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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of the potential of near ground measurements of CO_2 and CH_4 in London, UK for the monitoring of city-scale emissions using an atmospheric transport model" *by* A. Boon et al.

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Author response to Reviewer #2 We would like to thank the reviewers for their useful comments and their positive assessment of our study.

Reviewer comment: The manuscript is well-written, with precise terminology (see however the comments on the use of "misfits" and "signature" in the accompanying pdf) and detailed descriptions of the methodology and data analysis.

Author response: We thank the reviewer for this general comment. We will give a

more precise and more visible definition of what we called misfits (which will now be called discrepancies following the reviewer's suggestion below) and signature as early as possible in the revised manuscript.

Reviewer comment: The manuscript seems however "methods-heavy" which makes the results and discussion section seem a little thin at times.

Author response: Since we have conducted both measurements and model simulations, we feel that it was necessary to go into such a number of details on the method. Still, we think that our conclusions apply to a wide range of models and measurement situations, and we will clarify and highlight it better.

Reviewer comment: Interpretation of the data is sometimes too qualitative and speculative, especially for the discrepancies between measurements and model.

Author response: We will more systematically refer to the diagnostics statistics of the model-data discrepancies to support our interpretations.

Reviewer comment: As a result of this, the conclusions are a little disappointing (e.g. "this study strongly questions the ability to exploit a GHG network with near ground urban measurement sites alongside a state of the art atmospheric inversion system with atmospheric transport models at kilometric horizontal resolution.")

Author response: We will describe more specifically what "near ground", "urban", and "state of the art inversion system" mean here and we will add "currently" in the new sentence. However, we cannot realistically be more affirmative since the modeling of CO2 and CH4 within urban areas, where dedicated to CO2 and CH4 emission atmospheric inversions, is an emerging activity with a fast growing community and breakthrough improvements can be expected in the coming years.

Reviewer comment: and it would have been interesting to explore and report on ways to improve the results.

Author response: We will provide ideas directly derived from this study including pro-

moting measurements at more than 20magl, using networks with different types of measurements (e.g. integrated column measurements) or with sufficiently dense sampling that averaging their data could be informative about the spatial scales relevant to the model, using local (for each site) very high resolution simulations to help detect under which conditions the large scale signal vs. local signals could be filtered from the measurements.

Reviewer comment: As it stands, this work does not offer a credible alternative to more conventional bottom-up or top-down approaches for estimating greenhouse gas budgets at the city-scale.

Author response: We do not aim to propose an alternative to top-down approaches, but to help design it. We insist (and will make it clearer in the revised paper) that approaches using measurement sites outside the core of the urban areas have worked (see Bréon et al. 2015) but that the use of measurements at "cheap" (without much infrastructure) locations within the core of the urban area would strengthen the capabilities of the approach. The methods proposed above (in the answer to the previous comment) could help to make it work.

Reference: Bréon FM, Broquet G, Puygrenier V, Chevallier F, Xueref-Remy I, et al. (2015) An attempt at estimating Paris area CO2 emissions from atmospheric concentration measurements. Atmos Chem Phys 15: 1707-1724.

Reviewer comment: I anticipate however that this work should be of interest to the specialist scientific community. I therefore recommend that the manuscript be reconsidered for publication in ACP once the comments detailed in the attached document have been addressed.

Author response: We thank you for this recommendation.

Reviewer comment: My main concern with this manuscript is that it demonstrates a "non-proof" of concept in the sense that despite its rigour the methodology does not

deliver the anticipated solution.

Author response: The study still proposes techniques for defining the data to be assimilated in the inversion system. We will better highlight the "positive" results from the analysis. The title should be changed to reflect this. The existing title refers to the potential of the method which belies the ultimate conclusion that the proposed method does not advance the state of knowledge within the field.

Author response: We think that the situation is a bit more complex. As stated above, our analysis cannot indicate that we will never be able to use near ground measurement in the near future. It details the issues related to such a type of measurements with state of the art techniques but also approaches to better extract information from them and, thanks to the reviewer's comment above, it will raise some possible solutions for circumventing these issues. Our understanding of the expression "analysis of the potential" is that it will not necessarily demonstrate that this potential will be high. Therefore, we would prefer to keep such a title.

Reviewer comment: Whilst it is interesting to learn that the methodology did not work as well as anticipated, the manuscript needs finish on a high by either presenting credible improvements or at least suggesting new approaches.

Author response: As indicated above, we will follow this suggestion.

Reviewer comment: The data analysis needs to be more quantitative; the authors mention the "signature" of emissions at length but it is still unclear to me what this quality might be.

Author response: We will better refer to numerical values from our diagnostics when discussing the results, and as indicated above, we will provide at first mention a sort of systematic definition to the term signature (i.e. the amount of CO2/CH4 at a given time of location, and of its variation due to the emissions, also called "response function" in the inverse modeling community).

Reviewer comment: General comments 1. Inconsistencies with the cited literature have been found (see for example the comment about the Rigby et al. (2008) paper listed in the technical comments.

Author response: See the answer to the corresponding comment, we made a small mistake regarding this study and we will correct the text accordingly.

Reviewer comment: Please, check all references to ensure that the work and methods attributed to them is correct.

Author response: We have checked that there are no further mistakes in the literature survey. The revised manuscript will be updated for more recent studies and recent work in London, according to the comments below (see answers below).

Reviewer comment: 2. London has been the subject of several publications but the references to the literature are incomplete. Consider adding the following (the list is not exhaustive and you should conduct a thorough survey):

Kotthaus, S., and Grimmond, C. S. B.: Identification of micro-scale anthropogenic co2, heat and moisture sources - processing eddy covariance fluxes for a dense urban environment, Atmospheric Environment, 57, 301-316, 10.1016/j.atmosenv.2012.04.024, 2012.

Ward, H. C., Kotthaus, S., Grimmond, C. S. B., Bjorkegren, A., Wilkinson, M., Morrison, W. T. J., Evans, J. G., Morison, J. I. L., and Iamarino, M.: Effects of urban density on carbon dioxide exchanges: Observations of dense urban, suburban and woodland areas of southern england, Environmental Pollution, 198, 186-200, 10.1016/j.envpol.2014.12.031, 2015.

Reviewer comment: 3. The introduction should present the current state of urban research into GHGs more broadly (see for example Helfter et al. (2011) and Ward et al. (2015) for references) and list the different measurement and modelling approaches applied for completeness.

Author response: We will provide a more detailed literature survey regarding GHG fluxes and transport in London, including these papers and improving the analysis of Hefter et al. and Ward et al. Note, however, that most of them relate to types of scales, processes and objectives that are different from the those analysed in our study. In particular, there has been a significant number of studies mainly dedicated to eddy covariance flux measurements for the derivation of local flux estimates based on local scale transport processes (the link between the fluxes and the concentrations mainly relies on local vertical transport for such approaches). In contrast, the atmospheric inversion approach aims to filter the CO2 signal with a large scale representativity to derive city scale emissions (the link between the concentrations and the fluxes mainly relying on large scale horizontal advection within a well-mixed PBL). It is thus difficult to exploit studies on eddy covariance measurements for supporting our analysis. Lengthening the list of publications on such an activity would be outside of the scope of our study.

Reviewer comment: Specific comments Abstract Line 13 and throughout: Consider changing “misfits” into “discrepancies”.

Author response: We will do it throughout the text.

Reviewer comment: Line 14: “signature of the errors”. . . this is unclear. Line 27: again, it is unclear what the term signature refers to in this context.

Author response: As indicated above, we will provide a clear definition of this term as early as possible.

Reviewer comment: Introduction Page 33006 Lines 13-14: “Atmospheric measurements” is too vague. I interpret the sentence as meaning any type of atmospheric measurements but the references appended to that sentence do not reflect the broad variety of urban measurement sites and techniques used in the last 20 years.

Author response: The two first sentences of this paragraph will be merged to make it

clear that we speak about atmospheric inversions using GHG atmospheric concentration measurements.

Reviewer comment: Line 23: to my knowledge the Rigby (2008) study was conducted at the campus of Imperial College London and at Royal Holloway University of London and not the BT tower. Please check this reference and revise the manuscript if need be. In addition, clarify the measurement approach used by Rigby et al.

Author response: We made a mistake in the manuscript and we apologize for this. The text will be revised accordingly and we will give more details on this experiment that is relevant for our study.

Reviewer comment: Page 33008 Line 3-15: these bullet points sound like conclusions. Please reword them to make them sound like hypotheses.

Author response: We will do it.

Reviewer comment: Page 33009 Line 9: whilst offshore emissions due to gas production are used to derive the emissions inventory, these cannot of course be measured in the city and you should highlight this.

Author response: It would be a bit difficult to conduct such a discussion at this stage; it is just part of the general description of the NAEI inventory. Whether or not offshore gas production never impacts concentrations is not clear-cut and a discussion on this may not fit well in such a section. We will rather simply mention what the main sources are within the London area.

Reviewer comment: Page 33009 Line 16: I seem to remember that the 2009 dataset for CH4 was removed by the NAEI in 2011 or 2012. Could you confirm that the dataset you used is still available from the NAEI and provide the complete web address where it can be downloaded from?

Author response: We confirm that we accessed these 2009 CH4 and CO datasets in 2012–2013 when building these experiments (last access provided in the bibliography:

12/12/2013). Today, more recent data are available and we cannot access the 2009 version of the inventory we have used. However, this is documented in the report

Dragosits, U., Sutton, M.A. 2011 Modelling and mapping UK emissions of ammonia, methane and nitrous oxide from agriculture, nature, waste disposal and other miscellaneous sources for 2009. NERC/Centre for Ecology & Hydrology, 20pp. (CEH Project Number: C03614)

given in the following link: http://nora.nerc.ac.uk/14265/

and which will be added to the bibliography.

Reviewer comment: Page 33010 Line 12: give the percentage of wind occurrences from the south-west for the study period and longer term statistics if available.

Author response: Based on the Heathrow data, 52% of wind occurrences were from the south-west sector. This information will be added to the final manuscript. Deriving statistics in a similar way for a longer period would be quite demanding in terms of data access, treatment and analysis for a small added value on this topic. Shades have been added to Figure 6 to indicate when the wind is in the range chosen for the gradient filtering proposed in Section 3.6.

Reviewer comment: Page 33011 Line 16: this is a very large CO mole fraction! Please, provide a typical range for ambient CO mole fractions measured in London for comparison.

Author response: The CO mole fractions at the London sites ranged from 0.08 to 0.92 ppb according to the measurements. As already stated in the manuscript, unlike the calibration of CO2 and CH4 measurements, it was not possible for CO to use a reference gas within the ambient concentration range. The value of the calibration gas (9.71 ppm) is much higher than the observed values, leading to a larger uncertainty. However, it is important to note that the linearity of the G2401 analyzer has been evaluated by Zellweger et al. up to 20 ppm. Their results show that the CRDS analyzer remains

linear from 0 up to 20 ppm, with residuals from a linear fit not significantly different from zero (+/- 5 ppb) and showing no trend. We will indicate this in the paper.

Reviewer comment: Page 33013 Line 14: is “thickness” the technical term? Consider using height or equivalent instead.

Author response: We will use “vertical resolution” (which is a traditional technical term) instead.

Reviewer comment: Line 19-20: was there an explicit treatment of surface roughness? If so, at what spatial resolution and where did the data come from? If not, explain how the wind speed dampening was scaled to the “fraction of urban area”. What model/ assumptions were used?

Author response: We cannot say that we use an explicit treatment of the surface roughness. We just constrain the surface wind speed to 0 over the urban area, i.e. we rescale the surface wind speed, for a given 2 $\times$ 2 km model grid cell, by (1 $-$ x) where x is the fraction or urban land cover within this grid cell. The land cover is derived from the GLCF (Global Land Cover Facility) 1 $\times$ 1 km resolution database from the University of Maryland, following the methodology of Hansen and Reed (2000) and based on AVHRR data. This will be clarified in the revised manuscript.

Reference: Hansen MC, Reed B (2000) A comparison of the IGBP DISCover and University of Maryland 1km global land cover products. International Journal of Remote Sensing 21: 1365-1373.

Reviewer comment: Page 33015 Lines 18-19: Seasonality in CH4 emissions has been observed in London and elsewhere (see for example Lowry et al. (2001) and McKain et al. (2015)). Quantifying the seasonality might be difficult but you should acknowledge that it might exist.

Author response: In the revised manuscript, we will discuss the fact that these studies have indicated seasonal variations of the CH4 emissions. Processes underlying such

variations will be discussed.

Reviewer comment: Page 33017 Line 22: write “timeseries” as time series.

Author response: This will be changed throughout.

Reviewer comment: Page 33018 It would be useful to define the assumed extent of the “local scale”.

Author response: “Local” is associated with distances from the measurement sites over which the transport cannot be characterized by the Eulerian model. This primarily applies to distances smaller than the size of the model grids i.e. at less than 1–2 km. However, in principle, this can extend further depending on the type (strength and spread) of the sources and on the topography (ground topography and urban canopy) at a distance from the measurement sites. This will be better discussed in the revised manuscript and the local scale will be associated with the typical range of distances of 1–5 km.

Reviewer comment: Page 33019 The term “signature” is not used correctly; it implies a specific characteristic or quality but what you describe is a type of source apportionment. Please revise the manuscript with a more appropriate term.

Author response: We definitely associate “signature” of a given type of source to the source apportionment for the corresponding concentration time series of field. This is usually referred to as “response function” in the inverse modeling community. We would like to keep the term “signature” but to propose a clear definition of this term early in the text to avoid confusion.

Reviewer comment: Why not do a model run with measured boundary layer height rather than modelled ones and quantify the potential bias induced?

Author response: The BLH varies substantially in space in the modeling domain, and it would be difficult to extrapolate the BLH measured at a given site near London into a realistic 2D field. Mixing parameters within the BLH of the transport model are in-

fluenced by variables from the meteorological product whose vertical profile need to be, to some extent, consistent with the BLH. And the BLH used to force the model needs, to some extent, to be consistent with the wind field used to force the model. These consistencies are naturally ensured when using a meteorological simulation for the BLH and other variables. Therefore, it would be quite problematic to constrain the BLH of the model to the value measured at one or few stations near London.

Reviewer comment: You could also look at ratios of CO/CO2 (for wind sectors devoid of green spaces and where traffic can be assumed to be the main common source of the 2 gases) as atmospheric transport should have a limited impact on that quantity.

Author response: We do not have CO simulations and thus the CO/CO2 ratio must be examined with the measurements only, which prevents us from checking the skills of the model for catching it in principle. Furthermore, it is not possible to sort wind directions and speeds for which the urban CO and CO2 measurements would be unaffected by green spaces and traffic since, first, both HAC and POP have trees and housing all around in their vicinity, and, second, even though we highlight the large weight of local sources, the measurements are impacted by larger scale emissions. In particular, they both bear a significant impact from the natural fluxes in Southern England as demonstrated with the model. There is thus no reason to think that the ratio between measured CO over measured CO2 is indicative of the signature of the city anthropogenic emissions. Section 3.7 addresses the relationship between CO and the anthropogenic CO2 once the impact of natural fluxes has been decreased through the computation of the gradients during the afternoon.

Reviewer comment: Page 33029 Equation 1: the same equation appears twice in line with one another. Table 3: define FF-CO2 in the legend. Figure 2: include the units in the plots (not only in the legend). Figure 4: Insert the panel reference letters (b) and (d) for the top and bottom right plots respectively. The font size and line thickness are a bit small and make reading the graphs difficult. Define BC-CO2 in the legend. Figure 5: same comment regarding font size and line thickness as for Figure 4. Figure 6: Same

comment regarding font size and line thickness as for Figure 4 & 5. Define FF-CO2 in the legend (legends should be intelligible e in their own right without any reference needed to the main body of the manuscript).

Author response: These specific amendments will be addressed in the updated manuscript.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C13254/2016/acpd-15-C13254-2016-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 33003, 2015.