

# 1 Response to the Reviewer's Comments

We thank the reviewer for their comments and below is a response to the reviewer's specific and technical comments and highlighted changes in the manuscript. We hope that these responses are satisfactory and that polishing the introduction by removing any repetition and adding further text to captions/discussion of the figures has addressed the points raised by the reviewer.

## Report 1

The authors improved the manuscript by 1. providing a more detailed introduction, 2. describing in more detail the calibration setup, and 3. extending the scientific analysis by looking into the benzene to toluene ratio. The introduction needs to be polished since the authors often repeat sentences. Regarding the new scientific analysis, it will be great if the authors clearly mention which datasets/drives are used for each of the graphs, specifically for Figure 8- Figure 11.

### Specific comments

- 1.1 Line 18-19: *PAN is also known to thermally decompose and form NO<sub>x</sub> that can affect O<sub>3</sub> production.*

Some text has been added that states this.

“and it has also been shown to thermally decompose to form NO<sub>x</sub>, which leads to enhanced O<sub>3</sub> production (Heald et al., 2003)”

- 1.2 Line 20-21: *You are repeating that VOCs have health impacts...*

This sentence has now been changed

“Also, some VOCs can cause...”

- 1.3 Line 84-86: *Repeating what is already mentioned*

Any repetitions have now been removed.

- 1.4 Line 224: *Are these box-and whiskers generated for all 30 drives? Please specify.*

Yes the box and whisker plots are generated from the data containing all of the 30 drives and the caption now states this.

“Summary of measurements made by a) the SIFT-MS (in ppb) and b) the UGGA (in PPM) during 30 repeat drives around York.”

- 1.5 Line 226-227: *What about isoprene?*

The measurement route around York does not contain any large urban green spaces and therefore we expect the mixing ratios to be relatively low. We agree with our statement that there is a 'lack of emission sources for these compounds in York'.

- 1.6 Line 230-231: *Ethanol could also originate from VCPs.*

A sentence has now been added stating this.

“Ethanol variation could also result from use of VCPs.”

- 1.7 Line 254: *Is this the most populated area of the town? Some discussion on the population density may be of interest here since it may indicate human sources.*

This area of the centre would not be expected to be the most populated area and it is dominated by businesses and shops. It is likely therefore that increases here are due to commercial premises.

- 1.8 Line 264-274: *Please discuss the expected high background concentrations of the non-vehicle related compounds. Could this play a role in their grouping performed?*

It is difficult to say whether the grouping is due to high background concentrations or similar emission sources. But, it is worth noting that the correlation coefficients are quite low ( $> 0.5$ ).

- 1.9 Line 282-284: *Would you expect emissions of benzene and toluene from other sources? What about cooking? Biomass burning? Are these sources expected during the period of the measurements?*

We would not expect biomass burning to be an important emission of benzene and toluene as the measurements were made during the Summertime, so burning in homes would not be taking place. Toluene and benzene correlate the best to tracers from gasoline fuel evaporation or engine exhaust emissions (as shown in the correlation matrix), so cooking is not expected to be an important source.

- 1.10 Figure 6: *You could add the LOD as a marker per compound in the graph.*

We feel that keeping Figure 6 as presented is preferable as adding a marker for LoD may make the plot less clear for the reader.

- 1.11 Figure 8: *Provide more explanations in the caption. What are the numbers and colors indicating? Also, please indicate what the lines on the right are in more detail.*

Suggestion implemented.

“A higher correlation coefficient between species is represented by a higher number, a darker red colour and an ellipses (shape). The lines on the right-hand side show the hierarchical clustering between compounds and represent clusters of species with similar patterns/behaviours.”

## Technical comments

- 1.12 Line 177: *Delete “significantly” since the stringency index is an indicator but doesn’t directly describe the pollution reduction.*

Suggestion implemented.

- 1.13 Line 253: *Replace “, this” to “that”*

This sentence has now been changed.

- 1.14 Line 266: *Change to “evaporation or engine exhaust”*

Suggestion implemented.

- 1.15 Line 252-253: *I would rephrase to “Ethanol is at background levels. . .”*

Suggestion implemented.

- 1.16 Line 280: *Repeating “many studies”.*

We have now removed ‘many studies’ from the previous sentence.

## 2 Response to the Reviewer's Comments

We thank the reviewer for their comments and below is a response to the reviewer's general comments, which addresses the points that they have highlighted. We have also provided responses to the specific comments and highlighted changes in the manuscript. We hope that these responses are satisfactory and that reworking section 3.4 and adding further text to captions/discussion of the figures has addressed the points raised by the reviewer.

### Report 2

Review of "Application of a mobile laboratory using a Selected-Ion Flow-Tube Mass Spectrometer (SIFT-MS) for characterization of volatile organic compounds and atmospheric trace gases" by Wagner et al., for publication in AMT

This study presents the application of a mobile analytical platform equipped with two analyzers: an Ultraportable Greenhouse Gas Analyzer (UGGA) and a Selected-Ion Flow-Tube Mass Spectrometer (SIFT-MS) measuring carbon dioxide, methane, several VOCs and other trace gases. This mobile platform was deployed in the city of York, UK, where it completed a total of 31 surveys of the same route over a 10-day period.

Upon re-reviewing the article, I really appreciate that the authors made a thorough effort to address the several comments received during the first review process. The authors had particular care in further analyzing their measurements and developing the results section. Indeed, they added a section where they investigated the toluene to benzene ratio of their measurements to differentiate between emissions from vehicle exhausts and evaporative emissions from fuels and solvents. Even if I find this added section a bit confusing (see comments below) and think it should be reworked a bit, I am satisfied with the other edits and responses to most review comments. Overall, this manuscript should be published subject to these minor corrections:

- 1.17 : *There are many acronyms in this article (especially in the first two sections), some of them are defined once but never used after (CAFO) or only used once (PAN, ARI), some are defined several times (UGGA), some are not defined at all (PTR-TOF-MS). I would recommend simplifying all of these notations by getting rid of the ones that are almost never used. For PTR-TOF-MS, I would describe this technique, explain the difference with regular PTR-MS and indicate right away that it will be referred as PTR-TOF in the rest of the article.*

We have simplified and removed notations throughout the text. We have also added some text describing PTR-MS and PTR-TOF-MS and the differences.

*"A PTR-MS is a term used for an instrument which consists of an ion source that is directly connected to a drift tube and a mass analyzing system, which either consists of a quadrupole or time of flight mass analyser. Standard PTR-MS instruments are a PTR-QMS (Proton Transfer Reaction Quadrupole Mass Spectrometer), which can detect and resolve product ion masses at single unit mass resolution. ... A PTR-TOF can detect and resolve product ions at much higher mass resolution with currently available commercial instruments having a mass resolution  $\geq 4000$ ."*

- 1.18 *I would be consistent with the introduction of the different species' chemical formulas: some species have their chemical formula introduced properly (O<sub>3</sub>), other formulas are used in the introduction and introduced in section 2 (CH<sub>4</sub>, CO<sub>2</sub>), other formulas are simply never introduced (NO<sub>2</sub>, HONO, NH<sub>3</sub>...).*

Suggestion implemented.

- 1.19 *Section 3.4 is a bit confusing to me. In this section, the authors describe how the T/B ratio can be used to differentiate emissions from vehicle exhausts and evaporative emissions from fuels and solvents, and explain how both of these sources can be transient. I am not sure I understand what do the authors mean L294-296: "Taking the median value of concentrations will down-weight the contribution from less frequent, higher concentrations as used by Apte et al. (2017). However, there is potentially important information that can be missed when only considering the median",*

are they talking about using the median value of toluene and benzene in each 30 metre segment of road to calculate the T/B ratios rather than one of the regressions described later? Or using an ordinary least square regression? I think the authors should guide the reader by introducing the 2 possible approaches to estimate the T/B ratio (OLS and quantile regression), and then explain their advantages and drawbacks (transient emissions...). In the end, I am not sure the T/B ratio example on Hull Road is the best one to illustrate the advantages of the quantile regression over the OLS: in both case these ratios are over 2 which indicates that the main source of the emissions is from fuels and solvents. Overall, this section should be reworked a little bit to make it easier for the reader to follow.

Line 294-296 is discussing the median ratio used for spatial mapping, which has now been stated in the text. The T/B ratio plot shown in Figure 9 shows the OLS regression slope obtained from the data from all 30 repeat drives around York, but each point represents a 30 metre segment of road. We have reworked this section but have kept the same structure, we hope that it now reads more clearly. We feel that Hull road is a good example to show the differences between OLS and quantile regression as there is a clear difference in the T/B ratio seen at higher percentiles compared with the 50th percentile. The two regression approaches may show some similarities, but the quantile regression used along a gaussian kernel smoother ensures that transient emissions are fully captured.

1.20 Figure 10: *I suggest to improve the title of this figure or at least remove "A plot of...".*

Suggestion implemented.

1.21 Figure 11: *I would add the slope values for the different regressions, I find it relatively difficult to compare "Evaporative" and "Remainder" as it is currently.*

Suggestion implemented.

1.22 Line 338: *I would remove "We have demonstrated the correlation method". The authors did observe correlations between species but the method does not need a demonstration.*

Suggestion implemented.

## References

Colette L. Heald, Daniel J. Jacob, Arlene M. Fiore, Louisa K. Emmons, John C. Gille, Merritt N. Deeter, Juying Warner, David P. Edwards, James H. Crawford, Amy J. Hamlin, Glen W. Sachse, Edward V. Browell, Melody A. Avery, Stephanie A. Vay, David J. Westberg, Donald R. Blake, Hanwant B. Singh, Scott T. Sandholm, Robert W. Talbot, and Henry E. Fuelberg. Asian outflow and trans-Pacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft, and model perspective. *Journal of Geophysical Research: Atmospheres*, 108(D24):4804, 12 2003. ISSN 2156-2202. doi: 10.1029/2003JD003507. URL <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2003JD003507><https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2003JD003507><https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2003JD003507>.