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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Assessing London CO_2, CH_4 and CO emissions using aircraft measurements and dispersion modelling" *by* Joseph Pitt et al.

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This paper presents results of a single flight around London, comparing two techniques for evaluating emissions from the aircraft measurements. First, the authors apply Lagrangian modelling methodology to simulate the expected CO2, CH4 and CO mole fractions at each aircraft sampling location based on the UK NAEI inventory. The simulated values are compared with observations to evaluate the accuracy of the NAEI for each species. Second, the authors use the well-established mass balance method. They compare the two methods and argue that the Lagrangian modelling technique is more appropriate, particularly for a location like London where emissions outside of the

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metropolitan region are not negligible.

Thank you to the authors for the excellent writing, which makes it easy for the reviewers to focus on the science. Overall, this is a nice study and it presents a straightforward methodology that will likely be used by many others in the future. I do have some concerns about the details of the comparisons, and particularly in the choice of background. Overall, the paper needs only minor revisions, noted below.

Specific comments: Pg 1 ln 19-20. Consider rephrasing – many in the atmospheric greenhouse gas community are recognizing that the value of atmospheric measurements in the emissions reporting context is in working with existing inventories to evaluate and improve emissions reporting. Presenting these measurements as "independent verification" pitted against inventory methods is problematic.

Pg 2 ln 3-10. Please add some discussion here about sources/sinks that are not included in the NAEI. For CO2 this is mainly biogenic fluxes, which are noted later to be critically important. Please also include a discussion of what sources of CO and CH4 are included in the inventory, and which are not. For example, I suspect that oxidation of biogenic VOCs is not included in the CO inventory. These may be negligible in March, but should still be mentioned. As for the CH4 inventory, does it include all sources, or only anthropogenic sources, and how significant might non-anthropogenic sources be?

Pg 2 lines 21-22. While it is true that comparison of top-down estimates with bottom-up inventories is one important way to use the atmospheric observations, it is certainly not true that the only use of these measurements is to evaluate inventories! Please rephrase.

Pg 4 ln 25. "an altitude-latitude plane."

Pg 5 ln 6-20. Please add a sentence that explains in plain English the principle of what the equations do, rather than requiring the reader to wade through the equations to

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figure out the principle (although the detail of the equations is necessary too).

Pg 5 section 3. Please add some detail about the NAEI. It is spatially explicit, but does it have temporal variability? If so, what kind of temporal variability and how reliable might that be? Diurnal cycles? Seasonal cycles? Weekday/weekend? Are there any existing estimates of the quality of the inventory (and perhaps the quality is different for the different gases)? This becomes important in trying to understand the differences between the inventory and the observations.

Pg 6 ln 24-25. Again, please add a sentence that explains the principle in plain English rather than forcing the reader to work it out from the equations. Eg “The mole fraction enhancement is calculated by subtracting the background value”.

Pg 6 ln 24-32. The choice of background is known to be a key uncertainty in this type of measurement (eg. Cambaliza et al 2013; Heimburger et al, 2017). Unfortunately the research community has not yet come to any conclusion as to how to resolve this. The simple method of taking an average of the values measured on the downwind edges of the plume (as is done here) is far from perfect, even if it might be the best available option given the measurements that have been done. Heimberger et al (2017) showed that there can be significant differences in the values on the two edges, and that in that case, a simple improvement would be to linearly interpolate between the two edges to evaluate background. It is also entirely possible that the background is not uniform and that there are plumes from upwind sources that are not detected because they are inside the urban plume. From Figure 4, it’s apparent that there are a lot of methane emissions upwind of the city that could cause this. Further, there’s an implicit assumption that there are no emissions occurring in the footprint of the edge measurements. This is clearly a bad assumption for this dataset, and so the edge measurements will be biased high (or perhaps low in the case of CO2 if there is significant drawdown in the edges), resulting in an underestimate of the urban emission rate (or perhaps overestimate in the case of CO2). A forthcoming paper (in last phases of review) will discuss this further, but unfortunately is unlikely to be published in time

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to be referenced in this paper. My suggestion is to: (1) Add a figure that shows clearly the background values, how they were chosen, and whether there is any difference between the two edges. (2) A plot of the upwind measurements could also be included to show whether there is any particular concern with plumes coming in from upwind for this dataset. (3) Add figures that show the NAEI CO2 and CO emissions, similar to that shown in Figure 4 for CH4, to give a sense of upwind and edge emissions and how important they might be. (4) If there are no particular concerns with the points above, then stick with the current choice of background. (5) Add some discussion about the uncertainty associated with choice of background and how it might influence the results.

Pg 7 ln 24-30. Looking at figure 5, there's a clear spatial mismatch in the plume location between the obs and simulation. What might be the explanation for this? Given this mismatch, is it reasonable to average over the whole thing and then compare the two methods? This mismatch seems to imply a larger uncertainty than that given by just comparing the means.

Pg 8. Please emphasize throughout the discussion of the comparison that this analysis is for a single flight, and that care should be taken in drawing conclusions about the integrity of the inventory from a single comparison on a single day. Previous authors have shown that when multiple flights are considered, there can be large differences in the calculated flux that are likely due to uncertainties in the top-down flux estimate rather than day-to-day differences in the actual emissions.

Pg 8 ln 10-23. I agree that an incorrect spatial pattern in the inventory could explain at least part of the difference. However, I suspect that the choice of background may be more important and be biasing the top-down estimate low. See earlier comments. Does the NAEI include temporal variability and could lack of temporal variability in the NAEI be an explanation for the difference? See earlier comment.

Pg 8 ln 24 – 35. It's clear than biogenic CO2 will have an enormous influence on the

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calculated flux, and that it can bias the CO2 background quite dramatically (see e.g. Turnbull et al 2015, Cambaliza et al 2013). The statement here needs to be much stronger, “treated with caution” is an understatement! It is simply not possible to compare a flux based on total CO2 with an anthropogenic CO2 inventory unless the biogenic component can be accounted for, likely by either having a good biogenic model or being able to separate biogenic and fossil fuel CO2 in the observations (e.g. using 14C or CO). I would say something like “comparison with the NAEI is not appropriate for this dataset”.

Pg 9 ln 28-34. See earlier comments about choice of background. The same biases occur for this method as for the other method.

Pg 10 Section 3.2.2. Can you come up with a total emission flux for the flux dispersion method, so that the total flux from each method can be compared more directly? As written, the comparison is between the obs/model ratio for each of the two methods. Thus it can’t be determined whether the difference in the ratio occurs because the observed flux rate is different, or the modeled flux rate is different. You argue that the difference is in the modeled flux (actually that you’ve defined the modelled footprint differently in the two cases). By making a slightly different comparison, this could be argued more strongly.

Pg 10 section 3.3. This difference in how the footprint is defined is a good candidate for the difference. There are potentially ways to resolve this in the mass balance method. A good start would be to make an estimate of the footprint of the mass balance, rather than assuming that the footprint is an arbitrary metropolitan boundary.

Pg 11. Conclusions. Please restate the point that the CO2 comparison is invalid because biogenic CO2 is not accounted for. Otherwise the conclusions are very nice.

Figure 1. Please add a larger scale inset to show where this is in relation to the UK, Ireland, etc. Not all of us are well-versed in English geography!

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