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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Megacity and local contributions to regional air pollution: An aircraft case study over London" *by* Kirsti Ashworth et al.

Anonymous Referee #1

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Summary:

This paper discusses concentration measurements for several air pollutants (CO, VOCs, NMHCs and PM), and greenhouse gases (CO2, CH4), and derives Greater London emission fluxes for CH4, CO2, and CO, from aircraft sampling in a series of flights around South East England in July 2017. It uses world-class research facilities (the NERC FAAM aircraft) and calibrated instrumentation and combines the measured geospatial dataset with back-trajectory modelling and simple mass balancing approaches to discuss airmass history and emission sources. The focus is on quantitative emissions from London as a megacity, but the paper also discusses localised and spatially-contrasting sources qualitatively. The use of tracer:tracer ratios for pollution source attribution and ageing is very interesting and offers some very useful guidance

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for those following this work. Put together, this represents an excellent and rigorously compiled dataset, analysed using appropriate modelling and analysis methods, resulting in some incremental conclusions and comparisons with similar previous studies. The paper is rigorous and of high quality in terms of its presentation and analysis. I believe it would be of interest to readers of ACP, especially for those with interests in air quality emissions and chemistry and GHG flux methods. There are just a few suggestions to further improve the manuscript that the authors could consider below, and a couple of potentially important clarifications that may or may not require slight modifications to the analysis.

Specific comments: 1/ The paper compares CH4, CO2 and CO fluxes derived using mass balancing with those reported using highly analogous aircraft sampling and methods by O'Shea et al., 2014 and Pitt et al., 2019. The paper currently discusses the relative quantitative differences with those studies. A further useful dimension to the discussion surrounding that comparison could be more thought as to "why" they differ. Line 355 suggests that the method for defining the background concentrations for mass balancing may be a potential reason for discrepancy between O'Shea and this study, and that the spatial footprint of emissions may be different. I would agree that that is one potential source of difference. However, an equally valid reason is that the emissions from London are simply different at these two very different times. Would it possible to raise some hypotheses on the reasons for truly different emissions? Would a comparison with the NAEI inventory help to elucidate the changing sources with time between 2014 and 2017? If the authors do not wish to go to this next level of interpretation, then simply stating that the emissions are likely to vary with time would go some way to ensuring the reader is left with a more correct (or full and balanced) conclusion. In other words, I don't believe that difference in method and footprint solely explain the different fluxes, which is the message that is perhaps currently conveyed.

2/ The paper often refer to "spikes" or "spiking". I think words such as "concentration enhancement" or "transient enhancement" could be more intuitive.

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3/ All concentrations are reported in units of ppv (e.g. ppbv) in the paper. As far as I understand, most in situ instruments on the FAAM aircraft report molar mass concentration (e.g. ppb). The GHG kit certainly does. This does make a difference for flux calculations as ppbv is not equal to ppb and a molar ratio must be used to convert between the two units prior to use in the mass balance equation. Has this been done? If not, the fluxes may need to be corrected. Otherwise, please confirm how ppb etc was converted to its volumetric equivalent for CO, CO2, and CH4.

Technical comments: 1/ Some units do not have a space between quantity and unit, e.g. 450m (and elsewhere where m are used). 2/ Figure 3 – does not appear well on my screen. Can the axes lines be thickened? 3/ Legend on Figure 12 – hard to read – can a larger font size be used?

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