

Interactive comment on “Assessment of uncertainties of an aircraft-based mass-balance approach for quantifying urban greenhouse gas emissions” by M. O. Cambaliza et al.

Anonymous Referee #1

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General comments.

This was a difficult review to write, on a paper that will be a major contribution to the field once published. At first read the authors appear to do a careful job; it took several careful passes to realize that they do an excellent job identifying and quantifying only a subset of the uncertainty terms of an airborne mass balance calculation. I believe they neglect several key sources of bias and uncertainty:

1) they do not quantify mixed height in the downwind urban plume itself, so are blind to heat island effects that cause emissions to mix to a higher altitude (e.g., the Birm-

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ingham study described in Trainer et al. (1995)) than might be suggested by vertical profiles to the side of the urban plume;

2) they do not quantify detrainment (either episodic or continuous) of urban emissions from the boundary layer to the free troposphere, which can be exacerbated by elevated, buoyant plumes (e.g. the power plant CO₂ plume) and can further be exacerbated by the presence of an urban heat island, or in the presence of clouds;

3) They do not quantify uncertainties induced by non-constant wind field history, although they implicitly assume that winds measured during the downwind transects are unchanged since the time of emission;

The flight patterns described in this study are not sufficient to quantify these effects (or demonstrate they were negligible) but these effects appear to not contribute to the uncertainty estimate. These must be treated rigorously in the uncertainty terms before the draft can be considered for publication. Including these and other terms in the uncertainty is a large reason the Trainer et al. mass flux for CO from Birmingham has a stated uncertainty of 100%. The present draft claims a significantly higher accuracy, which is unjustified until these additional sources or potential bias are addressed, and further is contradicted by the large spread in their own estimates, especially compared to Mays et al., to Hestia, and to the much more well-known emissions from the Harding Street power plant.

It is possible that I've overlooked discussion of some of these points in the manuscript, so perhaps they simply need to be highlighted more clearly. In any case, I would be happy to re-review another submitted draft once these shortcomings have been addressed.

Specific comments.

1. Introduction

29898 line 25: for city-scale GHG estimates, should also cite Brioude et al. (2013) Top-

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down estimate of surface flux in the Los Angeles Basin using a mesoscale inverse modeling technique: assessing anthropogenic emissions of CO, NO_x and CO₂ and their impacts, *Atmos. Chem. Phys.*, 13, 3661-3677, doi:10.5194/acp-13-3661-2013.

2. Methods

29900 line 25: "The city of Indianapolis . . . fossil fuel signal above background should be relatively easily identified." This assertion here in the text is premature; it was likely the basis for selecting Indianapolis as the focus of this study, but the qualitative assertion here is not supported by data at this point in the text. Can the authors provide some context? Signal above background implies some level of knowledge of background variability, so what is the enhancement above that variability expected from a US city of X million inhabitants into a mixing layer of Y meters at a wind speed of Z meters per second, etc. This is a non-trivial assertion. In the absence of large point sources (e.g., the Harding Street Power Plant and Southside Landfill in Indy) the signal above background shown for Indy in Figs. 3 for CO₂ and for CH₄ appears to be quite small. Please rephrase here.

2.2 Aircraft-based measurements

29901 line 25 ff: "Ambient air . . . was pulled through 5 cm diameter PFA Teflon tube. . ." Teflon is not recommended for sampling CO₂ or CH₄. Why was Teflon used rather than stainless or Dekabon tubing, which are recommended and nearly universally used in the literature? Despite the short residence time in the Teflon, wall contact is the relevant parameter here – turbulence, esp. over the inlet entrance length, makes it probably that all of the sampled air sees an inlet surface. What effect could use of a non-recommended inlet material have on the stated accuracy for CO₂ and for CH₄? The discussion of comparability between CRDS and flask sample data on page 29903 does not quantify potential bias due to use of Teflon for sampling lines upstream of these two detection methods.

2.3 Experimental flight design

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29903 line 21: ". . . flight experiments were conducted between 11:00 LT and 16:00 LT when the boundary layer was essentially fully developed." This seems not to be the case for the flight of 1 June 2011 when the boundary layer grew by nearly 50% over the duration of the airborne study. Please rephrase, and define LT as local time please.

29903 line 28: ". . . transects . . . were conducted downwind. . . to the top of the convective boundary layer." A crucial but neglected assumption is buried here. The authors do not discuss the potential for detrainment of urban emissions from the convective (boundary) layer into the overlying free troposphere as a potential source of bias in their calculations of the urban flux. By neglecting to sample systematically above the boundary layer at the downwind transect location, they have no ability to detect or quantify detrainment of the plume. This potential loss process adds bias by removing plume mass from the boundary layer. Assuming no wind shear (in either velocity or direction) between boundary layer and free troposphere, a crosswind pass just above the boundary layer at the downwind transect location is key in detecting the presence of detrainment of urban plume mass. Given any wind shear between mixed layer and free troposphere, however, it becomes more difficult to detect (much less quantify) the potential for detrainment, as the plume aloft can experience very different transport than the plume remaining within the boundary layer. This additional source of uncertainty is not discussed in their error analysis and could potentially add significantly to the overall uncertainty. This process may have contributed to the large day-to-day variability observed for the Indy urban plume in CO₂ and in CH₄ in this work and the May et al. It is difficult to assess quantitatively the degree to which detrainment might have affected the results; however, it is a crucial, potentially large, and certainly variable term in the uncertainty that appears to be wholly neglected in this report. 29904 line 12: Another key, but unexamined, assumption is embedded here. Calculated boundary layer heights from a single location outside of the urban plume (two locations on 1 June) are used to estimate boundary layer height. The presence of any urban heat island effect could bias the flux calculation low, as the urban plume would be mixed to higher altitudes than suggested by the vertical profiles outside of the urban plume. This

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issue was identified in the Trainer et al. reference for the Birmingham urban plume, in large part driving the much larger uncertainty derived by Trainer et al. for the flux of CO from that city. To be fairly assessed for Indy, this report must additionally consider the effects of urban heat island driving a potential bias in downwind mixing heights (or leading to detrainment from the boundary layer over the city) that are not captured by the flight patterns in this study. This represents a second uncertainty term that can not be neglected in the error analysis here.

29905 Equation 1: The limits defining the vertical column over which the data are integrated are given as $z=0$ to $z=z_i$. Here $z=0$ is ambiguous – on page 29912 the lower limit to z is described as the ground height, but in Eqn. 1 $z=0$ could be interpreted as mean sea level. Suggest explicitly setting the limits as $z=z(\text{sfc})$ to $z=z_i$ in Eqn. 1. What limits were used for the calculations in the paper? If the surface height of Indy was used, great, otherwise using $z=0$ meters will introduce an error. Please clarify in Eqn. 1 and verify that the integrals have been calculated between surface height and z_i , not sea level and z_i .

29905 line 10: with transects at multiple altitudes at a single downwind location, the authors have chosen to interpolate in the vertical. This may be problematic (despite its use in May et al.) and certainly introduces non-physical features in the resulting 2D curtain plot. One, emissions from a non-buoyant source (such as landfill CH₄ emissions) are mixed by turbulent transport to the aircraft transect altitude. It is unlikely that enhancements observed aloft are actually disconnected from the ground – i.e., the kriging interpolation routine results in a non-physical lofted plume, and suggest a minimum in CH₄ below the minimum altitude of the aircraft transect, and other minima between each aircraft transect altitude. These are likely artifacts of the interpolation routine. While buoyant or lofted plumes such as the Harding Street power plant CO₂ plume (Fig. 4) can mix downwards from enhancements injected above the surface, these features should not persist downwind over multiple convective cycles in a well-mixed boundary layer, and likely should not be observed in Fig. 4 Two, the enhancements

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observed at different crosswind locations at different transect altitudes, which lead to variability in the interpolation, are just as likely due to wind speed or direction differences at the time of emission (remember, transects at different altitudes are separated by ~20 minutes or so) as they are due to incomplete vertical mixing. I'm left wondering - what quantitative value does this interpolation add to the data? It does produce a striking visual but the statistics are no longer robust in the interpolated field. The text must do a more thorough job justifying the quantitative use of interpolated fields, and correct the non-physical interpolation artefacts between transect altitudes and below the minimum flight altitude, or simply derive their fluxes using the measured data and not the interpolated fields.

29905 line 24: "... the section in the transect outside the projected city limits is used to calculate the mean background concentration..." This neglects a third major assumption: it appears that upwind variability is assumed to be zero for the purposes of the calculation, but this assumption is not stated and its uncertainty is not included in the error estimate. Later in the report the use of two aircraft is mentioned to permit a more thorough assessment of upwind variability, but it cannot be neglected here. Imagine a hypothetical upwind plume of a few ppm in CO₂ (equal to the variability in the background boundary layer) underlying part of the Indy plume. The background assessment from outside the projected city limits would not capture that, and its contribution would unfairly be ascribed to Indy sources. This is typically a negligible bias for point source plumes, characterized by extremely large enhancements compared to ambient variability, but it cannot be neglected when assessing the relatively smaller enhancements that make up a significant portion of the enhancement from an urban area. Since this cannot be corrected for by only flying downwind, it must at least be included in the uncertainty estimate, which appears to have been neglected in this report.

29907 throughout: Here the authors discuss boundary layer growth in good detail. However, this report appears to neglect an additional error term I was expecting to see here – how is detrainment, or mixing of urban CO₂ and CH₄ from the boundary layer

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into the free troposphere – handled? The lack of a horizontal transect above the mixing layer is a critical omission here. The Trainer et al. paper cited for the Birmingham plume analysis found a significant amount of urban emissions above the mixed layer due to detrainment. Here, vertical profiles outside of the downwind urban plume are unable to assess the extent to which detrainment may or may not have affected the observed Indy BL enhancements downwind. Since detrainment can be significant and episodic, and can be further exacerbated by urban heat island effects (see Trainer) its neglect in this report needs to be corrected. I suspect the measurements are not sufficient to assess this term quantitatively, but it must be included in the uncertainty estimate. Its contribution can range from negligible to significant, and will result in a low bias if not accounted for. I further suspect this contributes to the large variability in derived fluxes for CO₂ and CH₄ between transects and between flight days in this report.

29912 line 3: here the lower limit of column density is clearly indicated to be surface height, not mean sea level. What is the average surface height for Indy? What value was actually used in the calculations? See comment on 29905 Equation 1, above.

29914 line 5: It appears, given the invariant values of the backgrounds (392.6 ± 0.5 ppm CO₂ and 1880.6 ± 2.6 ppb CH₄) assumed on 01 June, that the signal from Indy is dominated by the point source emissions from the Harding Street power plant (for CO₂) and the Southside Landfill (for CH₄). Their contributions are quantified later in the text, but it is difficult to visualize the signal from the on-road mobile sources of CO₂ given the time series presented in Figure 3. Please consider modifying Figure 3 to give more space to the key chemical parameters (perhaps shrink or remove the H₂O and altitude panels) and include lines and shading to indicate background values of 392.6 ± 0.5 and 1880.6 ± 2.6 in the time series panels of Figure 3. This would illustrate graphically the magnitudes of the enhancements from non-point sources of CO₂ and CH₄ to the total signal from Indy, and indicate the sensitivity of the flux calculation to a non-zero upwind variability in the background. Put another way – how does the ± 0.5 ppm uncertainty compare to the actual enhancement shown in Fig. 3 for CO₂ (and

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similarly for CH₄)? 29914 line 14: Turnbull et al. (2013) is listed as “in preparation”, but is not available so difficult to judge appropriateness here. What is ACP policy on this kind of citation? Not sure if that’s a robust reference at this point. 29916 line 14: Cambaliza et al. (2013) is also listed as “in preparation” – not sure if that’s a robust reference, and it is not included in the references list at the end.

29918 line 2: “. . . the history of horizontal winds prior to the experiment can also be important in the mass balance.” Yet another critical point. The assumption is not stated clearly, but all the calculations assume that the winds measured during the horizontal transects are the same as the winds at the time of emission for the sampled air parcels. This assumption is very clearly spelled out in White et al. and in Trainer et al., but seems to be glossed over here. Any systematic change in wind speed between time of emission to time of measurement results in a direct bias (low or high, depending) in the flux calculation. Assuming measured winds from the aircraft are the correct value to use introduces additional uncertainty in the derived flux, and again I don’t see this explicitly included in the uncertainty analysis. The authors must revisit the error analysis to include the several additional sources of error identified in this review before this draft is ready for publication.

29918 line 24: having two aircraft, one upwind and one downwind, would help reduce errors in assumptions inherent in the flux calculation for area sources. But to realize the improvements described in this draft, the two aircraft would need to be exercised in a purely Lagrangian fashion, so that the same air parcels sampled on the upwind transect by aircraft 1 are sampled again on the downwind leg by aircraft 2. In practice this is difficult, and mixing creates additional hurdles to meeting the Lagrangian criterion in any case. Alternatively, rather than 2 aircraft, the authors neglect to mention that a single aircraft with longer endurance, better speed, and increased range (relative to the one used in this study) can perform both upwind and downwind sampling, thus affording the same advantage as would two aircraft.

29920 line 2: Please confirm the dairy cattle population data are relevant to the flight

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dates in question. Are these numbers averaged over any specific period of time? Some dairy farms can have very high fluctuations in their populations, which can add episodic variability to their emissions.

29921 line 19: mixing height is assumed to be equal to cloud base. Here again the potential for detrainment, or venting into the free troposphere, appears to be unquantified by the aircraft data and neglected in the uncertainty estimate. This should be corrected, especially given the Harding Street power plant CO₂ emissions are released at stack height and likely as a buoyant plume (thus more likely to be vented than a non-buoyant plume released at the surface).

29923 line 4: "However, the observed CO₂ flux... was 60% smaller than reported by EPA." Again, is detrainment or venting of the elevated, buoyant power plant plume a source of this discrepancy? This complication needs to be addressed in the uncertainty if not in the calculation itself. Further, in the recommendations section at the end, it would be appropriate to call for another horizontal transect above the mixed layer height to account for this issue in future Indy experiments.

Conclusions

The conclusions paragraph should be rewritten to reflect the expanded uncertainty budget, including the several additional uncertainty terms identified in specific comments above. The final paragraph seems highly speculative, especially in regard to an airborne mass balance flux experiment for a geographically larger source in complex terrain such as Los Angeles. Given the complexities of recirculation, stagnation, orographic lifting, and venting through multiple mountain passes (and indeed directly up mountain slopes) that are well documented in the Los Angeles basin, the simplistic flow-through model upon which INFLUX and this report are based on is likely inappropriate to apply to Los Angeles quantification. Recommend removing this paragraph – its assertions on the tractability of Los Angeles for this type of study are not supported by the known complexities of LA transport, from lidar studies in the 1980s through the

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NASA and NOAA airborne measurements in 2008 and 2010.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 29895, 2013.

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