

## **Responses to the reviewers**

We express our gratitude to both reviewers for their comments, as well as their critical remarks, which have helped us make this paper more organized and scientifically robust. We authors let the reviewer know that we got some feedback from the handling editor who asked us to submit this paper as the category of “measurement report” instead of “research articles”. Following the editor’s suggestion, we submitted this manuscript as the category of “measurement report” (please refer to the title, “Measurement report: Summertime and wintertime VOCs in Houston: Source apportionment and spatial distribution of source origins”) and thus, this manuscript includes measurement results, but where the implications of atmospheric chemistry and physics are less developed in this manuscript (refer to ACP manuscript types).

### **Anonymous Referee #2:**

#### **General comments:**

**This manuscript addressed source apportionment of VOCs in Houston, a petrochemistry condensed region in US, by using measurement data in 2018. It reads a quite routine source apportionment report, I regret to suggest a rejection of current version due to three points:**

- 1. The current version discussed only the data obtain in their monitoring site, with some discussion on roles of VOCs chemsity and transportation. I consider that an evaluation on trends of VOCs levels, chemical compositions and source, or features of source chenges for petrochemical industries in US ( Houston as an example) would be of more interest to community rather than a local study.**

Yes. That was a critical comment the reviewer mentioned. The authors clarified the motivations of this study to help the readers what encouraged us to conduct this research. We do think earlier studies showed the emissions of the VOCs over the Houston industrial region (Buzcu and Fraser, 2006; Leuchner and Rappenglueck, 2010). However, there are still some factors needed to be taken into account for an enhanced understanding of the VOCs of Houston. The variation patterns of VOCs depend on anthropogenic emissions, photochemical reactivity, and meteorology. Previous

studies showed that the emissions of VOCs in Houston are mainly from the industrial sectors of the Ship Channel, but the short period of campaign measurements limited those studies' ability to explore the seasonal variations of the anthropogenic and biogenic source emissions. We also looked (Na and Kim, 2001) and their investigation overlooked the variability of the VOCs concentrations and their photochemical reactivity over Houston (An et al., 2014; Guo et al., 2014; Pan et al., 2015; Baudic et al., 2016). In addition, these studies lacked the consideration of the different formation potentials of speciated VOC, which is necessary to address the causes of ozone episodes and mitigate urban air pollution.

**2. The methods used in the MS, PCA, PMF, OFP, ratio analysis and backward trajectory are sort of routine. And the dataset is for 2018, the reviewer didnot see measurements on OVOCs which cuold be important for petrochemical emissions. Therefore from methodological perspective, I didnot found see something new for VOCs source understanding.**

This study has tried to explore the seasonal characteristics of VOCs measurements, their emissions and contributions to ozone formation, and geographical locations in the industrial area of a metropolitan after the last campaign of VOCs characteristics in summer 2006. We used the available data of VOCs measurements in summer and winter and followed the editor's suggestion as to classify this manuscript as a "measurement report" instead "research articles". However, as we can do, we significantly modified and added many subsections to satisfy the need to explore the scientific points (please also refer to our responses to reviewer 1 and the revised manuscript with colored highlights). We hope the revised draft of this manuscript clearly clarifies the findings of our study.

**3. There is an important issue to discuss with the authors. The authors showed quite string chemical oxidation processes by using ratios of VOCs, e.g. Benzene/Toluene, and etc, this clearly means that conventional PCA and PMF could not be deployed for source apportionment in Houston, the authors needs to use chemical-loss correction to do reliable source apportionment. This problem is not discussed in the current version.**

The authors would not argue on capabilities of the PMF model since it is a well-regarded source apportionment technique that provides supportive and reliable information that enhances our understanding of the atmospheric chemistry of organic compounds in numerous recent studies

(e.g., Baudic et al., 2016; Buzcu-Guven and Fraser, 2008; Buzcu and Fraser, 2006; Koss et al., 2020; Leuchner and Rappenglueck, 2010; Li et al., 2019; Liu et al., 2020; Pernov et al., 2021; Pollack et al., 2021; Sinha and Sinha, 2019; Song et al., 2019; Verreyken et al., 2021).

However, we agree with the point the reviewer mentioned and are aware of the fact that the PMF algorithm treats the measured organic compounds as inert species, which addresses some limitations to the interpretation of the results. Hence, as the reviewer suggested, we discussed in the manuscript that processes such as mass production and atmospheric removal could cause a single emission source to appear as multiple factors or causing oxidized species from multiple emission sources to be grouped together and increase the uncertainty of the resolved source profiles (Sauvage et al., 2009; Wang et al., 2013; Yuan et al., 2012).

Thank you again for your detailed remarks; we have adapted the manuscript as suggested.

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