Response to Reviewer #2

In this manuscript, the authors present aircraft-based vertical observations of volatile organic compounds (VOCs) measured using high-time resolution mass spectrometry. Despite the technical challenges associated with such data collection, the authors provide valuable insights into the complex evolution of VOC species under the combined effects of surface emissions, chemical removal, and regional transport. The topic is certainly of interest, but the analysis does not yet sufficiently support the proposed conclusions. The existing data requires further exploration, and additional information is needed to strengthen the findings. There are also several aspects that require clarification. Below are my specific comments and suggestions.

Reply: We thank you for all the valuable comments. Each comment has been addressed in detail below and we believe the manuscript has been strengthened after the revision. We hope our replies are to your satisfaction.

Specific Comments:

1.My primary concern is that the current analysis does not robustly support the statement about the "complex VOC species evolution under the joint impacts of surface emission, chemical removal, and regional transport."

1.1 Features of VOC species profiles:

The VOC species selected for the study are appropriate, but a more thorough classification and combination would enhance the analysis. For instance, methanol and acetonitrile are largely chemically inactive and primarily derived from primary emissions. In contrast, acetaldehyde is active and predominantly the result of secondary chemical transformations.

To better address the relationship between VOC species profiles, emissions, and chemical removal, I recommend replotting Figures 4, 6, 8, and 9 to incorporate the ratios of VOC species based on their chemical reactivity (see Zhu et al., 2025) or the ratios of primary vs. secondary species (see Yang et al., 2024). Including CO and NOx profiles would also provide useful tracers for distinguishing between inactive and active species, provided the data are available.

Reply: We thank you for your suggestions. Unfortunately, there was no CO or NOx data available during the aircraft measurements. In order to incorporate the ratio analysis between primary species and secondary species as you suggested, we added an extra vertical profile of C8 aromatics-to-acetone concentration ratio in Figure 4 to address the chemical removal effect.

C8 aromatics represent primary VOC species with higher chemical reactivities that undergo fast removal in the atmosphere, while acetone represents VOC species with lower chemical reactivities and secondary formation pathways. Discussions for the vertical profile of C8 aromatics and acetone have been added in the revised manuscript (Lines 262-269):

"*The ratio between VOC species is commonly applied to address the impact of chemical removal and secondary formation during transport (Yang et al., 2024b;Zhu et al., 2025). The vertical profile of the C8 aromatics-to-acetone concentration ratio was plotted in **Figure 4** to demonstrate these effects. Both species can be emitted by vehicular and industrial emissions (Jiang et al., 2023;Wang et al., 2024), but C8 aromatics are more reactive, and acetone can be formed from secondary processes. The two effects both lead to a rapid decrease in the concentration ratio of C8 aromatics to acetone within the PBL before stabilizing as expected.*"

The purpose of Figure 6 is to compare the vertical profiles between Beijing and Baoding, and Figures 8 and 9 serve for discussions about anomalous enhancements. Thus, the proposed additional plot would not be relevant to these three figures. We would like to leave these figures unrevised. However, we have added additional discussions on the different reactivities between styrene and benzene, and the influences on the analysis in Figure 8 in Lines 360-365:

"*Since styrene is more chemically reactive than benzene and thus the lifetime of styrene is much shorter than that of benzene. The ratio of styrene to benzene would decrease during transport. As shown in Figure 8d, the enhancement ratios at higher altitudes still fall within the characteristic ranges of industrial sources and are significantly larger than those of vehicular emissions. Thus, the chemical influences do not change our conclusion here*".

The revised Fig. 4:

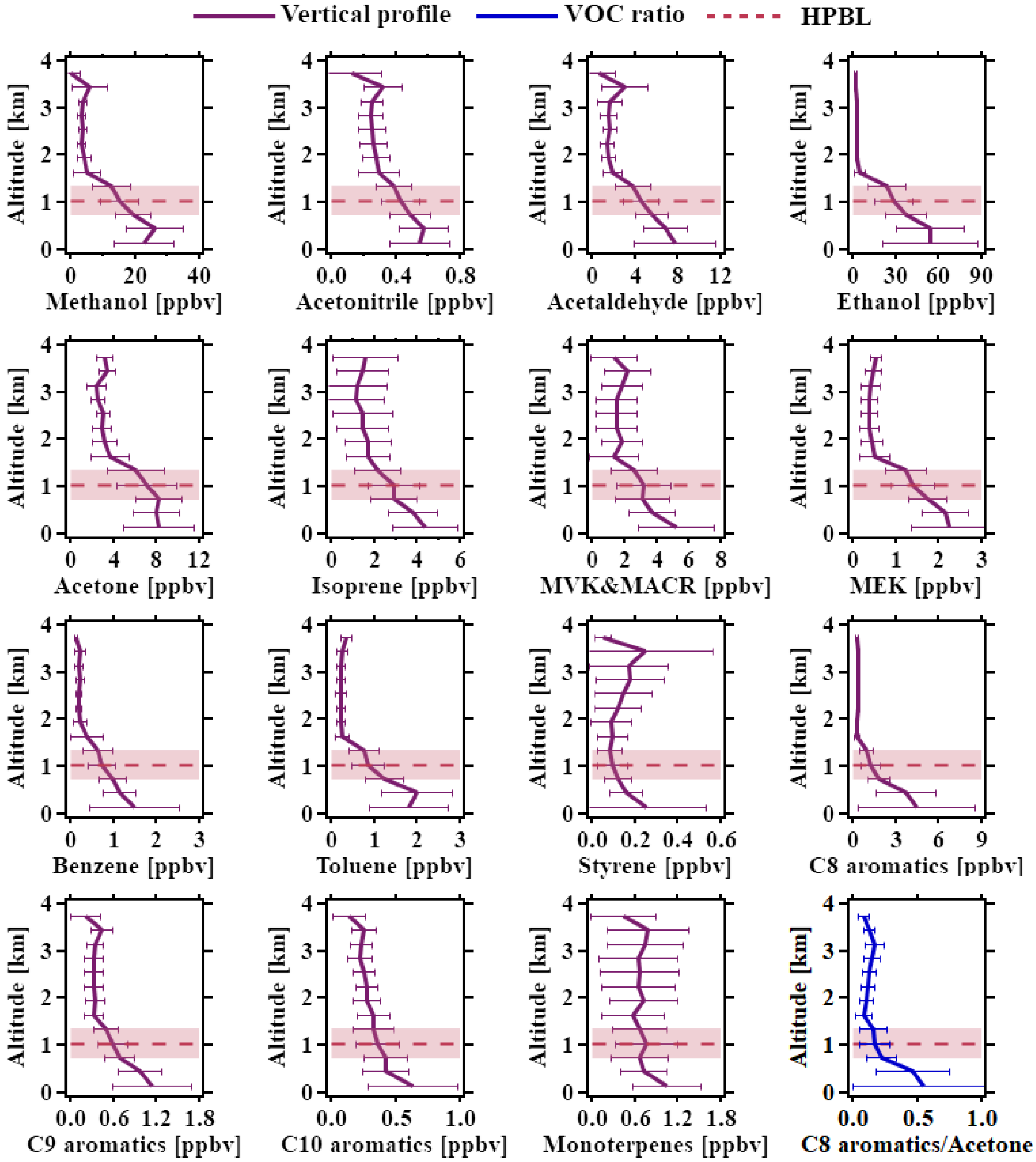

Figure 4. *Averaged vertical profile (purple lines) of VOCs in five aerial surveys above the Beijing area in Sep. 2017 with error bar. The blue line shows the average vertical profile of C8 aromatics-to-acetone concentration ratio with error bar. The red dashed line is the average of the HPBL, with the light red area showing the variation range of one standard deviation.*

1.2 Regional Transport of VOCs**:**

For the analysis of VOC regional transport, I suggest incorporating synoptic charts and diagnosing vertical velocity (including vertical transport and advection). In lines 192 and 222, the CO concentrations at ~3500 m and 2000 m could serve as effective tracers for regional transport.

Overall, the manuscript would benefit from better organization and additional data analysis to substantiate the points raised by the authors.

Reply: Thank you for your valuable suggestion. We added Figure S9 to show the wind field at 850 hPa during the periods with anomalous enhancements on Sep. 9th, 2017 and Jul. 14th, 2019. We also added additional evidence from the wind field plot to support the analysis in section 3.3 (Lines 358-360, Lines 389-392 in revised manuscript).

"*The synoptic chart in* ***Figure S9a*** *shows strong northerly winds at the 850 hPa level over the aircraft survey area, suggesting that the industrial emissions were likely transported from the north.*"

"*As shown in* ***Figure 9b****, winds were quite weak at 850 hPa in the aircraft survey area, suggesting that the elevated VOC concentrations were likely attributed to localized biomass burning emissions.*"

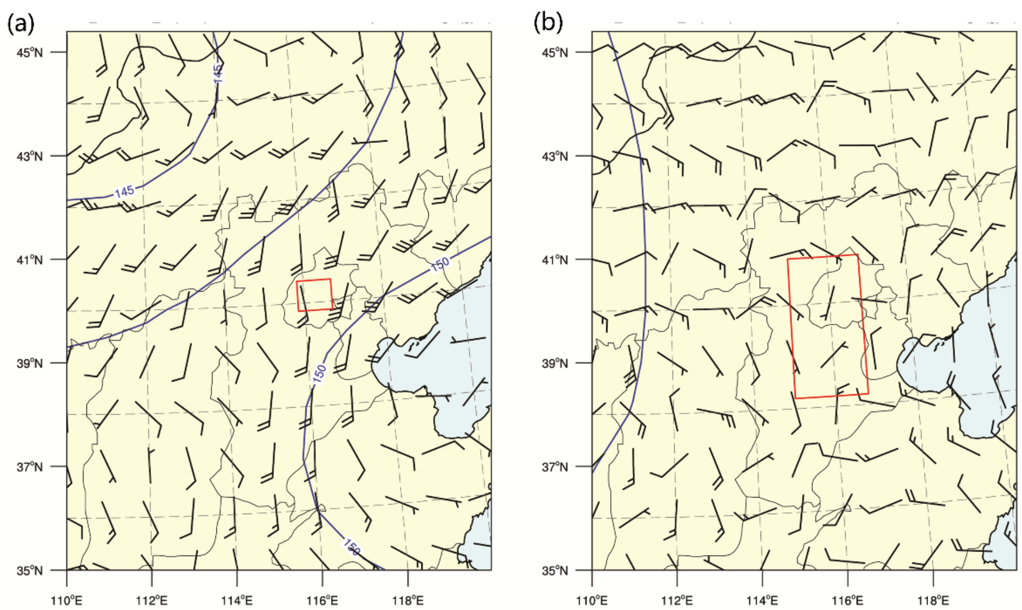

Figure S9*. The synoptic charts for the flight measurements on Sep. 9th, 2017 (a) and Jul. 14th, 2019 (b). The wind field at 850 hpa was plotted. The red box indicates the area covering the trajectory of each flight as shown in Figure 1 (Route 1 and Route 6). The meteorology data were downloaded from the Global Forecast System* *(https://www.ncei.noaa.gov/products/weather-climate-models/global-forecast).*

2. In lines 129-132, the criteria for determining the HPBL are necessary to include, but the equation for potential temperature seems unnecessary in this context.

Reply: We thank you for your suggestion. The equation for potential temperature has been removed, and we have added additional descriptions about how HPBL was determined in the revised manuscript (Lines 148-150):

"T_θ first decreases with height to a minimum and then increases. The HPBL is determined as the height where T_θ returns to its surface value. "

3.The analysis presented in lines 230-243 is intriguing. The highest ratio of ethanol between below and above the boundary layer (BL), compared to acetaldehyde, acetone, and MEK, may reflect the more uniform vertical distribution of secondary VOCs (OVOCs) from various chemical sources. However, if the ratios of active species (e.g., styrene and monoterpenes) to inactive species (e.g., toluene and benzene) were compared, a different ratio feature of OVOCs/ethanol might emerge. These features could provide further meaningful insights that are directly related to the "complex VOC species evolution under the joint impacts of surface emission, chemical removal, and regional transport."

Reply: Thank you so much for your constructive suggestions. We have added additional discussions about the ratios of reactive species to inert species and revised the related sentences in the revised manuscript (Lines 283-290):

"*C8 aromatics are quite chemically reactive, so a higher ratio suggests strong chemical removal, while for less reactive species, such as acetone, its ratio is closer to 1, indicating a weak impact of chemical reactions and potential contribution of secondary formation. Notably, the data points of styrene are clustered near the 1:1 ratio line, and occasionally, the concentrations above the PBL could be higher than those within the PBL. Since styrene is a primary species emitted at the surface, this pattern suggests a great contribution from transport, which will be discussed in* ***Section 3.3***."