the cylinder (Fig. 1c). Mass coming in and out of the cylinder should be conserved if there is no leak through the top or the bottom of the cylinder (i.e., the flow into the cylinder balances with the flow out of the cylinder). More precisely, the surface integral through a closed system is *equal* to the volume integral of the divergence over the region inside the surface. Since the atmosphere has no upper boundary, we assume that vertical mass transfer is accomplished through entrainment from the top of the PBL, and surface flux from the bottom of the cylinder near the surface. In this way, the oval cylinder we design over the city has a closed surface, and the flux inside the cylinder is equal to the sum of the emission flux at the bottom.

In Section 3.1 we will first describe the "base case" calculation of fluxes, and in Section 3.2 we report the sensitivity of the fluxes to variations in several aspects of the method.

3.1 Base case experiment

Our base case experiment used the entire gridded, enclosed elliptical data curtain using kriging as both the interpolation and extrapolation method. We averaged the measured wind in vertical layers 100 m thick so that air (mass) coming into the cylinder equaled air leaving the cylinder (which we refer to as "mass-balanced wind"). We assigned the background to be the minimum concentration found in each 100 m layer. PBLH was determined as the altitude of the maximum gradient from a vertical profile of potential temperature (Wang et al., 2008) obtained from the MMS measurements during each flight. We included entrainment from the top and surface flux from the surface. The results of this base case are displayed in the top rows of Tables 1 (urban scale) and 2 (local scale).

Here we define the entrainment (surface) flux as the turbulent flux of the scalar at the boundary layer height (surface) (Faloona et al., 2005). Then we compute the entrainment flux at the top of the cylinder by multiplying the area of the top of the cylinder with $E = \overline{(w'c')}_{z=h} \cdot A$ where A is the area of the top of the cylinder, $c' = (C(t, z) - C_{bg}(h))$, $C(t, z)$ is the CO₂ (or CH₄) mass concentration (g m⁻³) converted from the CO₂ (or CH₄) mixing ratio (ppmv) at a given point, t , along the perimeter of the top of the cylinder at $z=h$, and $C_{bg}(h)$ is the background concentration of CO₂ (or CH₄) at the top of the boundary layer. The CO₂ (CH₄) mass is calculated from the CO₂ (CH₄) mixing ratio (ppmv). Using this, we could make direct observations of the entrainment flux by measuring vertical velocity together with the trace gas mixing ratio throughout the boundary layer. The surface flux is computed at the surface ($z = 0$) in a similar manner.

We determined kriged data for each field from the measured CO₂, CH₄, wind, temperature, and pressure, and then subtracted background values from the trace gas data at each grid point. To convert the volume mixing ratio [ppmv] to a mass concentration [g m⁻³], the number of CO₂ or CH₄ molecules were computed based on the ideal gas law using the kriged temperature and pressure. Then, the net mass flow [g m⁻² s⁻¹] was integrated in the horizontal and vertical directions from the surface up to the top of the cylinder.

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$$F = \iint \bar{U}(\theta, z) \sin(\alpha) \cdot (C(\theta, z) - C_{bg}) L d\theta dz \quad (2)$$

where L is the difference between two points on the ellipse, $\bar{U}(\theta, z)$ is the wind speed, α is the angle of the wind velocity relative to the flux surface, C is the concentration (g m^{-3}), and C_{bg} is the background concentration at each level z . The component of the wind perpendicular to the flux surface was used in the flux calculation.

Figure 4 shows the measured methane over Sacramento, CA, for November 18, 2013, the projection to the ground, and the computed “flux surface”. The kriged CH_4 not only captures the measured CH_4 mixing ratio but also fills the unsampled area based on the observed data characteristics. Maximum values were found at 38.73° N , 121.2° W to 38.68° N , 121.45° W at 300 m. The high CH_4 region corresponds with highways, landfills and dairy farms.

For the same flight, Fig. 5 shows the observed and kriged CO_2 mixing ratio and kriging uncertainty at each grid point. The kriged CO_2 field captures the main features of the observed CO_2 plume well. The CO_2 mixing ratios were much larger (up to 25 ppm higher at most spots) on the downwind side than the upwind side. The observations in Figs. 5(b–e) suggests that the vast majority of the emission sampled by the flights originates in the region identified as traffic regions (Roseville), airports, metropolitan areas (Arden-Arcade, Roseville, North Highlands, Fair Oaks) (see the map in Fig. 5a). Uncertainties of CO_2 were large near the surface, small from 200–900 m, and grew larger near the top of the sampled domain.

The vertical stretching pattern of CO_2 mixing ratios in Fig. 5(d) appears to be due to the large scale difference between the horizontal length ($> 120 \text{ km}$) and the vertical length ($< 1 \text{ km}$). When we applied our method to the local scale (horizontal scale $< 3 \text{ km}$, see Fig. 7), or took a small horizontal portion of the large oval (see Fig. S3 in Supplementary Material), the vertical stretching pattern disappeared.

As shown in the top row of Table 1, we determined urban-scale flux values of $25.6 \pm 2.6 \text{ Mt CO}_2 \text{ yr}^{-1}$ and $87.1 \pm 8.7 \text{ Gg CH}_4 \text{ yr}^{-1}$ for this base case experiment. Note that we do not consider the uptake of CO_2 by vegetation, but the biological impact on CO_2 flux will be important especially during summer.

3.2 Sensitivity Tests

3.2.1 Sensitivity of calculated flux to interpolation/extrapolation method

As shown in Fig. 3b, there is a large source of uncertainty and difference due to extrapolation of the data between the lowest flight level and the surface. For example, CO_2 mixing ratios span more than $\sim 80 \text{ ppm}$ near the surface among the methods ($\sim 20\%$); CH_4 ranges $> 0.15 \text{ ppm}$ ($\sim 8\%$). This implies that a proper choice of extrapolation scheme requires knowledge or presumption of the mixing ratio behavior in this region when there is no ground-based data. Gordon et al. (2015) proposed that the case of elevated sources beneath the lowest flight level is best suited to constant extrapolation of mixing ratio to the surface (cyan curve), while a ground-source should be represented with an exponential-fit extrapolation (red). Note that the differences of the mixing ratio between interpolation schemes where data exists (above $\sim 250\text{--}380 \text{ m}$) are smaller than the differences of the mixing ratio between various methods below the lowest flight data for both CO_2 and CH_4 .

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While it is important to make the best possible choice among different GHG mixing ratio extrapolations, similar uncertainty in how best to treat the wind below the lowest flight level makes calculations propagating the effect of each extrapolation scheme challenging, and exploration of the isolated impact of the extrapolation of GHG mixing ratios is of limited value. In general terms, and making reasonable assumptions for the estimation of wind, boundary layer height, and background, we expect the sensitivity of calculated flux to the interpolation method is small above the lowest flight level, but the sensitivity of calculated flux to the extrapolation method below the lowest flight level will be more than 20% depending on the usage of wind, leading to difference in the final flux estimate.

3.2.2¹ Sensitivity of calculated flux to wind treatment

Wind variability and measurement assumptions can lead to errors in the CO₂ and CH₄ flux estimates (Mays et al., 2009; Cambaliza et al., 2014, 2015; Nathan et al., 2015; Karion et al., 2013, 2015), and the way in which winds are estimated and quantified especially matters. To test the sensitivity of fluxes to the treatment of wind, we applied the measured high-resolution (1 Hz) in-situ wind data to the flux calculation in two different ways. We averaged horizontal wind on each vertical level (100 m for the base case, 500 m (not shown), or the whole cylinder as one layer), so that air (mass) coming into the cylinder equaled air leaving the cylinder. We also evaluated the calculated fluxes when the measured wind was used without any averaging (hereafter we refer to it as “raw wind”). In this case, inflow and outflow are not required to be balanced.

For November 18, 2013, the wind was southwesterly at the low altitudes, but it changed its direction to southeasterly as height increased. Figure 6 demonstrates the clear difference in flux estimates when the 2-D raw wind or the mass-balanced wind is used. The right column in Fig. 6 shows that we captured high fluxes when we used the mass-balanced wind (middle and bottom rows), while we were less likely to obtain a strong emission signal when using the raw wind data (top), which might be attributed to an imbalance of inflow and outflow to the cylinder. The total flux was ~7 times different between wind cases: 3.7 Mt CO₂ yr⁻¹ and 13.0 Gg CH₄ yr⁻¹ calculated with raw wind, and 25.6 Mt CO₂ yr⁻¹ and 87.1 Gg CH₄ yr⁻¹ using mass-balanced wind with 100 m vertical average, leading to 86% and 85% difference compared to the base case (see Table 1).

The importance of wind data on the flux calculation is also seen in local-scale emission calculations, but not as dramatically as in those for the urban scale (see Table 2). For the small cylinder over the landfill site on July 29, 2015, Figure 7 shows the observed and kriged CH₄ mixing ratio and the flux estimation using the raw wind and mass-balanced wind over the landfill site. As before, the kriged CH₄ is a good representation of the local characteristics of the CH₄ field. Reassuringly, the elevated CH₄ concentration was reconstructed over 121.19° W, 38.52° N, which was close to the nearby landfill (See also Fig. 4, Fig. S6 in Supplementary Material). Considering light wind conditions (< 4 m s⁻¹) and high temperature during July, the high flux estimates are attributed to the local emissions. For local-scale, the difference in the flux estimate using the raw wind and mass-balanced wind is relatively small. For example, even when using raw-wind over the landfill, the difference of the calculated flux from

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base case is ~25 % for CH₄, which is about 1/3 smaller than the difference of calculated flux from the base case for urban-scale (~ 85%) for CH₄. For CO₂, when using raw-wind the difference of calculated flux from base case gets larger, but it is still smaller than the difference for urban-scale (See Table 1).

350 Another interesting finding here is the importance of the vertical averaging effect of wind, which is also shown in Figs. 6 and 7. Even when using the mass-balanced wind, the whole-column-averaged wind can underestimate or overestimate the final flux estimate depending on the situation. Certainly, care needs to be paid when treating wind as a mean in both the horizontal and the vertical. Many previous studies estimated CO₂ and CH₄ fluxes based on the mean wind vector at the dominant wind direction (positive and one direction) and speed (Turnbull et al. 2011; 355 Karion et al. 2015), often using simulated wind obtained from a coarse resolution model. Even when using high resolution measured winds in place of coarse resolution model data, we can see the impact of averaging wind on the flux estimate. However, overall, the choice of raw wind or mass-balanced wind is more important than the thickness of the vertical averaging for mass-balanced wind for flux estimate for both urban- and local-scale. Furthermore, the flux estimates using raw wind are more sensitive to the choice of the background for both urban- and local scale. For 360 example, when we use raw wind with average background concentration, the flux estimate is about the same as the base case flux estimate (See bottom rows in Table 1 and Table 2).

3.2.32 Sensitivity of calculated flux to the choice of background concentrations

Background values are one of the most important factors in obtaining flux estimates, and theoretically, the background values should be cancelled out for the enclosed-shape mass-balance flight. Here we used several distinct 365 methods to determine background values and calculate emission fluxes for each gas to assess if our method could remove some of the uncertainty due to assigning the background. As in the base case, we used the *minimum* concentration over the layer height (e.g., 100 m or whole column averaging). In comparison, we also calculated fluxes using the *average* concentration in each layer as the background. Third, we also tested two different, vertically invariant, constant values.

370 Tables 1 and 2 show the calculated CO₂ and CH₄ emission fluxes using two different wind methods and two different background treatments. The rows labeled "min" were generated using the minimum kriged mixing ratio in each altitude band as the background for all data at that level. The rows identified by "avg" used the average mixing ratio in each altitude band as the background on that level.

375 The bottom two rows of Tables 1 and 2 show the sensitivity of calculated flux to the choice of the background treatment was significant when we used raw wind; the estimate using average concentration for the background closely matched the base case, but using the minimum concentration for the background resulted in significantly different calculated fluxes, as we mentioned earlier. This was true both with the vertical mass transfer (Table 1) and without (not shown). In contrast, when we use the mass-balanced wind, the emission estimates for both CO₂ and CH₄ are nearly identical for either choice of background treatment. Interestingly, when an average mixing ratio at a 380 given vertical level is used for the background concentration, emission estimates with raw wind are similar to emission estimates with mass-balanced wind. To satisfy mass conservation, we also computed the entrainment flux

from the top ($z=h$) and the surface flux from the bottom of the cylinder ($z=0$). The data from Table 1 is also shown in Fig. 8 as the non-hatched bars.

3.2.43 Sensitivity of calculated flux to vertical mass transfer

Many previous studies assume that vertical mass transfer can be neglected (Cambaliza et al., 2014; Conley et al., 2017). To quantify the validity of this assumption, we compare in Fig. 8 the flux determined when including or neglecting the entrainment and surface fluxes. Multiple implementations were tested, as shown in different colors. The differences between CO₂ and CH₄ fluxes calculated with (left side of each panel) and without (right side, hatched bars) vertical mass transfer were determined to be about 11 % for CO₂ and 21 % for CH₄ on the urban scale and less important for the local emission estimates (< 8 %, not shown).

3.2.54 Sensitivity of calculated flux to the PBLH estimate

We also considered the sensitivity of the calculated flux to the PBLH. The potential temperature profile, which indicates atmospheric static stability and which significantly affects pollutant diffusion, is one of the most common operational methods to determine PBLH. No significant sensitivity was found using several different PBLH detection algorithms, such as the parcel method (the interaction between dry adiabatic lapse rate and temperature), rapid decrease in water vapor (Wang and Wang, 2014), or Richardson number method (Wang et al. 2008). A simple example is shown in Supplementary Material Fig. S5. When we determined the PBLH based on the largest gradient of the vertical profile of the potential temperature, the uncertainty due to PBLH estimate for [the](#) urban scale is about ~10 %, and that for the local-scale is about 1-5 %, thus the change of PBLH does not affect the total flux estimate, especially for the local-scale. As seen in Fig. S5, the vertical range of the largest gradient of potential temperature is very small for the local-scale, compared to the urban-scale. This leads us to another important message: the uncertainty can increase when we consider urban-scale flux estimates.

3.2.65 Sensitivity of calculated flux to the closed shape

Our city-wide estimate of about 25.6 ± 2.6 Mt CO₂ yr⁻¹ (e.g., using the base case of mass-balanced wind with minimum background concentration) is higher than the result by Turnbull et al. (2011), who reported 13.46 Mt CO₂ yr⁻¹ (3.5 MtC yr⁻¹) over Sacramento in February 2009 (See Table 3). When we examine only the small downwind portion of the ellipse which shows the highest CO₂ mixing ratio (e.g. 121.45–121.20° W and 38.65–38.76° N in 2013, See Figs. 5(b, d)), CO₂ fluxes calculated using mass-balanced wind with *minimum* concentration for the background were about 17.3 ± 1.7 Mt yr⁻¹ in 2013. When calculating fluxes using mass-balanced wind with *average* concentration for background, the "downwind side" emission estimates were 8.9 ± 0.9 Mt CO₂ yr⁻¹. According to these calculations, the fluxes from the downwind portion of the cylinder were responsible for only ~35–68 % of the total emissions. The Turnbull et al. (2011) data were collected in 2009; the value given here was converted from the mean reported value of 3.5 Mt C yr⁻¹ with a 1.1% yr⁻¹ increase in CO₂ flux to adjust to 2013 ([Turnbull et al., 2011](#)). Bottom-up inventory estimates of the annual total emissions from Sacramento County from Vulcan (Gurney et al., 2009) and the California Air Resources Board CEPAM database (Turnbull et al., 2011) are

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420 included for comparison for CO₂ in Table 3. The Vulcan inventory is available only for 2002, and the CEPAM database is available for 2004. We applied a 1.1% yr⁻¹ increase in CO₂ flux to adjust to 2013. The Vulcan inventory is available only for 2002, and the CEPAM database is available for 2004. We applied a 1.1% yr⁻¹ increase in CO₂ flux to adjust to 2013.

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425 Our city-wide estimate of 87.1 Gg CH₄ yr⁻¹ (e.g., flux estimate using 100 m vertically averaged mass-balanced wind) on November 18, 2013 corresponds to 52% of the 167 Gg CH₄ yr⁻¹ (~ 140–220 Gg yr⁻¹) reported by Jeong et al. (2016) over Region-3 (Sacramento Valley) for 2013-2014. The value of 89.7 Gg yr⁻¹ we estimated from the Gridded (0.1° x 0.1°) 2012 National U.S. Environmental Protection Agency (EPA) Inventory, increased by about 0.6% yr⁻¹ (Stocker et al., 2014) to adjust November 2012 to 2013 (Maasakkers et al., 2016), is in excellent agreement with our whole-cylinder observations. Slight adjustments to the grid boxes we choose from Maasakkers et al. (2016) give a range of 58.8–120.5 Gg yr⁻¹.

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430 Our city-wide estimate of 87.1 Gg CH₄ yr⁻¹ (e.g., flux estimate using mass-balanced wind, 100 m vertically averaged wind) on November 18, 2013 corresponds to 52% of the 167 Gg CH₄ yr⁻¹ (~ 140–220 Gg yr⁻¹) reported by Jeong et al. (2016) over Region 3 (San Joaquin Valley area including Sacramento). Direct comparison between different flux estimates is challenging due to various factors, such as i) differences in the areas covered, ii) differences between bottom-up inventory and top-down estimates, iii) the variance of measurement methods (tower, aircraft, and model), iv) underestimation of the emissions from known sources, v) seasonal and interannual variability, and vi) lack of understanding of unidentified sources. Consideration of these factors will be one of the most important areas for improvement for establishing better emission estimate databases in the future.

3.3 Flux uncertainties

440 Figure 5e shows the uncertainties in the individual kriged CO₂ values are large near the surface (i.e., below ~ 200 m), small from 200–900 m, and grow larger near the top of the sampled domain. Because there were no measurements on the ground, the estimates below 200 m were dependent only on the data around 200 m, leading to the largest uncertainty near the ground. In addition, uncertainties are increased where the data were observed far from the elliptical path.

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445 Here we consider the uncertainty of the kriged CO₂ and CH₄ mixing ratios as the sources of uncertainty in overall flux estimates. We also consider uncertainty from the wind measurement (Mays et al., 2009; Karion et al., 2015; Tadić et al., 2017), estimation of the PBLH, and vertical fluxes. We did not consider the uncertainty of the grid resolution, measurement error, and the selection of the variogram model (such as gaussian-cosine, linear, exponential, and exponential-bessel variogram), because these all have been shown to be small, (< 4%, e.g., Nathan et al. (2015)). We also did not consider the uncertainty of the background in isolation, because this is somewhat coupled to the choice of wind treatment in this study, and the uncertainty is small when we chose the mass-balanced wind. However, in general, the uncertainty of the background can contribute significantly to the overall flux uncertainty, especially for the curtain-shape mass-balance flight. The uncertainty can be also impacted by meteorological conditions and distance from the emission sources, which are not considered in this study.

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455 The uncertainty in the kriged results was assessed using the variance (and the standard deviation) of the kriged
estimate. By assuming that the errors of each factor are gaussian in nature, each measurement (e.g., CO₂ and wind)
is independent; we estimate the total uncertainties in the calculated flux by adding the fractional uncertainties of the
individual kriged CO₂ (or CH₄), and winds in quadrature (Nathan et al., 2015). We also added the fractional
460 uncertainty of the PBLH estimates and vertical fluxes in quadrature to the uncertainty of the flux. For urban scales,
the fractional uncertainty of fluxes due to kriged interpolation was 4%, PBLH estimation was 10%, and vertical
fluxes <1%. For local scales, the fractional uncertainty of fluxes due to kriged interpolation was (35%, 17%), PBLH
estimation (1%, <5%), and vertical fluxes (<1%, <1%) over the landfill and rice field. By including all these factors,
the overall uncertainty of the emission flux estimate over the urban scales is about 11% for both CO₂ and CH₄. The
465 overall uncertainties over the local scales for both CO₂ and CH₄ on were about 35% at the landfill site and 18% at
the rice field location.

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The uncertainty in the kriged results was assessed using the variance (and the standard deviation) of the kriged estimate at each point, as in Mays et al. (2009) and Nathan et al. (2015). In a statistical sense, the interpolated CO₂ and CH₄ concentration are one of the largest sources of uncertainty in flux error estimates because the flux calculation requires interpolated values at unsampled locations. Another well known significant source of uncertainty comes from wind measurement (Mays et al., 2009; Karion et al., 2015; Tadić et al., 2017). The grid resolution can also be a source of uncertainty. Nathan et al. (2015) reported that changing the grid resolution by a factor of 2 in either direction resulted in a 4 % absolute change in the emission rate, and showed that the grid size does not significantly bias the interpolated emission rates for their study. However, emission estimates may depend on scales of variability in the measured quantities and the grid resolution, in that the grid resolution has to be sufficiently fine to capture the observed scales of variability. They also demonstrated that the selection of the variogram model they used, such as gaussian cosine, linear, exponential, and exponential-bessel variogram, did not affect the final emission estimate substantially (the difference is less than 5 %) in their case study. Moreover, uncertainties in greenhouse gas mixing ratio measurements, as well as in wind speed and direction observations, directly propagate the emission rates uncertainties.

480 Uncertainties in the individual kriged CO₂ values are large near the surface, small from 200–900 m, and grow larger near the top of the sampled domain. Figure 5e shows the uncertainty is largest near the ground, in particular over the unmeasured area (e.g., November 18, 2013, below ~ 200 m), and when the data were observed far from the elliptical path. Furthermore, there were no measurements on the ground, so the estimates below 200 m were dependent only on the data around 200 m, which was an additional source of the uncertainty.

485 By assuming that the errors of each factor are gaussian in nature, each measurement (e.g., CO₂ and wind) is independent, we estimate the total uncertainties in the calculated flux by adding the fractional uncertainties of the individual kriged CO₂, CH₄, and winds in quadrature (Nathan et al., 2015). We also added the fractional uncertainty of the PBLH estimates in quadrature to the uncertainty of the flux; they are about 10% for the urban scale and 1–5% for the local scales, so that the change of PBLH does not affect the total flux estimate, especially for the local scale. Furthermore, when we include the entrainment flux at the top and the surface flux at the bottom of the cylinder in

495 addition to the flux in the lateral part, the total uncertainty was increased by about <1% or remained the same for both CO₂ and CH₄. This appears to be because the contribution of the vertical mass transfer through entrainment and the surface flux to the total flux estimates is relatively small. By including all these factors, the overall uncertainty of the emission flux estimate over the urban scales is about 10% for both CO₂ and CH₄. The overall uncertainties over the local scales over landfill and rice field for both CO₂ and CH₄ are about 35% and 17%.

500 Although we used much more accurate in-situ wind measurements than most past studies for flux calculation, the wind was still the most important variable for the uncertainty of flux estimates, consistent with previous studies. This partially stems from the uncertainty in the wind at interpolated locations or the sparsity of the measurements. Cambaliza et al. (2014) estimated the uncertainty of the emission rates from kriging analysis is about 50%. Nathan et al. (2015) also estimated the overall statistical uncertainty of the emission rate over a compressor station in the Barnett Shale as $\pm 55\%$.

4. Conclusions

505 We have estimated CO₂ and CH₄ fluxes over Sacramento, California, on two days using an airborne in-situ dataset from the Alpha Jet Atmospheric eXperiment (AJAX) project and have tested the sensitivity of emission estimates to a variety of factors. We deployed cylindrical flight patterns of two sizes that differ from common curtain flights to estimate the total flux at urban and local scales. We also applied a kriging interpolation method to the data, capturing the characteristics of the data at both observed and unsampled locations. Then, we tested the sensitivity of flux estimates to the wind treatments (either raw wind or mass-balanced wind) and background concentrations and found these two factors were the dominant factors in determining the total flux uncertainty. 510 When we used the mass-balanced wind for flux calculation, the sensitivity of the emission estimate to the choice of background was minimal (Table 1). Raw wind produced similar flux estimates when the background mixing ratio was set to the average value on each vertical layer. In contrast, choosing the background as the minimum value observed on each level led to calculated fluxes that were substantially different.

515 Additionally, we took into account not only the inflow and the outflow through the cylinder around the city, but also the vertical mass transport (e.g., entrainment and surface flux) and tested the sensitivity of the total flux estimate to the vertical mass transfer for both urban and local scales. The winds observed on November 18, 2013 came from the southeast, showing high concentrations of CO₂ downwind of industrial facilities. CH₄ over a rice field showed lower emission rates than those over the landfill, and this may be due to the relatively high wind, no particular point source, and reduced CH₄ emissions as a result of low humidity. Considering the wind speed was much lower in July (especially over the landfill), this indicates that most of the emission was produced from local sources for the July 29, 2015 case. 520

525 The advantage of the closed shape (i.e., elliptical in this study) approach over a curtain flight is to make a more precise “total” emissions estimate possible by taking into account all unknown sources of emissions. Regarding the balanced incoming and outgoing fluxes within a closed volume, we suggest that emission estimates using mass-balanced wind computed over a closed shape can be beneficial for several reasons. First, the flux estimates calculated using mass-balanced wind show reduced sensitivity to the choice of background. Figure 6 and Tables 1

and 2 show that the background value is one of the major sources of variability in both CO₂ and CH₄ emission estimates when using raw wind, but not when using mass-balanced wind. Vertical averaging of wind also affects the flux estimate, but the choice of raw wind or mass-balanced wind is more important than the thickness of the vertical averaging for mass-balanced wind on both urban and local scales. Second, when we analyze only a small portion of the large loop (e.g., downtown hot spot region) to mimic the curtain flight style, the final flux estimates are highly sensitive to the background choice no matter how the measured wind data are treated. Thus, we propose that the flux estimates for the closed elliptical loops have a reduced sensitivity to the choice of background values in comparison to the curtain geometry.

The spatial variation of CO₂ and CH₄ observed in the cylindrical flight pattern measured over Sacramento reveals that there were several local sources throughout the entire city, not only concentrated on the downwind side. Our sensitivity study reveals that the unbalanced wind varying with time and space may be a source of methodological uncertainty. Thus, use of constant wind speed or unrepresentative coarse resolution of wind (e.g., model output) by focusing only on the downwind side may lead to significant uncertainty in the estimation of the greenhouse gas emission fluxes. The size of the ellipse measuring urban emission appeared to be another factor affecting flux estimates. In general, the vertical mass transfer does not significantly contribute to the total emission estimate (especially at local scales), but it can modify total emission estimates by up to 11% for CO₂ and 21% for CH₄ urban scales in our cases. For the local scale (~ 3 km), the vertical mass transfer was not important due to the small turbulent fluxes. The Planetary Boundary Layer Height (PBLH) was calculated using the vertical profiles of potential temperature and was used together with the vertical fluxes for computing the entrainment from the top and the surface flux from the bottom of the cylinder.

There are still several issues to be addressed in future studies. First, sector-specific emissions and their uncertainties for CO₂ and CH₄ need to be further identified (Miller and Michalak, 2017). Second, the [variability seasonality](#) of emission estimates to various factors [with season](#) needs to be examined. We expect that the biological impact on CO₂ flux by the CO₂ uptake by vegetation will be important especially during summer. Finally, understanding the sources of uncertainties in emission estimates, and how different they can be under various meteorological conditions (such as temperature, atmospheric stability) need to be investigated further. In this sense, the changing climate over California makes it harder to predict future emission patterns. The use of aircraft measurements presented here provides a tremendous opportunity to measure the entire urban plume.

This effort is not limited to one particular city. There has been increasing interest in performing inter-city comparisons to validate datasets in a more efficient and adequate manner, to create a uniform database that is useful for emission controls (Urban greenhouse gas measurements workshop, 2016). Given that data are available over several cities which have different conditions, we can test how to obtain emission estimates from several cities. Differences in the socio-economic, geologic, and industrial characteristics of cities lead to a need to compare emission estimates between them, as together they can contribute significantly to the total GHG emission at national and global scales. Thorough comparison among datasets and a customized sharing system between different research groups will lead to reducing the uncertainty of emission estimates.

565 **Author contribution**

Ju-Mee Ryoo, Laura T. Iraci, Tomoaki Tanaka, Josette E. Marrero, Emma L. Yates, Warren Gore designed the experiments, and they carried them out. T. Paul Bui and Cecilia S. Chang prepared MMS instruments and Jonathan M. Dean-Day processed the data. Anna M. Michalak and Jovan Tadić participated in experiment design and supported the interpretation of the statistical analysis. Inez Fung gave insightful comments, and Laura T. Iraci ~~providesgave helpful guidance~~[substantial guidance and support](#) in formulating the structure [and contents](#) of this study. Ju-Mee Ryoo developed the statistical model code and performed the analysis as well as prepared the manuscript with contributions from all co-authors.

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Competing interests

The authors declare that they have no conflict of interest.

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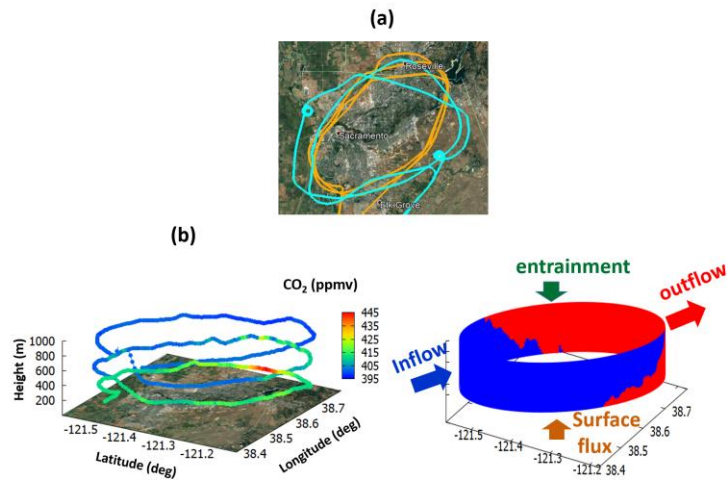


Figure 1: (a) Map of AJAX flight tracks on November 18, 2013 (orange) and July 29, 2015 (cyan) plotted in Google™ Earth. (b) Vertical measurements of CO₂ mixing ratio on November 18, 2013, and (c) simple illustration of airflow [$\text{kg m}^{-2} \text{s}^{-1}$] passing through cylinder (over Sacramento). The color represents the air mass flux (density [kg m^{-3}] multiplied by wind vector [m s^{-1}]) normal to the cylinder. The blue and red represent inflow and outflow, respectively. The vertical mass transfer through the top and bottom are referred to as the entrainment and surface flux, respectively.

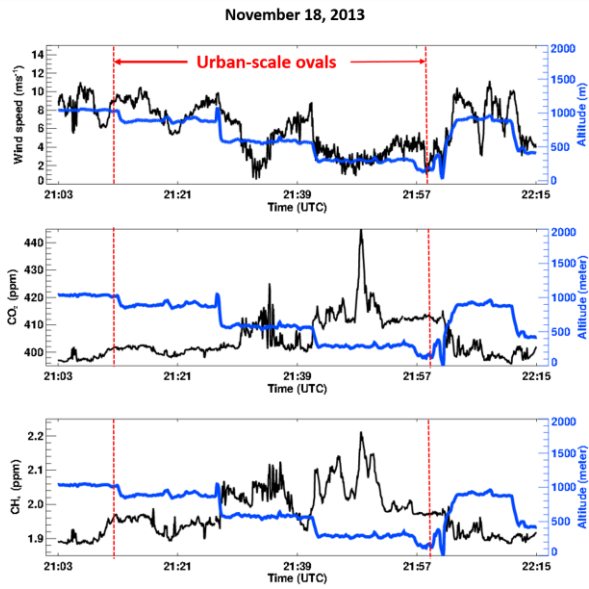
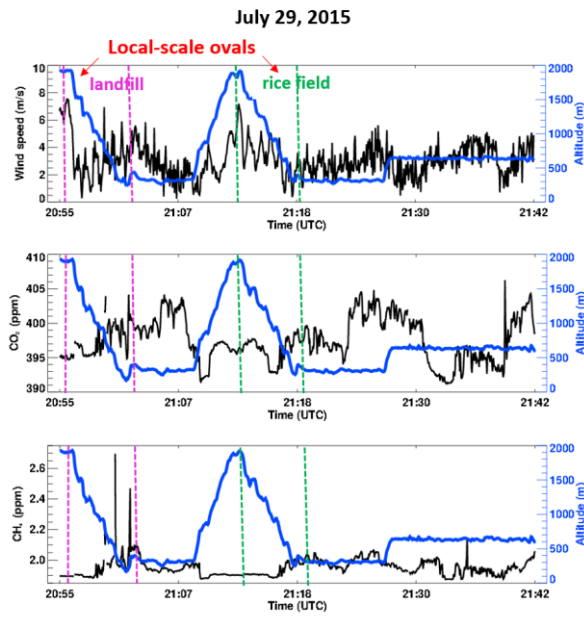


Figure 2a: A time series of (black lines) CO_2 , CH_4 , horizontal wind speed and (blue lines) altitude of the aircraft for November 18, 2103. The red dashed lines represent portion of the flight over Sacramento.

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760 Figure 2b: The same as Fig. 2a except for July 29, 2015. The magenta dashed lines indicate the portion of the flight over the landfill, and the green dashed lines mark the start and end times of the rice field measurements.

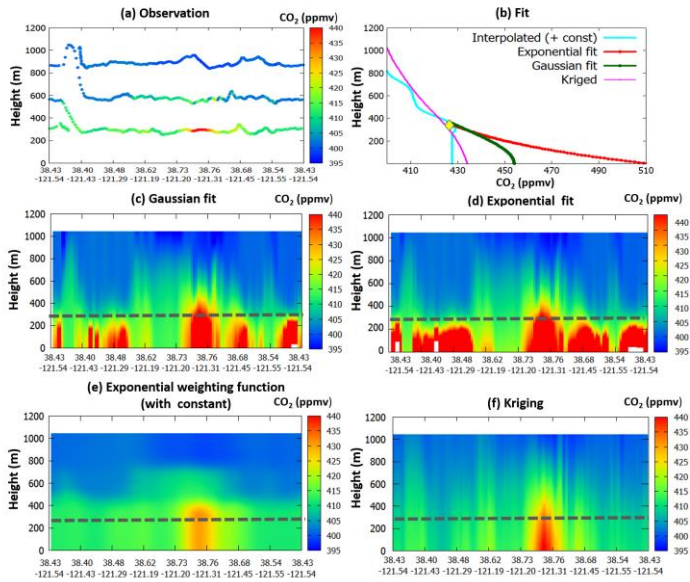


Figure 3: (a) Observed CO₂ over the Sacramento loop on November 18, 2013. (b) The vertical profiles of calculated CO₂ mixing ratios around 38.75° N, 121.27° W. The yellow diamond indicates the altitude of the lowest flight data. The kriged values (magenta), interpolated values with exponential weighting function and extrapolated values using constant (cyan), gaussian fit (green), and exponential fit (red) are compared. The CO₂ mixing ratio obtained from (c) the gaussian fit, (d) exponential fit, (e) exponential weighting function with constant, (f) kriging method. The empirical fits were generated based on the approach by Gordon et al. (2015). In panels (c) and (d), the white boxes result from no fit due to the lack of the data points. [The gray dashed line in \(c\)-\(f\) depicts the rough lines of the lowest flight altitude.](#)

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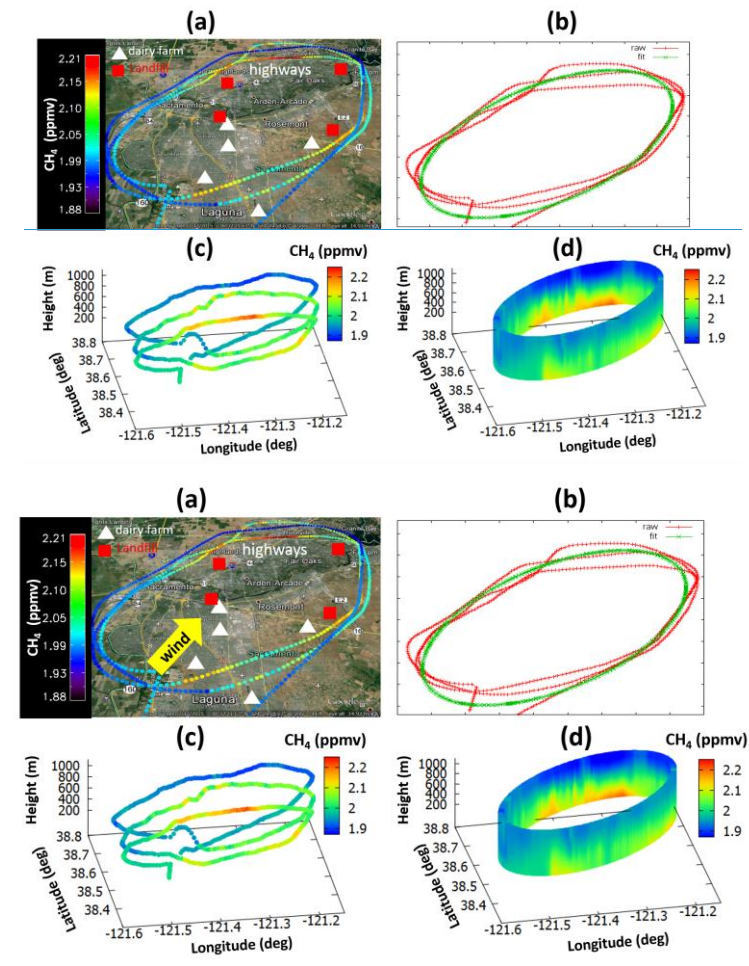


Figure 4: (a) Map of AJAX flight tracks colored by CH₄ mixing ratio for November 18, 2013, plotted in Google™ Earth. (b) The data (red) fitted to an oval (green). The observed CH₄ mixing ratios (c) are kriged to generate the cylindrical surface (d). The axes of the oval are approximately 25 and 40 km. [The yellow arrow represents the dominant wind direction.](#)

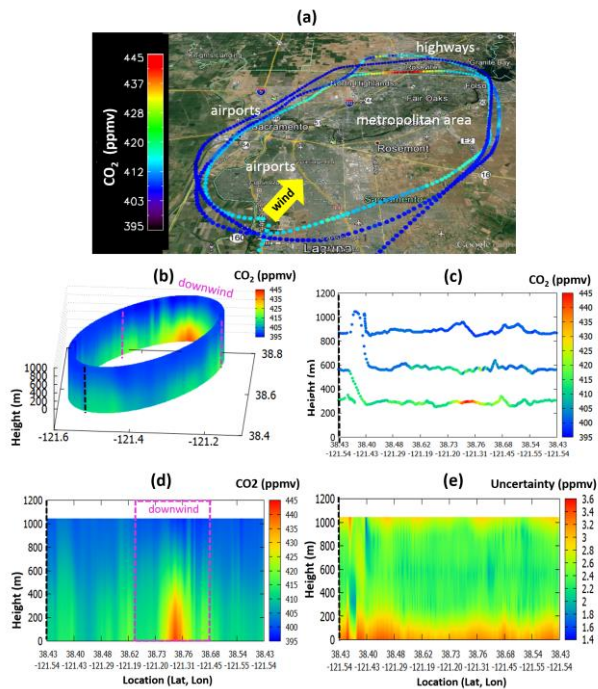


Figure 5: (a) A map of AJAX flight track overlaid by the CO₂ mixing ratio, plotted in Google™ Earth, (b) the kriged CO₂ mixing ratio, (c) Measured CO₂ mixing ratio, (d) the same as (b) except plotted in two dimensions, and (e) the kriging uncertainty at each grid point on November 18, 2013. The yellow arrow represents the dominant wind direction, and the black dashed lines indicate where the surface is split open. The area enclosed by the magenta dashed lines represents the area we used as a downwind portion.

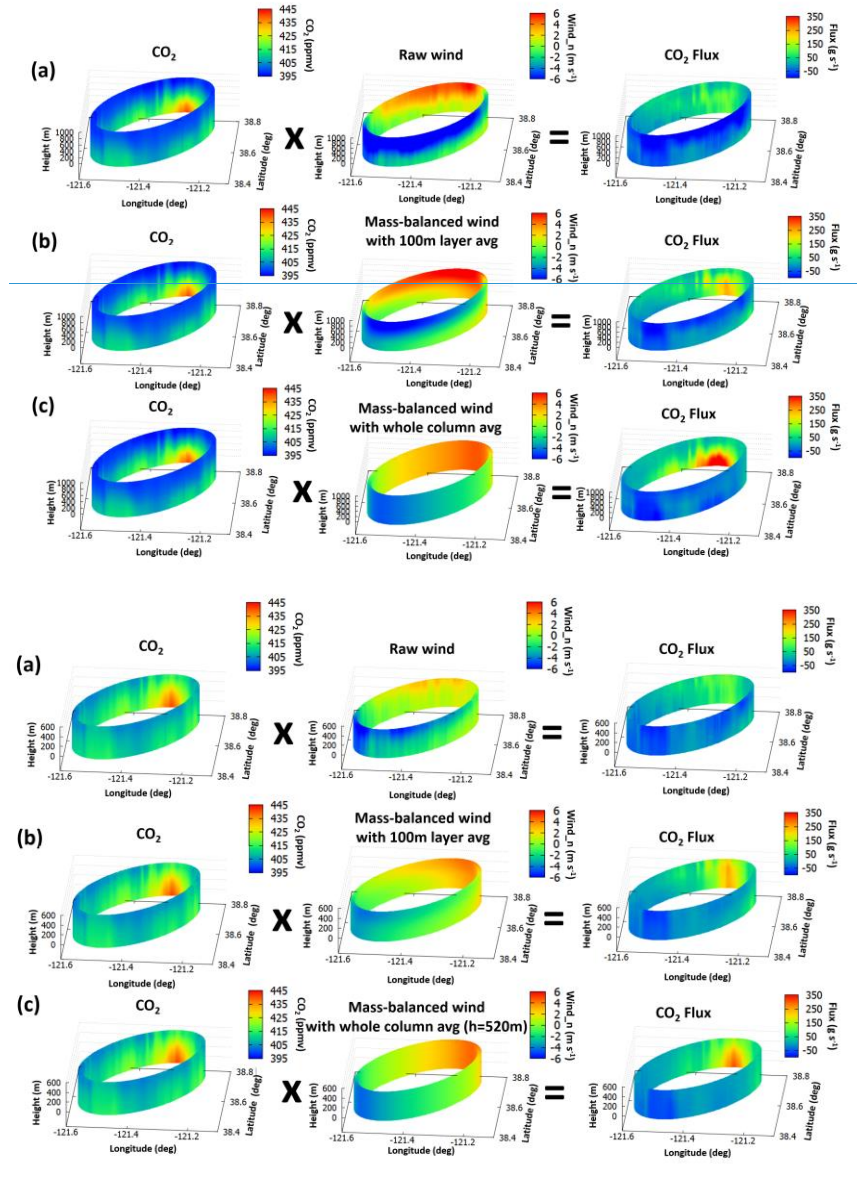
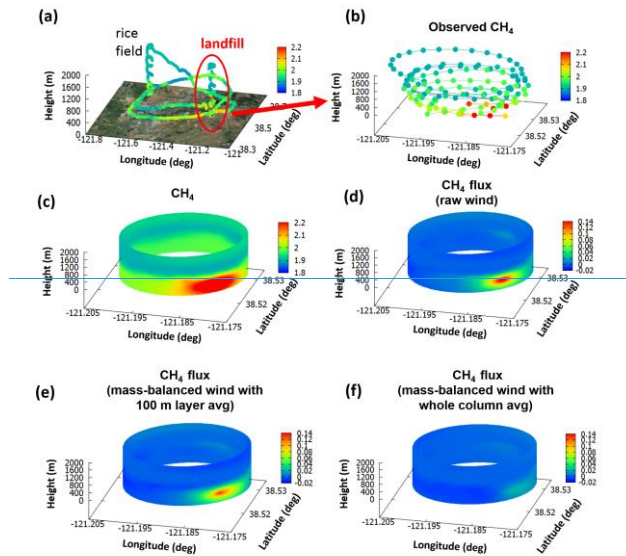


Figure 6: (a) Kriged CO₂ mixing ratio, the raw wind, and CO₂ flux using the raw wind, (b) Kriged CO₂ mixing ratio, 100m vertically averaged mass-balanced wind, and CO₂ flux using the mass-balanced wind, and (c) Kriged CO₂ mixing ratio, whole column averaged mass-balanced wind, and CO₂ flux using the mass-balanced wind on November 18, 2013. In the middle columns, the blue color represents the inflow toward (and red outflow from) the cylinder so that it is defined as negative (positive) wind. The background CO₂ was chosen as the minimum mixing ratio at each vertical layer. The whole column average means the vertical average from the surface to the top of the planetary boundary layer height (PBLH). Figures are only shown below the PBLH.



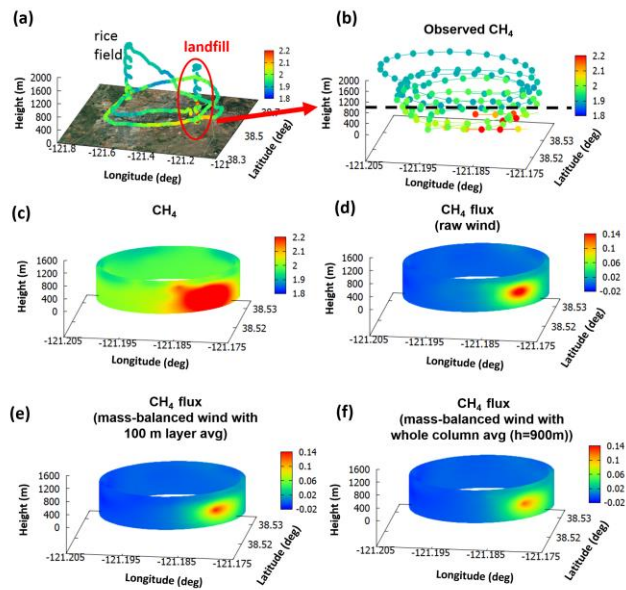


Figure 7: (a) The map of AJAX flight track with the observed CH_4 mixing ratio. (b) The observed CH_4 mixing ratio, (c) kriged CH_4 mixing ratio, (d) CH_4 flux using raw wind, (e) CH_4 flux using the mass-balanced wind with vertically-100m vertically averaged wind, and (f) CH_4 flux using mass-balanced wind (vertically-averaged through the entire PBLH every 100m) over the landfill location on July 29, 2015. The fluxes are computed based on equation (2). The background value was chosen as the minimum value at each vertical layer. The approximate diameter of the cylinder is 3 km, and the color scale is capped at 2.2 ppmv in panels (b) and (c). The black dashed line represents the top of the PBLH for this flight. Figs. (c-f) are only shown below the PBLH.

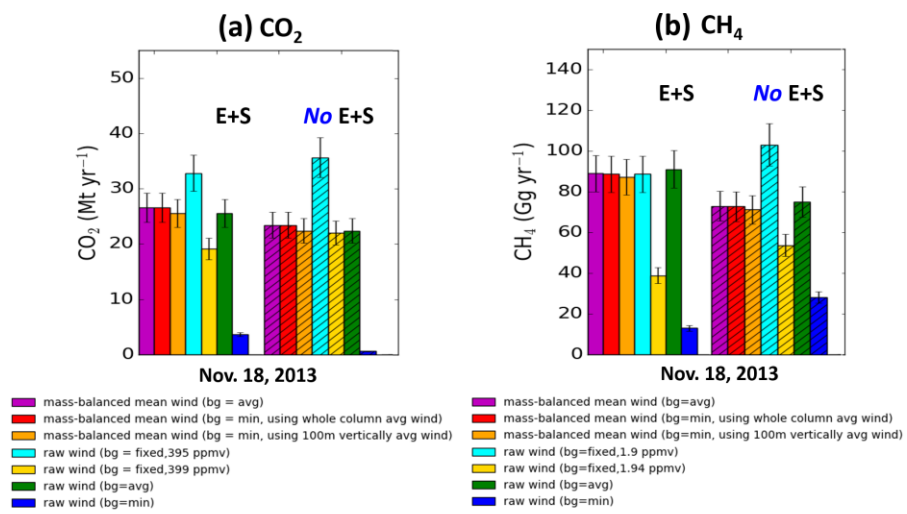


Figure 8: Urban-scale (a) CO_2 [Mt yr^{-1}] and (b) CH_4 [Gg yr^{-1}] emission rate estimates using raw wind and mass-balanced wind, with different background treatments. Solid bars represent emission estimates including entrainment and surface flux (E+S), and hatched bars represent the corresponding emission estimates without consideration of entrainment and surface flux (No E+S). Error bars represent the uncertainty of the total emission fluxes. The average and minimum values for background are computed at each vertical layer (100 m or whole column average), and the fixed value alternatives are 395 or 399 ppm for CO_2 and 1.90 or 1.94 ppm for CH_4 for all altitudes.

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Table 1. Urban scale CO₂ and CH₄ fluxes over Sacramento using different wind treatment (raw wind: “raw”, or mass-balanced wind: “mass-balanced”) and different background values (minimum or average values) on November 18, 2013. The vertical mass transfer (entrainment and surface flux) is included in these calculations. “Whole column avg” means the vertical column average up to the estimated PBLH.

Background	Wind		Urban Scale (large loop)			
			November 18, 2013			
			CO ₂ (Mt yr ⁻¹)	Difference from base case (percent)	CH ₄ (Gg yr ⁻¹)	Difference from base case (percent)
min	Mass-balance	100 m layer avg	25.6±2.6		87.1±8.7	
		Whole column avg	26.6±2.7	3.9%	88.7±8.9	1.8%
avg	Mass-balance	100m layer avg	25.6±2.6	<1%	87.4±8.7	<1%
		Whole column avg	26.6±2.7	3.9%	89.0±8.9	2.1%
min	Raw		3.7±0.4	86%	13.0±1.3	85%
avg	Raw		25.5±2.6	<1%	91.1±9.1	4.6%

Background	Wind		Urban Scale (large loop)			
			November 18, 2013			
			CO ₂ (Mt yr ⁻¹)	Difference from base case	CH ₄ (Gg yr ⁻¹)	Difference from base case
min	Mass-balanced	100 m layer avg	25.6±2.6		87.1±8.7	
		Whole column avg	26.6±2.7	3.9%	88.7±8.9	1.8%
avg	Mass-balanced	100m layer avg	25.6±2.6	<1%	87.4±8.7	<1%
		Whole column avg	26.6±2.7	3.9%	89.0±8.9	2.1%
min	Raw		3.7±0.4	-86%	13.0±1.3	-85%
avg	Raw		25.5±2.6	<-1%	91.1±9.1	4.6%

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Table 2. Local scale CO₂ and CH₄ fluxes over landfill and rice field over Sacramento using different wind treatment (raw wind: “raw”, or mass-balanced wind: “mass-balance”) and different background values (minimum or average values at each level) on July 29, 2015. The vertical mass transfer (entrainment and surface flux) is taken into account. [“Whole column avg” means the vertical column average up to the estimated PBLH.](#)

Background	Wind		Local Scale (small loop): July 29, 2015							
			Landfill				Rice Field			
			CO ₂ (10 ³ Mt yr ⁻¹)	Difference from base case (percent)	CH ₄ (Gg yr ⁻¹)	Difference from base case (percent)	CO ₂ (10 ³ Mt yr ⁻¹)	Difference from base case (percent)	CH ₄ (Gg yr ⁻¹)	Difference from base case (percent)
min	Mass -	100 m layer avg	2.2±0.8		7.1±2.5		2.5±0.4		2.5±0.4	
	balance	Whole column avg	2.2±0.8	<1%	6.9±2.4	2.8%	2.5±0.4	<1%	2.6±0.4	2%
avg	Mass-	100 m layer avg	2.2±0.8	<1%	7.1±2.5	<1%	2.5±0.4	<1%	2.5±0.4	<1%
	balance	Whole column avg	2.2±0.8	<1%	6.9±2.4	2.8%	2.5±0.4	<1%	2.5±0.4	<1%
min	Raw		3.7±1.3	68%	8.9±3.1	25%	1.7±0.3	30%	1.8±0.3	28%
avg	Raw		2.2±0.8	<1%	7.1±2.5	<1%	2.6±0.4	2%	2.5±0.4	<1%

Background	Wind		Local Scale (small loop): July 29, 2015							
			Landfill				Rice Field			
			CO ₂ (10 ³ Mt yr ⁻¹)	Difference from base case	CH ₄ (Gg yr ⁻¹)	Difference from base case	CO ₂ (10 ³ Mt yr ⁻¹)	Difference from base case	CH ₄ (Gg yr ⁻¹)	Difference from base case
min	Mass -	100 m layer avg	2.2±0.8		7.1±2.5		2.5±0.4		2.5±0.4	
	balanced	Whole column avg	2.2±0.8	<1%	6.9±2.4	-2.8%	2.5±0.4	<1%	2.6±0.4	2%
avg	Mass-	100 m layer avg	2.2±0.8	<1%	7.1±2.5	<1%	2.5±0.4	<1%	2.5±0.4	<1%
	balanced	Whole column avg	2.2±0.8	<1%	6.9±2.4	2.8%	2.5±0.4	<1%	2.5±0.4	<1%
min	Raw		3.7±1.3	68%	8.9±3.1	25%	1.7±0.3	-30%	1.8±0.3	-28%
avg	Raw		2.2±0.8	<1%	7.1±2.5	<1%	2.6±0.4	2%	2.5±0.4	<1%

Table 3. Flux estimates for the Sacramento urban area from measurements made on November 18, 2013.
 The two "curtain" rows below used the same wind treatments as the "whole cylinder" rows (mass-balanced wind).

^a Turnbull et al. (2011) data was collected in 2009; the value given here was converted from the mean reported value of 3.5 Mt C yr⁻¹ with a 1.1% yr⁻¹ increase in CO₂ flux to adjust to 2013.

^b Bottom-up inventory estimates of the annual total emissions from Sacramento County from Vulcan (Gurney et al., 2009) and the California Air Resources Board CEPAM database (Turnbull et al, 2011) are included for comparison for CO₂. The Vulcan inventory is available only for 2002, and the CEPAM database is available for 2004. We applied a 1.1% yr⁻¹ increase in CO₂ flux to adjust to 2013. Gridded (0.1° x 0.1°) National U.S. Environmental Protection Agency (EPA) Inventory for November 2012 is included for comparison for CH₄. We applied a 0.6 % yr⁻¹ increase in CO₂ flux to adjust to 2013.

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		CO ₂ (Mt yr ⁻¹)	CH ₄ (Gg yr ⁻¹)
Whole cylinder-AJAX	(bg = min, 100m layer avg)	25.6 ± 2.6	87.1 ± 8.7
	(bg = avg, 100m layer avg)	25.6 ± 2.6	87.4 ± 8.7
Curtain-AJAX	(bg = min)	17.3 ± 1.7	64.4 ± 6.4
	(bg = avg)	8.9 ± 0.9	24.1 ± 2.4
Turnbull et al. (2011)		13.4 (with uncertainty of ~ 100%)	
Vulcan estimates for Sacramento		11.5	
CEPAM estimate for Sacramento		10.0	

		CO ₂ (Mt yr ⁻¹)	CH ₄ (Gg yr ⁻¹)
Whole cylinder-AJAX	(bg = min, 100m layer avg)	25.6 ± 2.6	87.1 ± 8.7
	(bg = avg, 100m layer avg)	25.6 ± 2.6	87.4 ± 8.7
Curtain -AJAX	(bg = min)	17.3 ± 1.7	64.4 ± 6.4
	(bg = avg)	8.9 ± 0.9	24.1 ± 2.4
Turnbull et al. (2011)		13.4 (with uncertainty of ~ 100%)	
Vulcan estimates for Sacramento		11.5	
CEPAM estimate for Sacramento		10.0	
Maasackers et al. (2016)			89.7 (58.8 – 120.5)
Gridded EPA data (November)			

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