Responses to the comments of Reviewer #1:

We would like to thank the anonymous referee for his/her comprehensive review and valuable suggestions. These suggestions help us to present our results more clearly. In response, we have made changes according to the referee's suggestions and replied to all comments point by point.

1. the conclusion about MEGAN overestimates NMVOC emissions seems to be case-specific, and in the revision the authors emphasize that it is due to severe heatwave conditions. So I would suggest that 'August 2022' should be clearly stated in the title and the abstract. In the discussion, it is also suggested to mention that the conclusion is for august 2022 case.

Response: Thanks for your suggestions. We have added "August 2022" to the title and modified the title to " Constraining Non-Methane VOC Emissions with TROPOMI HCHO observations: Impact on Summertime Ozone Simulation in August 2022 in China".

We also added corresponding statements to the abstract and conclusion

See line 33, page 2.

"Here, we extended the Regional multi-Air Pollutant Assimilation System (RAPAS) with the EnKF algorithm to optimize NMVOC emissions in China in August 2022 by assimilating TROPOMI HCHO retrievals."

See lines 38-41, page 2.

"NMVOC emissions exhibited a substantial reduction of 50.2%, especially in the middle and lower reaches of the Yangtze River, revealing a prior overestimation of biogenic NMVOC emissions due to extreme heatwave."

See line 592, page 26.

"The application of TROPOMI HCHO observations as constraints led to a substantial

reduction of 50.2% compared to the prior emissions for NMVOCs in August 2022."

2. The data of Figure 5 should be more clearly stated in the text. Average across how many sites? It would be good to illustrate in a few key sites/cities and put in SI, so that the readers can get more detailed information.

Response: Thanks for your suggestions. We have provided detailed statistics and the number of sites in the text. Additionally, we conducted statistics on seven key cities selected from the seven major regions constituting China. The detailed changes are shown below:

See lines 457-462, pages 18-19.

"Constraining the NMVOC emissions also led to better model simulations in terms of RMSE throughout the entire study period. Time-averaged BIAS and RMSE decreased from 20.6 and 37.3 μg m^{-3} to 10.6 and 31.0 μg m^{-3}, respectively. We also evaluated the simulation results for seven key cities (i.e., Beijing, Shanghai, Guangzhou, Wuhan, Chongqing, Yinchuan, and Changchun, which represent key cities in North, East, South, Central, Southwest, Northwest, and Northeast China, respectively), and the biases in the VEP with posterior emissions all showed a significant reduction (Figure S8). Overall, the assimilation of HCHO column observations effectively reduced NMVOC emission uncertainties and consequently improved simulations of HCHO and O_3."

See lines 470-472, page 20.

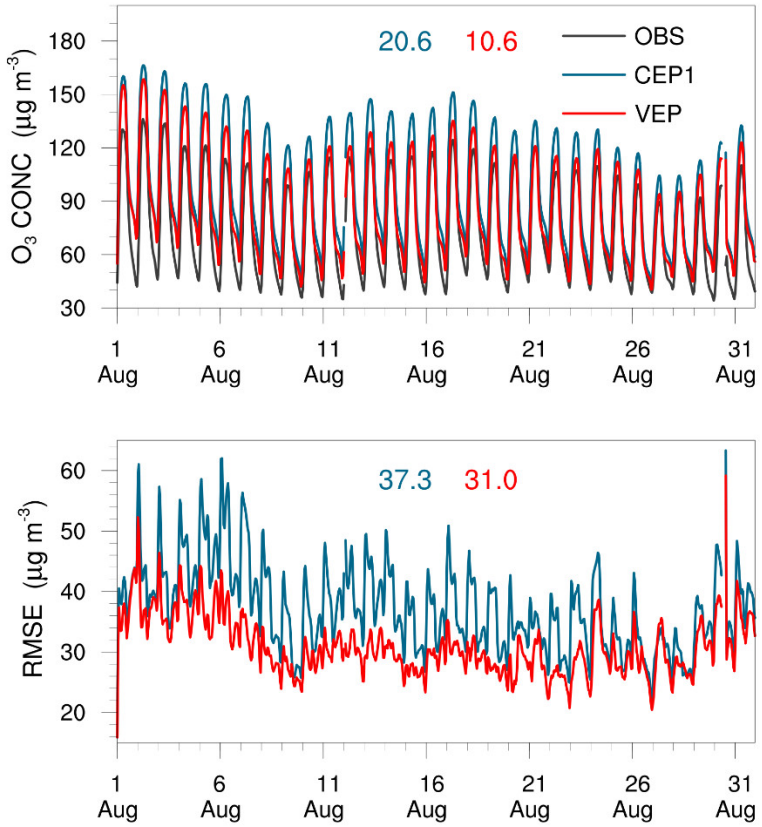

Figure 5. Time series comparison of hourly surface O_3 concentrations (μg m^{-3}) and RMSE (μg m^{-3}) from CEP1 and VEP experiments against all observations at 1701 monitoring sites. The blue and red values on the graph represent the time-averaged statistics in the CEP1 and VEP experiments, respectively.

See Figure S8 in the Supplementary Information.

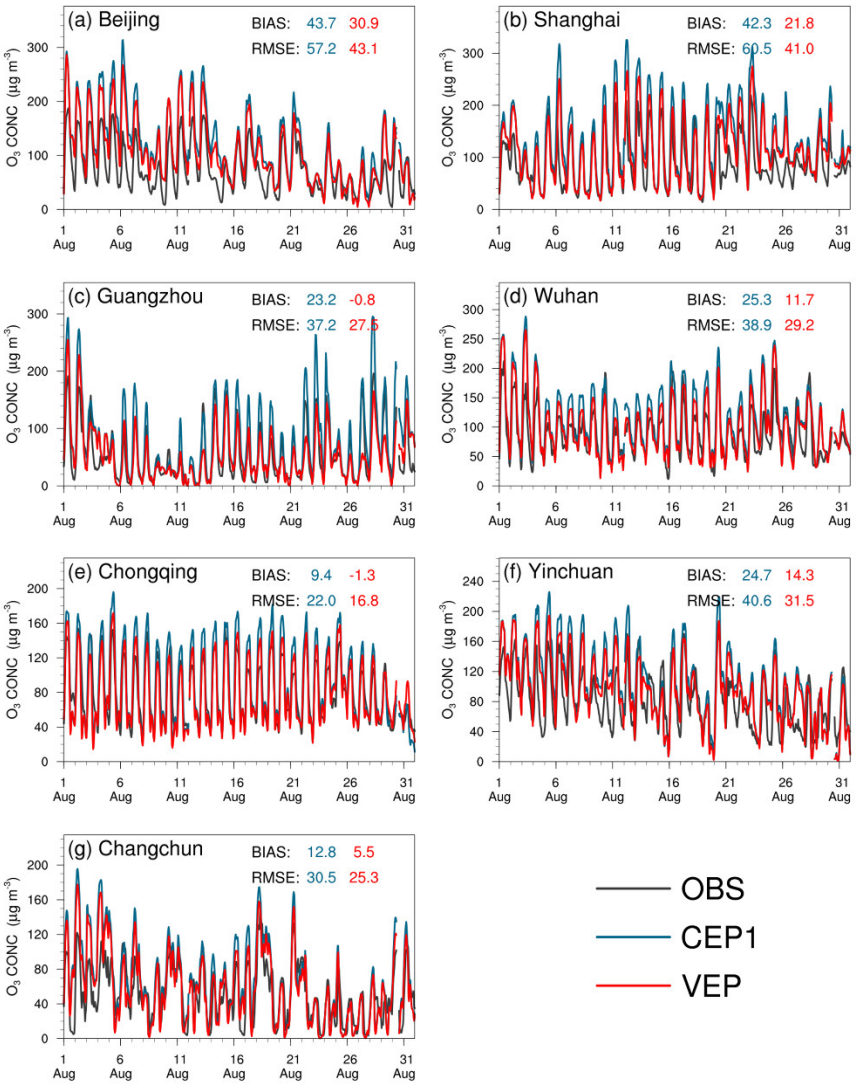

Figure S8. Time series comparison of simulated and observed hourly surface O_3 concentrations (μg m^{-3}) from CEP1 and VEP experiments over (a) Beijing, (b) Shanghai, (c) Guangzhou, (d) Wuhan, (e) Chongqing, (f) Yinchuan, and (g) Changchun, representing key cities in North China, East China, South China, Central China, Southwest China, Northwest China, and Northeast China, respectively.

3. Espeically in urban areas, where anthropogenic emissions become dominant for VOCs, are the anthropogenic NMVOCs emissions also overestimated? How is it compared to rural areas where biogenic emissions dominate?

Response: Thanks for this comment. Accurately distinguishing between biogenic and anthropogenic NMVOC emissions remains challenging. This difficulty arises because the same model grid contains both biogenic and anthropogenic emissions, and observations cannot easily differentiate the mixed signals in the atmosphere. In this study, we derived the posterior anthropogenic emissions based on the prior emission ratio information and the total posterior emissions (Figure R1). It can be observed that in most regions of the central and eastern parts of China, especially in areas where anthropogenic sources absolutely dominate the total emissions, such as Beijing-Tianjin-Hebei, the Yangtze River Delta, and the Pearl River Delta, there is a significant decrease in posterior NMVOC emissions. The overall NMVOC emissions in China have decreased by 43.4%, but this is within the uncertainty range of MEIC (±68-78%) (Li et al., 2019). Additionally, the overestimation of anthropogenic emissions in urban areas is less than that of biogenic emissions in rural areas (53.7%).

We have added following descriptions in the revised manuscript. See lines 371-374, page 13.

"Ultimately, the biogenic NMVOC emissions decreased by 53.7%, which was higher than the 43.4% decrease in anthropogenic NMVOC emissions (Figure S3). Overall, the large magnitude of emission decrease of 50.2% in our inversion is comparable to studies … …"

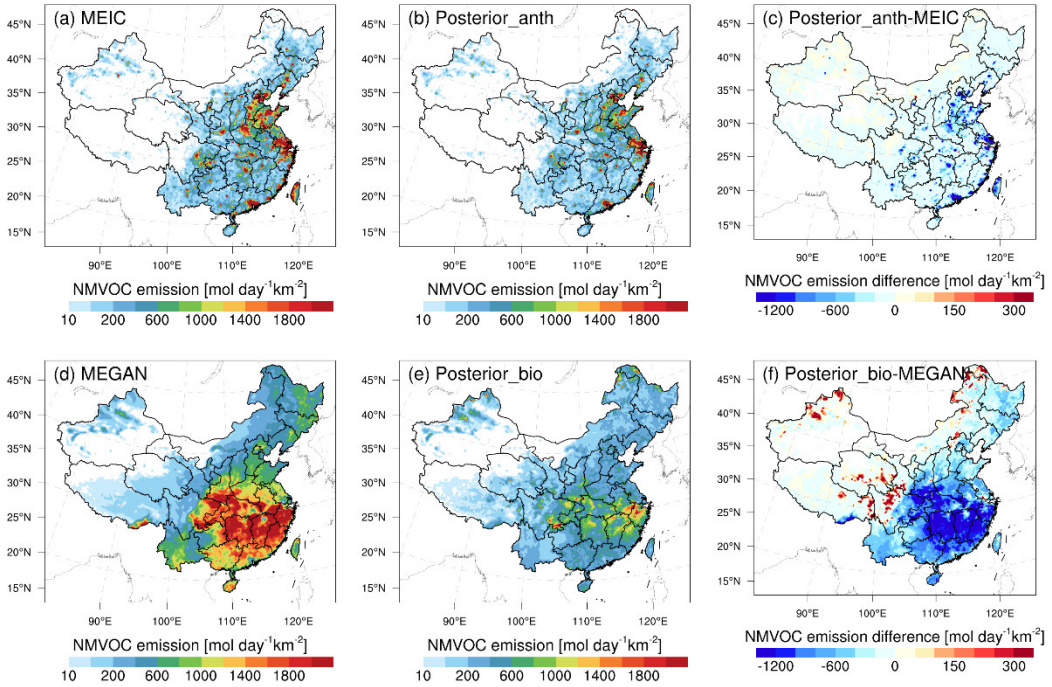

Figure R1. Spatial distribution of the time-averaged (a) prior anthropogenic emissions (MEIC 2020), (b) posterior anthropogenic emissions, (c) difference between prior and posterior anthropogenic emissions (posterior minus prior), (d) prior biogenic emissions (MEGAN), (e) posterior biogenic emissions, (f) difference between prior and posterior biogenic emissions (posterior minus prior) of NMVOCs over China. (Figure S3 in the Supplementary Information)

Li, M., Zhang, Q., Zheng, B., Tong, D., Lei, Y., Liu, F., Hong, C., Kang, S., Yan, L., Zhang, Y., Bo, Y., Su, H., Cheng, Y., and He, K.: Persistent growth of anthropogenic non-methane volatile organic compound (NMVOC) emissions in China during 1990–2017: drivers, speciation and ozone formation potential, Atmos. Chem. Phys., 19, 8897-8913, 10.5194/acp-19-8897-2019, 2019.