

Fluxes and concentrations of volatile organic compounds above central London, UK.
Langford et al.

This is an interesting paper which presents results from the city flux contribution to the REPARTEE campaign in London. The authors briefly describe diurnal variations in the concentration of 8 VOC compounds measured at a height of 200 m in the urban boundary layer in central London. They then present fluxes of these compounds going on to discuss variations in the fluxes in relation to traffic densities and the depth of the boundary layer.

Although well written, this paper could be strengthened by increased reference to meteorological data and other studies thereby avoiding the need to make unsubstantiated inferences about underlying processes.

Detailed comments:

Introduction

The introduction is very short and does not review the relevant literature in sufficient detail. For example the authors later discuss the impact of the break down of nocturnal boundary layer and it would be useful to explore this in more detail in the introduction. Also the authors rely heavily on the assumption that traffic emissions are the dominant source of VOCs – it would be useful to see whether this is true in other locations.

Methods

It would be useful to have a section here which focuses on the mean surface air pollution conditions observed during the campaign. The authors also comment that surface measurements of at least some of the VOCs studied and CO are available. It would be helpful to provide annual data and data for October to establish the how typical the conditions observed were.

The exact dates of the measurements are not provided; neither is a section on the typical meteorological conditions during the campaign and how they compare to the rest of the year. This is important as later the authors make strong temperature related arguments and use the data as the basis for modelling annual emissions. Thus there is a need to establish that meteorological conditions during the observation period were not only typical for that time of year, but that they were representative of the annual condition.

In the calculation of the flux footprint only a very limited range of conditions were used. The authors do not comment on the grid to grid variability of the emissions estimates, but if this is significant it would be useful to use a wider set of conditions to determine the typical footprint. This is especially true for the night time period when the potential for a weakly developed convective nocturnal boundary layer may complicate the identification of source areas.

A brief comment on the emissions inventory data would also be useful to establish the methods used to create the inventory and determine the predominant sources for the region.

Results

P17306 line 18 The authors comment that the aromatic compounds show two peaks with the second larger peak occurring in the evening. The increased magnitude of the second peak for these compounds is not obvious from Fig 2.

P17307 The authors comment on the long term trends that methanol and toluene show in the data set. These are not provided for the reader, and not compared with other compounds. Reasons for these trends could also be usefully explored and supported by the appropriate data sets.

P17308 The authors discuss the comparison of their data with surface data, however typical diurnal cycles or scatter plots are not provided. The differences between these two data sources could usefully be explored in more detail, especially as the authors later go on to discuss the potential impact of the layering of the boundary layer at night and resulting decoupling of the two layers. If these processes are operating there should be evidence in the mean concentration data sets – notably the diurnal variation in concentration ratio between the two levels and difference in the lag between morning and evening concentration peaks. The authors also argue that measurements at the tower height may be influenced by sources outside the city. This not supported by data but could be effectively explored by examining the relationship in mean daily concentration with wind direction or using back trajectory modelling.

P17308 3.2 VOC fluxes : The authors comment that during some nights the site becomes decoupled from the street canyon activity. However no evidence is provided for this. Model data for the BLD presented in Fig 5 is not convincing as it indicates that the BLD is always above 200m. The details of the model used are not provided to the reader, including whether or not an urban parameterisation has been employed to correct nocturnal estimates of BLD. Further the authors comment that a limitation of the model is that the lowest level it resolves is 250m – thus little is known about conditions when this is likely to be below this value. As a consequence this model was not a suitable choice to support their analysis of the night-time conditions. The authors refer to Barlow et al 2009 as a source of data – perhaps a case study of observations would provide more convincing evidence. Further, there should be evidence of decoupling in the sensible heat flux data. If the authors can show a peak in the heat flux coincident with the peak in the pollutant fluxes this would support their argument more effectively.

As it currently stands the arguments for the decoupling of the nocturnal boundary are based on speculation, which although supported by pollutant flux data is not convincingly supported by mean pollutant concentrations. Why would you expect mean concentrations to start to increase so early if the layers are separated?

P17310 lines 10-20 The authors comment that the differences in the diurnal flux cycles observed between the VOC may result from changes in the sources at a diurnal scale. Please support this statement with references and emissions inventory.

P17311 It would be useful to see the diurnal cycle of CO that is used for the ratio calculations.

P17312 The authors argue that VOC processing is limited at night due to the absence of sunlight and titration of O₃ by NO. This implies that the layer is connected to the surface at night counter to earlier arguments. Perhaps it would be useful to look at ozone concentrations as an indicator of the coupling of the surface layers with those above.

Comparison with emissions inventory: This section would be stronger with the inclusion of more details in the methodology to establish the how representative the data observed during the campaign are of annual conditions. See earlier suggestions.

In summary, the paper presents an original and interesting data set. When supported by a wider data set and an accompanying process based discussion this paper will be a useful contribution to the literature.