**Comments on manuscript number acp-2015-686 (Boon et al.)**

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.

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.

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. 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.

Under these considerations, I recommend that the manuscript be reconsidered for publication in ACP once further analyses have been conducted and the comments raised in this document have been addressed.

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

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.

**Specific comments**

**Abstract**

Line 13 and throughout: Consider changing "misfits" into "discrepancies".

Line 14: "signature of the errors"… this is unclear.

Line 27: again, it is unclear what the term signature refers to in this context.

**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.

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.

Page 33008

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

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.

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?

Page 33010

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

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.

Page 33013

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

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?

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.

Lowry, D., Holmes, C. W., Rata, N. D., O'Brien, P., and Nisbet, E. G.: London methane emissions: Use of diurnal changes in concentration and delta c-13 fo identify urban sources and verify inventories, Journal of Geophysical Research-Atmospheres, 106, 7427-7448, 10.1029/2000jd900601, 2001.

McKain, K. K., Down, A., Raciti, S. M., Budney, J., Hutyra, L. R., Floerchinger, C., Herndon, S. C., Nehrkorn, T., Zahniser, M. S., Jackson, R. B., Phillips, N., and Wofsy, S. C.: Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts, Proceedings of the National Academy of Sciences of the United States of America, 112, 1941-1946, 10.1073/pnas.1416261112, 2015.

Page 33017

Line 22: write “timeseries” as time series.

Page 33018

It would be useful to define the assumed extent of the “local scale”.

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.

Page 33020

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

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 in their own right without any reference needed to the main body of the manuscript).