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Atmospheric Chemistry and Physics Discussions

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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.

A. Font et al.

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Dear reviewer,

We appreciate the detailed comments you made on the manuscript. We have reviewed the manuscript following your suggestions and a revised version is now prepared.

Please find below the answers to the specific comments raised, along with an expla-

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nation of how the suggestions have been incorporated into the revised version of the manuscript.

Yours sincerely,

A. Font on behalf of the co-authors.

Reply to specific comments from Reviewer #2:

» The paper by Font et al. utilizes aircraft-based measurements of CO_2 concentrations, as well as surface-based eddy covariance measurements, to calculate the CO_2 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/m2 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 However, I feel that this paper should not be published in its current form, for largely two reasons - 1.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.

Re the methodology presented in the current manuscript. We do recognise that this has been applied previously in other regions and landscapes. However, we are not aware of studies using it in urban areas to calculate surface fluxes from aircraft observations. Cleugh and Grimmond (2001) did use a similar methodology to derive turbulent sensible and latent heat fluxes in Sacramento (US) from radiosonde profiles. In a second study, Zimnoch et al. (2010) used a similar approach to calculate night-time surface fluxes of CO_2 and CH_4 in the city of Krakow (Poland) from measurements taken at 20

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m above the ground level. Surface fluxes of CO_2 were compared to respiration fluxes, but not with anthropogenic emissions given the time when fluxes were calculated for.

Previous studies have calculated urban fluxes from observations taken from light aircraft using a different top-down approach. Mays et al. (2009) and Turnbull et al. (2011) calculated surface fluxes downwind of the cities of Indianapolis and Sacramento in the US by a mass balance approach. This calculates emissions using wind speed data and the difference between the concentration in the downwind city plume and the background.

In this paper we aim to evaluate the applicability of the boundary layer budget method in a dense urban area to calculate fluxes at the city scale. We believe the study does present a novel set of data of CO_2 mixing ratios in the urban mixing layer over a megacity in Europe. The limitations of the method are now discussed more fully (see further comments below). Guidelines for future surveys are also given.

The uncertainty analysis has been redone following the reviewers' comments. Upon reflection, we agree that propagation of the standard deviation in each term of the IMBL equation did not accurately account for the unknowns of the methodology. The main unknowns from the method are the determination of the CO_2 concentration representative of the urban mixing layer, the uniformity of the spatial gradient in time, the mixing layer height in the urban region and the vertical wind speed on top of the boundary layer. These uncertainties are now analyzed and a sensitivity test has been carried out. A range of IMBL fluxes are given for each day.

» 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 CO_2 field on

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the calculated fluxes, and the associated uncertainties.

The footprint area of the IMBL fluxes is stated clearly for each survey (see Sect. 3.2). The following lines have been added to the text with respect to the day-to-day variability in relation to the aircraft footprint (lines 386-399): "Surface fluxes calculated for GL over four days in October 2011 ranged from 46 to 104 μmol CO2 m-2 s-1. Except for the 13 October, IMBL FCO2 were calculated for the time period around 10 am. Day-to-day differences in FCO2 are attributed to variations in the area for which the fluxes were representative of. From the DECC annual CO2 emissions inventory, the spatial pattern is seen to have a concentric pattern with central boroughs of GL having emissions between 40 and 120 μmolCO2 m-2 s-1 ; the boroughs surrounding the centre emissions from 20 to 40 μmol CO2 m-2 s-1; and the outer borough having emissions less than 20 μmol CO2 m-2 s-1. The highest FCO2 was calculated for 25 October when the footprint area encompassed the high emission central boroughs. Although the footprint for 17 October also sampled central London, the calculated residence times of the air were shorter (10-30 s, Fig. 3c, d) compared to 25 October (70-140 s, Fig. 3f). The 17 October the probable footprint also includes the lower emission area of south-west GL. The probable footprint for the 24 October extends over the most outer boroughs to the west and south-west of GL (average annual emissions < 40 μmolCO2 m-2 s-1)."

In order to account for the impact of the vertical and horizontal variability in the CO2 field, fluxes have been calculated to taking into account i) the concentration measured at the ground-level sites in London; ii) changes in time in the 5th and 95th percentile of CO2 concentrations measured in the horizontal transects as a measure of the range of CO2 concentrations in the urban mixing layer. The fluxes resulting from these calculations have been compared to the ones calculated from mean aircraft concentrations only.

» 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.

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The word “validation” has been removed from the manuscript. The issue of uncertainty is addressed elsewhere.

»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 do you assess the uncertainty from the 3D spatial variation in the measured $[CO_2]2$ and $[CO_2]1$? 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?

The uncertainty analysis undertaken in the previous version has been replaced by a sensitivity analysis which quantifies the change in the IMBL fluxes when changing one of the different parameters. We agree with the reviewer that the propagation of errors using standard deviations does not quantify the overall uncertainty of the method.

In order to account for the 3D spatial variation in the $[CO_2]$ field, the $[CO_2]$ from one of the stations located in the urban canopy layer has been used to calculate both $[CO_2]1$ and $[CO_2]2$. In order to assess the sensitivity of the method due to the horizontal variability in the urban mixing layer, $[CO_2]$ is calculated using the 5th and 95th percentile of the $[CO_2]$ measured from the aircraft.

The sensitivity of the IMBL method due to the determination of the urban mixing layer is

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evaluated by increasing the value of h in Eq. (4) by 50 m and 100 m. These values have been chosen based on the findings of Spanton and Williams (1988), who documented an increment of between 50 and100 m in the boundary layer height in London and 50 km north.

The sensitivity of the method due to the determination of the vertical wind speed is evaluated by considering its value zero and twice the mean vertical velocity at the top of the boundary layer from the profiles undertaken outside London.

The range of surface fluxes for each survey resulting from changing the above parameters is calculated. Results are described in Section 3.2.3 and summarized in Table 3.

» 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?

The IMBL approach that calculates surface fluxes from upwind-downwind profiles is based on the changes in the concentration of an air mass as it travels across a specific area. The enhancement of the CO2 concentration measured downwind is attributed to emissions taking place in the surface area of the footprint. Instantaneous fluxes in the city will vary in time but the downwind enhancement offers an integrated concentration due to the emissions from the footprint area. Fluxes calculated from the downwind enhancements are spatially and temporally integrated. Additional text is now added to clarify this in lines 179-185: "Second, by quantifying changes of [CO2] above upwind conditions by sampling a vertical profile downwind of the city. This approach, also termed the "column model" (Jacob, 1999) is based on the chemical evolution of a well-mixed column of air extending vertically from the surface to the mixing depth, travelling across the surface. Changes in the chemical composition are attributed to surface emissions taking place in the footprint area. The enhancement in concentration in the downwind profile is a spatial and temporal integrated measurement of the fluxes taking

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place in the footprint region.”

» 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.

We agree that sampling only one horizontal transect does not quantify the spatial variability in the entire urban mixing layer. Fluxes too are not representative of the entire city but for the footprint area.

We agree that propagating the standard deviations does not fully account for the uncertainty in the spatial gradient. Details of how these are now analyzed are given above.

»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.

The term “mean urban-regional CO2 surface flux” has been removed from the text; it is not accurate. The fluxes calculated by the IMBL method are representative for the footprint area and not for the entire urban region. In the paper, for each day it is clearly stated the area that the fluxes are representative of and the percentage of the total urban region covered.

The emissions from the DECC inventory used in this study are given at the borough level. The emissions in the footprint area are calculated from the residence time (calculated by the FLEXPART model) of the air masses over each borough multiplied by the emissions at the borough. That is explained in the text (lines 215-219): “In order to compare IMBL with bottom-up fluxes, a spatially integrated surface flux from the emissions inventory was calculated for each survey. Annual emissions are scaled by the residence time that air masses spent in each borough as given by the FLEXPART model, and divided by the total surface and by the total residence time that air masses spent in the GL surface layer."

Comparison of IMBL fluxes with emissions in the footprint area according to DECC are given in Table 2.

»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.

The explanation about the Gaussian plume has been removed from the text as we agree that it did not reflect the problem. The discussion about the limitations of the methodology is explained as follows (lines 446-453): "The assumption that aircraft observations were above the neighbourhood blended internal boundary layers and within a city-wide UML may not be appropriate. This may suggest that in a megacity such as London it may be necessary to consider internal boundary layers (IBL) within the city. The atmosphere above the outer boroughs, which are more extensive and typically have shorter roughness elements (e.g. buildings), may be well mixed, but over the central business district areas where the buildings are much taller the BL at the flight height may be the IBL for that area rather than the fully mixed UML. Thus more detailed knowledge of the BL dynamics over urban areas is critical. Downwind [CO2] enhancements over a background concentration sample a more representative concentration of the mix of emissions taking place in the urban environment. The footprint analysis of downwind profiles allows calculation of the urban areas potentially influencing the CO2 enhancements. A single pair of upwind-downwind profiles does not sample the entire urban area (Fig. 3e and f). In order to overcome this, multiple downwind profiles

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should be sampled in future surveys”

» 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 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 the revised paper the propagation of errors is not calculated from the standard deviation, hence this figure is now not in the text and a range of IMBL fluxes for that day is now given in Table 3. Fluxes for that day ranged from 33 to 51 μmolCO2 m-2 s-1. The range of fluxes is $\sim$50% the median value of IMBL fluxes for that day.

Unfortunately, data below 360 m for the location where the vertical profiles were carried out are not available. However, the CO2 mixing ratios measured at the urban canopy layer (UCL) at Tower Hamlets (TH) were similar to the CO2 mixing ratios measured in the aircraft transects near the UCL site (see new Figure 2): 400 ppmv (TH) and 400-402 (aircraft) on the 24 October; 396 ppmv (TH) and 395 ppmv (aircraft) on the 25 October. This indicates that the CO2 field below the aircraft cruise height was well-mixed and little variation in the calculated IMBL fluxes would be expected.

» 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

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possible.

The emissions inventory at the borough level used in this study is derived from the UK National Atmospheric Emissions Inventory (NAEI). The uncertainty claimed by NAEI is 2%. However, the uncertainty of large point sources such as urban areas are expected to be higher (100%) (CMEGGE, 2010). Additional text has been added to clarify this (lines 210-214).

»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 CO_2 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.

The comparison between IMBL and EC fluxes is discussed as follows (lines 365-385): "The airborne based IMBL and tower based EC fluxes are complementary because of their different spatial and temporal resolutions and limitations (Lloyd et al., 2007, Desai et al., 2011). As there were two EC systems in central London the variability in the CO_2 fluxes within the vicinity of these sites can be assessed (Fig 6.a). The sensors at the two EC sites are located at different heights. The lower KSK site is expected to be more influenced by micro-scale variability than the taller KSS sensor (Kotthaus and Grimmond, 2012, 2013). Thus, typically the footprint for the former is smaller than that latter (Kotthaus and Grimmond, 2013). These neighbourhood scale fluxes have much a smaller source area (~10ˆ0 km2) than flight based IMBL flux estimates (10ˆ2-10ˆ4 km2) (compare Fig. 15 in Kotthaus and Grimmond, 2012; and Fig. 3 with aircraft footprints). It should be noted that at both scale the sources area locations should be regarded as probabilistic, with the relative size and location being approximate. As the EC flux data are from central London where there is little vegetation and large sources of vehicle emissions the timing of human activities have an important control. At the city-scale the timing of these activities are also important but the larger area extent means that their impact is ore spatially and temporally integrated" The statistical comparison of EC fluxes and IMBL estimates have been removed from the text, as have the contradictory sentences. The title of the manuscript has changed to: "Daytime CO2 urban surface fluxes from airborne measurements, eddy-covariance observations and emissions inventories in Greater London in October 2011."

» 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?

It meant fluxes during daytime. It has been amended in the manuscript (line 21).

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

The word "validate" has been removed from the manuscript.

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

The word "method" has been added (line 143 of the manuscript).

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

In order to account for the uniformity in space and time of the spatial gradient (ΔCO2/Δx) and the advection term, the spatial gradient has been calculated for both time 1 and time 2. The advection term (Fadv) has been calculated using the mean con-

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ditions of wind speed and spatial gradient for the first and second transect undertaken over London. The resulting fluxes are summarized in Table 3.

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

We have attempted to improve this sentence and the paper more generally (lines 230-232).

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

The method does not account for fluxes for the entire urban area. The calculated fluxes are representative for the footprint area. This is explained in the manuscript and the percentage of urban area covered for each day is now stated. In order to avoid confusion the term "surface fluxes at the regional scale" has been removed from the manuscript and from the title.

» 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.

Significant figures have been amended.

» 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%?

We meant that "the CO2 mixing ratio was homogenous below the cruise altitude". The sentence has been amended accordingly (lines 268).

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

These comments have been removed from the manuscript.

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» 10. Section 3.2.3 - again, you say the two approaches can't 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?

Our paper compares fluxes calculated from the IMBL method (representative of a scale ∼10ˆ2-10ˆ4 km2) with continuous eddy-covariance measurements in central London (representative of an area of ∼10ˆ0 km2). Comparison of regional fluxes from boundary layer budget methods with local-scale fluxes from eddy-covariance flux towers has been undertaken in landscapes with mixed vegetation such as the Amazon forest (Lloyd et al., 2007) and a mixed boreal forest in central Sweden (Levy et al., 1999). Levy et al. (1999) argued that although the correspondence would not be exact for the two fluxes, as the source areas are different for the two methods, results should be within the same range and show a similar variation day to day.

This has been added in the text (lines 380-385).

The statistical analysis comparing the two means has been removed from the revised paper. However, Figure 6a is retained to show the order of magnitude and the day-to-day variation of EC observations and IMBL estimates.

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

The reviewer is correct, flight tracks were not repeated hence the atmospheric variability in the surface fluxes was not quantified. This sentence has been removed from the text as it referred to the previous uncertainty analysis that has been rewritten in this version of the paper.

» 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

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of CO_2 between the surface and 360m? Could there be a significant gradient in that range?

In order to evaluate the impact of the distribution of the CO_2 concentration between the surface and the aircraft cruise altitude, the CO_2 measurements within the urban canopy layer at Tower Hamlets have been taken to calculate [CO_2] (see new Figure 2). The resulting flux is referred to in Table 3 as mean {[CO_2]a+ [CO_2]s}.

» 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 CO_2, so, aren't they irrelevant to your measurements?

These comments have been removed from the text.

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