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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.

Anonymous Referee #2

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The manuscript "Assessing London CO2, CH4 and CO emissions using aircraft measurements and dispersion modelling" by Pitt et al. uses an aircraft atmospheric measurement campaign to estimate the CO2, CH4 and CO emissions from the Greater London area. They propose a new approach to estimate these emissions from the airborne concentration and wind measurements and they compare the results from this new approach to that from the more traditional mass balance technique. Their results seem to indicate that the mass balance approach suffers from the lack of knowledge on the footprint of the corresponding flux computation when the targeted source is not isolated. In contrast, their new approach takes advantage of atmospheric transport simulations to better connect the fluxes computed along the aircraft transects to the

surface emissions.

First, I would like to mention that I really appreciated the concision, clarity and quality of the text. The study is based on a well planned measurement campaign and on robust principles of computation. The analysis of the two estimation methods is interesting. This study is clearly worth being published. However, I would encourage to deepen the discussions and maybe analysis in order to better characterize the concepts, strengths and weaknesses of the methods.

1) My understanding is that the new approach is merely a combination or trade-off between two traditional approaches : the mass balance approach and the atmospheric transport inversion (Brioude et al. 2013 provide an example of inversion applied to aircraft data around a city). Conceptually, the major difference between this approach and the traditional atmospheric transport inversion is related to the fact that the observed variables to be fitted by rescaling the surface fluxes are fluxes at the measurement locations rather than concentrations. This requires some additional assumptions for the computation of such fluxes, but this enables to account for wind measurements when assimilating the observations. Another difference is that rather than assimilating all local fluxes at the aircraft measurement locations in a Bayesian statistical inversion framework, the method consists here in summarizing them into an average value which is used to rescale the map of surface fluxes. This simplification could lead to a loss of information but it can also help control the inversion behavior. One of the strength of traditional atmospheric inversions and of this new approach is the ability to extrapolate the information from the sparse measurements by accounting for the atmospheric transport and for the emissions spatial distribution, while the traditional mass balance approach makes coarser extrapolations (here based on a kriging technique).

I think that such a comparison to the atmospheric inversion is worth being discussed since the comparison to the mass balance approach only could lack of hindsight regarding the panel of methods that have been tested to exploit aircraft data. Furthermore, from my point of view, this new approach is closer to the atmospheric transport inversion than to the mass balance approach.

2) One of my main concerns is that by rescaling the total of the NAEI emissions according to measurements whose surface footprint extends well beyond the Greater London area, the new approach does not really inform on the emissions from this area either. Given the distances from the section A-B to London, and as illustrated by Figure 4, the results from this method are driven by emissions from a large part of the South of England that extends to the sea, despite the removal of the "background" concentrations (whose sensitivity to the Western part of the South of England seems much smaller than that of the measurements used to constrain the estimate of emissions according to Figure 4).

The computations are conducted in March so that ignoring the natural CO_2 fluxes might be fine. But similar computations based on the same aircraft campaigns in spring and summer would be highly hampered by the CO_2 uptake upwind and downwind London (not only by the differences between the natural fluxes within the urban part of the measurement footprint vs. within the background footprint that are discussed in section 2.3). While the lack of account for natural CO_2 fluxes is mentioned in section 3.1.2, the major issues raised by these fluxes for spring / summer deserve a discussion, and the topic could deserve some indications in the method sections (in particular in section 2.3) and maybe a coarse look at estimates of the CO_2 natural fluxes in the UK.

I feel that the manuscript is a bit severe with the mass balance approach by crudely attributing the flux estimate from this method to the Greater London area, and maybe by deriving an estimate of the background concentrations for this approach in a crude way. More cautious interpretations of the flux estimates from this approach are usually made, especially for situations like that of London. I would recommend the authors to comment on the paper by Font et al. (2015) who also made estimates of the emissions from London using aircraft data, and who used FLEXPART simulations to assess the footprint of their measurements. O'Shea et al. (2014) also used NAME to analyze the footprint of their aircraft measurements, and discussed the issue that would be raised

by the crude assumption that these measurements would correspond exactly to the greater London area.

Therefore, I would be ready to agree that the mass balance approach and its associated type of aircraft measurement tracks is not very well adapted to the monitoring of the emissions from a city surrounded by other cities and productive ecosystems, especially if flight regulations impose measurements to be conducted far downwind. However, I feel that by relying on the same type of measurements and by avoiding to solve for the spatial distribution of the emissions, the new approach may bear the same fundamental limitation which is the lack of ability for isolating the budget of the emissions from the targeted city. In this regard, I think that the conclusions are a bit optimistic.

3) A critical variable in the study is the wind which is used to compute fluxes. Comparisons between measured and modeled (UK Met Office) winds along the transects but also all around the London Greater area could potentially provide some strong insights on the robustness of the transport model, of the estimate of the measurements spatio-temporal footprint and of the estimate of the surface emissions (in particular if biases arise in the comparisons). I feel that it deserves some analysis.

More detailed comments:

* Introduction

- p2l1: explain that "top-down" relates to methods based on atmospheric measurements and models ?

- p2l5-6: do power plant represent a large fraction of the CO_2 emissions in the greater London area ? on the same topic: I had in mind that the city had large power plants in its vicinity that could represent a major share of the emissions in the measurement footprint (http://naei.beis.gov.uk/data/gis-mapping): is it the case ? if yes, it would feed my main concern (2).

- p2l15: I am not sure about the meaning of "bulk area flux" here. What would prevent atmospheric inversion to provide such a bulk area flux based on the same data ? see my main point (1)

- p2l21-33: I feel that the problem of defining the footprint of the estimated flux is presented in an "inverted" way which makes things more complicated than they are. In particular there is no reason to necessarily involve inventories in this problem.

* section 2.2

- this section should provide the duration and the period of the day corresponding to the flight. Maybe I missed it in the following, but the time of the measurements is a critical information that can raise questions regarding the temporal representativity and the robustness of the computations

- p4l11: I do not understand the end of the sentence ("so as to assess the representativeness . . .") in its context

- p4l13: "less than 24 hours" -> 24 hours is large if considering the need to connect the measurements to an emission footprint both in space and in time, and given the strong diurnal variations of the fluxes. Can the statement be more precise based on NAME simulations ?

* section 3 (beginning)

-p5l30: maybe you should already clarify here the fact that NAEI provides annual budgets of the emissions only, while the measurement were made during daytime in March, which corresponds to a period of relatively high emissions (this information is limited to the discussions on the CH4 results, and just ignored for CO and CO2 in section 3.1.2). Using constant emissions in the model may also be problematic because the duration of the measurement campaign is about 2.5h, during a period of the day when emissions could be highly variable.

-p6l1-3: this will be forgotten when discussing the results, while this potentially weak-

ens the confidence in the results from both methods; but this inter-annual change at the national scale may be negligible compared to the seasonal, day-to-day and diurnal variations biasing the comparison between annual budgets in NAEI and the flux estimates for daytime in March (see my comment above)

* section 3.1.1

-p6l22-23: The sentence (especially "enabling us to specifically assess accuracy. . .") seems to ignore the significant fluxes upwind and downwind London; see my point (2)

-p6l31: the latitudinal gradient is not fully accounted for since the background to be removed from local concentrations is taken as a constant value (the average between the north and south backgrounds) rather than as a linear interpolation between the north and south backgrounds; these north and south backgrounds sometimes seem to strongly differ: isn't it an issue (at least as significant as the one raised on p7l1-2) ?

* section 3.1.2

- p7l17: do the measurements and/or simulations show a significant change of vertical gradients in the concentrations when crossing this BLH $\sim$750m (it does not seem to be the case in Figure 2) ? does the vertical distribution of the concentrations say something about the reliability of the model ?

- p7l29: see my main point (2), you need strong assumptions to apply the scaling factors derived for a large part of the South of England to the Greater London area.

- p7l33: I think that this statement is a bit extreme, especially since several investigations could be led to provide insights on the transport uncertainties: the analysis of the wind fields (see my main point (3)), of the 2D vertical structure of the concentration measurements, and, maybe, of the measurements around the Greater London area that are not exploited in this study

- p8l20-23 are a bit confusing. I do not really catch how the spatial distribution will be tackled along with the temporal variability.

- p8l30: the human respiration could also be listed as a source of mismatch ?

- p8 in a general way: the authors should try to better connect and discuss together the results for CO and CO_2: why the scaling factors are so different for these two species ? is it due to the natural CO_2 fluxes only ? would not it say something about these natural fluxes ?

- p8l31-32: "we can expect them to underestimate" -> shortcut

* section 3.2.1

-p9l30: "horizontal boundaries" could be rephrased for clarity. Could the definition of the background as the average concentrations over the 15-km boundary sections be too crude for focusing the emission estimate on the Greater London area (is the 15 km distance too short) ? does this background fit well with the background estimated with the flux dispersion method ?

-p10l1-3: the discussion goes a bit too fast for me. One could also assume that the upwind concentrations are more suitable to define a background for the measurements downwind London because they would characterize a section across their footprint that is relatively close to the sea (Figs 1 and 4). Discussing the impact of BLH on background concentrations could mean that these background concentrations are mainly driven by fluxes that are relatively close to the measurements. However, the concentrations North and South of the transects A-B are mostly influenced by fluxes North and South of London that are hardly seen by the measurements downwind London, as indicated by Figure 4. In a more general way, I think that the characterization of the "background concentrations" and footprint for the measurements downwind London could be better discussed (see my main point (2)).

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