Review of "Megacity and local contributions to regional air pollution: An aircraft case study over London," Ashworth et al., ACP (2019)

## <u>Summary</u>

Ashworth et al. Present observations of atmospheric composition from a series of three flights conducted around London in July 2017. They apply a combination of trajectories and correlation analysis to illustrate the emission and transport of pollution from both London and other point sources in the area. The topic is appropriate for ACPD. The English is good. There are a few more figures than are necessary to convey the information.

This paper is largely a description of the observations, with some analysis of emissions that carries large caveats and uncertainties. These particular measurements have not been presented before, but there have been previous similar observations. It is not clear to the reviewer what new or novel things we have learned, or can learn, from this dataset. Some reorganization would help, as would focusing more on current open questions.

## Specific Comments

Use of WAS data for source identification: there are many examples in the literature of the use of specific hydrocarbon ratios to identify distinct emission sources. Why was this not done here?

Flight segments: It is helpful to number flight segments when comparing the map (Fig. 1) and time series (Figs 5 – 7). However, there are so many flight segments that the map looks cluttered and it is sometimes difficult to identify features discussed in the text. Rather than using one for every 5 minutes, how about 4 - 5 sections per flight?

L342 onward: discussion seems to indicate that the flux estimates shown in Table 3 are not comparable because of differences in the methodology and data used to do the calculation. Is this really the case, or is it just that the footprint is different? Also this makes Table 3 itself questionable, as it is comparing numbers that are not comparable. Statements regarding the comparison could also be more honest. For example, while it is true that the mean CO2 estimate is within 10% of O'Shea, the uncertainties are large. It would be better to state that the numbers agree to within combined uncertainties, which is something like 25 - 30%. This is also true for the abstract.

L420: This should not be surprising given that the observations were in the afternoon, when the diel cycle of ozone typically peaks.

L432: The discussion from here to the end of the section could benefit from improved organization. In particular, I recommend organizing paragraphs and order-of-presentation by plume rather than by chemical species. All paragraphs should have topic sentences. And, it might be clearest if the most likely source(s) are stated at the beginning, followed by evidence to support that identification.

L535: what is meant by "methods that can provide improved quantification of surface interaction"? Fluxes? Please be more specific.

## **Technical Comments**

The word "spike" is used throughout the manuscript to refer to features in the observed time series. In typical usage, this word refers to artifacts (e.g. due to electrical noise). Recommend replacing these words with "features" or "enhancements" or similar.

L219: "that WAS"

L245: "Fig. 4"

L294: which segments are the "background" ones?

- L322: "southwesterly wind"
- L403: replace "the profile of VOCs by altitude" with "the vertical profile of VOCs."
- L418: please quantify "far higher"
- L506: which numbered section is this in the time series/map?

L554: replace "known" with "assumed"

L556: "These three flights demonstrate"

L556: This data is not "remote sensing"

Figures in general: text is very hard to read in many cases. Too small. please fix.

Figure 2: line colors for GPH and coastlines are identical. Please change one.

Fig. 4: please mark London with a symbol. Also, do we need all 4 trajectories to get the point across?

Fig. 8 & 11: what is the triangle?

Fig. 9 & 12: Please flip so altitude is on the y-axis. Also, is the "total VOC" bar really that useful a metric?

Fig 14 & 15: Not sure we need both plots; just 15 would suffice. Also, blue color bar clashes with background.