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Interactive Comment

Interactive comment on "Cross-validation of inferred daytime airborne CO_2 urban-regional scale surface fluxes with eddy-covariance observations and emissions inventories in Greater London" *by* A. Font et al.

Anonymous Referee #2

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The paper by Font et al. utilizes aircraft-based measurements of CO2 concentrations, as well as surface-based eddy covariance measurements, to calculate the CO2 emission flux for the Greater London area, using a mass balance method, which they refer to as the Integrative Mass Boundary Layer (IMBL) method. They find a range of fluxes from 46 - 104 umol/mˆ2-s. The integrated mass flux measurement, for individual days, largely derives from two transects across the London area, at one altitude, 360m, for six different days in October of 2011. There is a great need for development and assessment of the uncertainty in urban greenhouse gas flux measurements, and the

ability to compare surface EC fluxes with those derived from aircraft is an interesting and important challenge. However, I feel that this paper should not be published in its current form, for largely two reasons - 1. It does not present anything new in terms of methodology, since this method has been applied in various forms for various cities and gases, and, far more importantly, 2. The uncertainty analysis is inadequate, and I believe likely grossly underestimates the actual uncertainty in their data, as discussed below.

With regard to the uncertainty, the mass flux measurements are calculated from two passes over the city in each case, at one altitude. There is little discussion of the aircraft footprint at this altitude, and how that variable footprint impacts day-to-day comparisons, and comparison to the EC fluxes, and the inventory. Nor is there discussion of the impact of vertical variability in concentrations. The paper presents very little analysis of the impact of horizontal and vertical variability in the CO2 field on the calculated fluxes, and the associated uncertainties.

The authors use "validate" in the text (e.g. line 11), and in the paper title. This word should not be used in this context. Methods such as this are not valid, or not; they simply have some level of uncertainty, which I think is poorly defined for this work. The authors should define carefully what they mean when they use the word "uncertainty". Do they mean precision, or do they mean an uncertainty which includes some assessment of the impact of systematic errors resulting from the approach? Regarding the propagation of errors for equation 4, the authors say they use the standard deviation of each of the components. But this does not provide a realistic assessment of the uncertainty in the measurement of the urban flux, it is more a calculated precision on the measurement data that was used to calculate the flux. It, and the text, says nothing about the temporal variability of the actual measured flux, for the footprint that applies. And, since no tracks were repeated, there is a no real measurement of the flux measurement precision. Of course the accuracy of the measurement is likely to be significantly worse than the calculated measurement precision. For equation 4, how

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do you assess the uncertainty from the 3D spatial variation in the measured [CO2]2 and [CO2]1? What fraction of the city does your footprint correspond to? For the entrainment flux, how well do you know w+ above h1 when you are flying below that height? You measure the upwind concentrations at a different time relative to the in-city or downwind concentrations - does the concentration field and/or the fluxes change in that time? Since you only measure the gradient across a small part of the city, what do you think is the real uncertainty in the spatial gradient? What does "standard deviation" mean in this context? It seems there are a large number of unknowns about the spatial and temporal variability in the quantities in equation 4, so, saying that you propagated the standard deviations is a bit unclear. You report "mean urban-regional CO2 surface flux", but don't specify the area that comes from FLEXPART, and what you know about the spatial variability of emissions in the footprint. Are the footprints representative of the whole of GL? This should be clarified. Figure 2 articulates quite clearly how limited the data sets are, from the flight tracks. I do note that the section in the middle of page 13479 does articulate the problem quite well! But the discussion is also odd, in that the idea that the plume is actually Gaussian would apply to a point source. You don't actually assume a Gaussian distribution for your calculations, do you? I think (not sure) you assume that the enhancement in concentration is uniform over some width of the city. The quantitative aspects of your calculation assumptions are not well-articulated in the paper.

For the October 24 flight, you report an uncertainty in the calculated flux of 9%. This is a better uncertainty than what, as far as I know, has ever been reported in the literature, from aircraft-based measurements of surface emissions. I could be wrong, but I think typically the best that has ever been achieved is 3-5 times worse than this. The paper should compare the reported uncertainties to what others have achieved, and explain the differences. If 9% is correct, you should discuss why your method is superior to what has been done previously. For this particular day, where you calculate a flux from the upwind and downwind vertical profiles, you have no data between 360m and the surface. So, how do you fill in this data, and how do you do that to an uncertainty of

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9%? While I could be wrong, I don't think that is possible, and thus the paper, to be defensible, should be more rigorous in how it assesses measurement uncertainties. In addition, you compare to an emission inventory, saying that that inventory has an uncertainty that "is estimated to be 2%". That would be a highly accurate inventory, indeed. My experience is that emissions inventories can be uncertain to perhaps 20-50%, even for a species as straight forward as CO2. You might explain how that is possible. You also compare to the EC fluxes, without comparing the aircraft and EC flux footprints, and discussing that comparison in quantitative terms. On page 13477 you do state that "EC and IMBL fluxes cannot be directly compared because of the spatial and temporal mismatch...", but then you proceed to directly compare them, and say that they are statistically similar (line 13) on that page and in the abstract. I think this component of the paper is the referral to "cross-validation" in the title. I note that, as an example, you have tiny uncertainties in the IMBL method, and in the inventory, for October 24, yet, as shown in Figure 6b, the results differ by far more than the combined stated uncertainties, and you don't discuss this. It is noteworthy that the last sentence in the Conclusions says that the agreement between EC and IMBL "endorse the use of these direct methods to verify CO2 emissions at the urban scale". There are serious contradictions here, that would have to be dealt with before this paper can be defensible and published. If these issues can be repaired, I present more minor issues that should be considered, in the order they arose in the manuscript, below.

1. Page 13466, line 9 - what does "'diurnal flux" mean?

2. Page 13467 - remove the word "validate" from the paper.

3. Page 13471 - is there a word missing after (IMBL)? "method", perhaps?

4. Page 13472 - Can you tell us how you assess the uniformity in space and time of the spatial gradient?

5. Some of the English usage and sentence structure in the paper is poor; for example, see lines 10-13 on page 13474.

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6. Page 13474, lines 18-20 - how do you know the enhancement is spatially uniform, to apply to the whole city?

7. Top page 13475 - given the discussion above about uncertainties, and your stated uncertainties here, the reported fluxes should be to not more than two significant figures.

8. Related to uncertainties and heterogeneity in the CO2 field, on page 13475, you use the term "well-formed". What does that mean? Well-formed, but it grew by over 50%?

9. What is the point of the NOx-O3 discussion on page 13476? It seems like an odd aside.

10. Section 3.2.3 - again, you say the two approaches cant be directly compared, but you do this in Figure 6a, and without discussing the two flux footprints. This should be repaired. Can they be compared, and if so, how?

11. Page 13478, line 19 - how do you determine the "atmospheric variability"? You don't repeat any of the flight tracks, right?

12. Regarding the first line of page 13479, "the uncertainty of the IMBL method" is a propagated/calculated precision, but not, in my view, the uncertainty in the method, including systematic errors. For example, CO2 in the boundary layer directly over a city is almost never perfectly mixed. So, how do you account for the unknown distribution of CO2 between the surface and 360m? Could there be a significant gradient in that range?

13. Bottom of page 13479 - I don't see how methane data would aid interpretation. Would better spatial coverage help you much more? "Gas system emissions" don't emit CO2, so, aren't they irrelevant to your measurements?

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 13465, 2013.