# 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 major points that they have highlighted. We have also provided responses to the specific comments and highlighted changes in the manuscript.
We have now included a comparison of SIFT-MS to PTR-MS in the introduction and highlighted SIFT-MS as an easier to use instrument that could be used by non-research organisations. Importantly SIFT-MS benefits from the use of three reagent ions, which can be switched in real-time, therefore allowing for measurements of a greater variety of species and separation of complex mixtures in atmospheric air. We thank the reviewer for highlighting mobile measurement studies carried out using a PTR-TOF-MS and we have now included these in our introduction.
We thank the reviewer for suggestions of scientific analysis that we could include in the paper. In the paper we have now expanded on the results and analysis section by developing a new method that is complementary to the existing analysis in the paper. This analysis is now in the paper as an additional section (Section 3.4) and involves investigating the toluene to benzene (T/B) ratio using quantile regression modelling. This has been used alongside a gaussian kernel smoother to show the T/B ratio along the distance of a road of interest to reveal intermittent sources that would be missed through only spatially averaging compounds.
We hope that these responses are satisfactory and that incorporation of a further discussion of the differences of SIFT-MS compared to PTR-MS and further development of the results section has addressed the points raised by the reviewer.

# Reviewer 1

The authors present measurements using a mobile laboratory equipped with a Selected-Ion Flow-Tube Mass Spectrometer (SIFT-MS) to measure 13 volatile organic compounds (VOCs) including aromatics, monoterpenes, nitrogen oxide, and more abundant species like methanol, ethanol, and acetone together with other instrumentation to measure CO2 and CH4. They provide details on the mobile laboratory setup including the online calibration of compounds. A set of measurements is presented where they perform a correlation analysis of all compounds and hierarchical clustering.

My main concern is that this study is not presenting a new method that isn't already published by previous work. Although the SIFT-MS is a great addition to the mobile lab such measurements have been performed before by higher-resolution instruments. An example is the proton transfer reaction time of flight mass spectrometer (PTR-ToF-MS) measuring hundreds of VOCs at 1-sec resolution that has been extensively used by the NOAA team but not referenced at all in the introduction. Recent example publications are from Coggon et al. (2016); Yuan et al. (2017); Coggon et al. (2018); Shah et al. (2020); Gkatzelis et al. (2021a); Gkatzelis et al. (2021b); Stockwell et al. (2020). TOFWERK has also been using the VOCUS for real-time measurements on their mobile lab in Europe (`https://www.tofwerk.com/vocus-ptr-mobile-laboratory-video/`), Aerodyne in the US e.g. for profiling natural gas production (`https://www.tofwerk.com/oil-and-gas-well-emissions/`), and the same type of measurements have been performed by Montrose (`https://montrose-env.com/services/testing-lab-services/ptr-tof-ms-mobile-laboratory/`), and the RJ Lee group (e.g. `https://rjlg.com/2019/06/philadelphia-refinery-explosion-air-quality-impact/`). Furthermore, a characteristic paper similarly discussing this method was published last year by Richards et al. (2020) on mobile measurements of VOCs using a PTR-ToF-MS and a membrane introduction mass spectrometry (MIMS) where they also perform more detailed Principal Component Analysis (PCA) to source apportion VOCs. This literature can be easily found by just googling "mobile laboratory measurements of VOCs" so I am surprised it is currently not covered by the authors. I am therefore not convinced that this is truly a new method. I could see how this work could be published as a complementary, possibly cheaper (?) way to perform such measurements. Of course, I leave this to the editor to decide. Nevertheless, carefully promoting all the existing work and the benefits and advantages of other studies compared to this one will be crucial, something the paper is currently lacking.

Regarding the scientific analysis of the derived data I find it to be limited to just correlations that are not fully discussed. A benefit I see from the SIFT-MS is that it can measure VOCs but also NO2. The slopes of the correlations of VOCs to NO2 could provide more insights into the pollution source. This correlation analysis would also be more informative when performed at higher resolution as a rolling correlation function. Analysis to prove whether the measurements obtained here are influenced by traffic should focus on comparisons to previous literature. What are the obtained slopes from the correlations for every few minutes of data and how do they compare to a vast literature of traffic emissions? These correlations could be done both for VOCs vs. NO2 but also VOCs vs. CO2. Also, I would expect enhancements of emission when moving to the city center. Did the authors observe that as has been seen before by various other groups (e.g. `https://www.fz-juelich.de/iek/iek-8/EN/Expertise/Infrastructure/MobiLab/MobiLab_node.html`)? Some timeseries plots or enhancement box-and-whiskers would be great additions here. Furthermore, the authors at points of the manuscript discuss the influence of other sources like paints and in general volatile chemical products. It would be great to compare their results to inventory estimates by McDonald et al. (2018) and paint studies e.g. Stockwell et al. (2020). While this is a measurement technique paper I do consider that science that validates the importance of performing such measurements should be included and in my opinion is currently not sufficient. Statistical methods to separate to different sources would also be valuable e.g. the use of positive matrix factorization. If this is not an option then I would consider discussing in detail the benefits of performing such statistical approaches in the future.

Overall, rewriting the introduction, focusing on what is new from these measurements, discussing the benefits compared to other published work as well as the disadvantages, and performing further analysis of the obtained data would be the main improvements before this publication is suited for AMT. Below some additional specific comments.

1.1 *Acetone, methanol, and ethanol are abundant species coming from multiple sources including atmospheric chemistry. This may drive the correlations observed here. Slopes would be interesting to have and compare to other studies. More detailed timeseries of compounds for specific events would be helpful to further conclude on possible sources.*

    Since publishing the paper, we have applied a compound specific zero offset to the measurements for each individual day. When re-plotting the correlation plot, the correlations between these species was not as strong as it was previously, so no further analysis has been carried out into these species.

1.2 *Table 4 should be a box and whiskers figure with the y-axis representing the compound names and the x-axis the concentrations. In-city and out-of-city box and whiskers would be valuable and provide urban enhancements during the COVID-19 pandemic.*

    Table 4 has now been changed into a box and whisker plot.

1.3 *Also, a more detailed discussion on the lockdown conditions during the period of the measurements would be great. A suggested reference could be the stringency index `https://ourworldindata.org/grapher/covid-stringency-index`).*

    We thank the author for the suggested reference and a sentence has been added which comments on the lockdown conditions.

    " It should be noted that during the measurement period the Covid-19 stringency index was 64.35 (taken from: `https://ourworldindata.org/grapher/covid-stringency-index?tab=chart&country=~GBR`), indicating significantly reduced economic and traffic activity. This could affect both the concentration and detection of different VOC species due to possible decreased emissions. "

1.4 Line 10: *Correct to "sources".*

    Suggestion implemented.

1.5 Lines 22-32: *The authors could further discuss here VOC emissions from other pollution sources in more detail. Volatile chemical products, cooking emissions, residential wood burning, and industry with their respective citations would be of value here. Also, studies that focus on the contribution of different sectors of VOC emissions would be valuable too.*

    A discussion of the contribution to VOC emissions from different emissions sources has been added to the introduction.

"Mass balance analysis by McDonald et al. (2018) concluded that emissions from VCPs now account for half of the fossil fuel VOC emissions in industrialised cities. Lewis et al. (2020) estimated that in the UK, VOC emissions from solvent use and industrial processes account for 63% of total VOC emissions."

1.6 Line 29: *Add references. Examples for volatile chemical products could be McDonald et al. (2018), Stockwell et al. (2020), Gkatzelis et al. (2021a,b).*

We have added the reference from McDonald et al. (2018) (as shown above) when discussing the contribution to VOC emissions. We have also added additional VOC sources in the text, but did not feel including references for all of the different emissions sources was necessary. We have instead included these references further along on the introduction discussing the use of PTR-MS in mobile laboratories to identify emissions sources.

"Nevertheless, the reduction in vehicular emissions means it is likely that other sources of emissions, such as volatile chemical products (VCPs), solvent use, cooking, residential wood burning and industry, have become more important."

1.7 Lines 57-58: *Many recent studies use PTR-ToF-MS in mobile laboratories that are currently not discussed. See comments above.*

This section has been re-written to include studies that use PTR-ToF-MS in mobile laboratories. We thank the reviewer for a comprehensive list of references.

1.8 Line 123: *Correct to "formulas".*

Suggestion implemented.

1.9 Line 161: *delete "instrument"*

Suggestion implemented.

## References

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