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. All the page and line number for corrections are referred to the revised manuscript, while the page and line number from original reviews are kept intact.

1. Accurate NMVOC emissions are essential for predicting air quality. Currently large uncertainties exist in NMVOC emissions, both in the anthropogenic and biogenic sources, as compared to other pollutants, such as SO2 and PM. In this study, the authors use the RAPAS assimilation system incorporated with the EnKF assimilation algorithm to optimize NMVOC emissions using TROPOMI HCHO retrievals. They use MEIC 2020 for anthropogenic emissions and MEGANv2.1 for biogenic sources as the priori NMVOC emissions. They find that NMVOC emissions are largely overestimated, especially biogenic NMVOC emissions. They also find O_3 predictions would be lowered using the posterior NMVOC emissions.

The study seems interesting, however, I have a few concerns about some of the key results in this study.

First, the CMAQ model overpredicts O_3 in China largely (over 20 ug/m^3) in most sites of China (Figure 4a) with the WRF-MEIC-MEGAN setups. The overpredictions are consistent over the whole month (Figure 5). Such 'large' overprediction problem of O_3 in CMAQ in China (or any other countries/regions) has not been reported. The overprediction seems very consistent in space and time. The spatial distribution of VOC emissions (in Figure 2a) also looks very uniform. More evaluation and check on the model setups and results should be performed and provided to fully understand this problem. Honestly, attributing such large O_3 predictions to VOCs emissions is somewhat dangerous. How are the predictions on CO/ SO2/EC (the species that are less

chemically reactive)? What about the meteorology predictions?

Response: We appreciate the reviewer for his/her constructive and up-to-point comments. Actually, it has been observed that in studies involving chemical transport models, there is a tendency for O_3 to be overestimated over China. For example, Li et al. (2019) and Akimoto et al. (2019) conducted a model evaluation and intercomparison of surface-level O_3 in East Asia in the context of MICS-Asia Phase III. They discovered that 14 state-of-the-art chemical transport models, including the WRF-CMAQ v5.0.2 model with MEIC-MEGAN emissions used in this study, widely overestimated surface O_3 over China by 10-30 ppb (20-60 μg/m^3), especially in the North China Plain and the Pearl River Delta. Liu et al. (2018), Qiao et al. (2019), and Xiong et al. (2023) also found similar O_3 overestimation (~ 20 μg/m^3) in the Sichuan Basin and the Yangtze River Delta region with the WRF-MEIC-MEGAN setups. Alternatively, in some studies, O_3 simulations have been implicitly improved by adjusting model chemical mechanisms or adjusting precursor emissions. For example, to study the effects of emission changes on the worsening of urban ozone pollution in China, Liu and Wang (2020) modified the original CMAQ model to update heterogeneous reactions to weaken the atmospheric oxidation capacity and thus inhibiting O_3 formation.

To facilitate the comparison of prior and posterior NMVOC emissions, we used a consistent legend in Figure 2. In fact, although NMVOC emissions are prevalent across much of central and southern China, the higher emissions in Figure 2a are concentrated in places with lush vegetation cover, such as Hunan, Jiangxi, and Zhejiang provinces, as well as in places with intensive anthropogenic activities. Despite our optimization of O_3 precursor emissions, the posterior simulations still show some degree of overestimation, indicating the presence of a systematic bias. We agree that the model-data mismatch error not only originates from the emissions, but also from variations in meteorological fields, spatial resolution, model treatments of nonlinear photochemistry and other physical processes. We utilized surface meteorological measurements from 400 stations, including temperature at 2 m (T2), relative humidity at 2 m (RH2), and wind speed at 10 m (WS10), and planetary boundary layer height (PBLH) measured by

sounding from 84 stations to evaluate the performance of WRF simulations (Figure S9 and Text S2). The results showed that the WRF model satisfactorily reproduced T2, RH2, WS10, and PBLH, with small biases of -0.5 °C, -5.3%, 0.3 m/s, and -42.4 m, respectively. The underestimated PBLH may lead to an overestimation of O_3, but the overestimated WS10 somewhat compensates for this overestimation. Additionally, due to the relatively coarse spatial resolution, NO titration effects in urban areas may not be well represented in the model, leading to an overestimation of O_3 in these areas. Model inherent errors arising from the model structure, parameterization, and the simplification or lack of chemical mechanisms inevitably affect O_3 simulations (Li et al., 2020). For example, Li et al. (2018) reported that heterogeneous reactions of nitrogen compounds could reduce surface O_3 concentration by 10–20 ppb for the polluted regions over China. These reactions have not been fully incorporated in CMAQ chemical mechanisms. However, there is still a lack of reasonable and effective algorithms to solve the model error in atmospheric data assimilation (Houtekamer and Zhang, 2016).

Due to inconvenient access to EC observation data, we only show the mean bias (BIAS), root mean square error (RMSE), and correlation coefficient (CORR) for simulated CO and SO_2 concentrations in the CEP and VEP experiments (Figures R1 and R2). There is a significant underestimation of CO in the CEP with prior emissions. However, a notable underestimation of prior CO emissions (MEIC) of about 100% has been confirmed by inversion estimations (Feng et al., 2020; Tang et al., 2013; Wu et al., 2020) and model evaluations (Kong et al., 2019) in previous studies. The BIAS of SO_2 is relatively small. Overall, after optimization, the BIAS and RMSE of CO were reduced from -0.27and 0.36 to -0.09 mg/m^3 and 0.21 mg/m^3, respectively, and the BIAS and RMSE of SO_2 were reduced from -0.36 and 7.0 μg/m^3 to -0.35 and 3.34 μg/m^3, respectively.

We have added following discussions in the revised manuscript. See lines 545-568, pages 25-26.

"O_3 simulations over China have a tendency to be overestimated in studies involving

chemical transport modeling. For example, by intercomparing 14 state-of-the-art CTMs with O_3 observations within the framework of the MICS-Asia III, Li et al. (2019) identified a substantial overestimation of annual surface O_3 in East Asia, ranging from 20 to 60 μg m^{-3}. Notably, the NCP exhibited substantial overestimations, with most models overestimating O_3 by 100–200% during May–October. Despite our optimization of O_3 precursor emissions, the posterior simulations still exhibit some degree of overestimation (Figure 4), suggesting that there may indeed be an effect of systematic bias, such as meteorological fields, spatial resolution, model treatments of nonlinear photochemistry and other physical processes. The WRF can generally reproduce meteorological conditions sufficiently in terms of their temporal variation and magnitude over China (Figure S9), with small biases of -0.5 °C, -5.3%, 0.3 m/s, and -42.4 m for temperature at 2 m, relative humidity at 2 m, and wind speed at 10 m, and planetary boundary layer height, respectively. However, due to the relatively coarse spatial resolution, NO titration effects in urban areas may not be well represented in the model, which can lead to an overestimation of O_3 in these areas. Additionally, model inherent errors arising from the model structure, parameterization, and the simplification or lack of chemical mechanisms inevitably affect the O_3 simulations. For example, Li et al. (2018) reported that heterogeneous reactions of nitrogen compounds could weaken the atmospheric oxidation capacity and thus reduce surface O_3 concentration by 20–40 μg m^{-3} for the polluted regions over China. These reactions have not been fully incorporated in CMAQ chemical mechanisms. However, there is still a lack of reasonable and effective algorithms for addressing model errors through assimilation (Houtekamer and Zhang, 2016).”

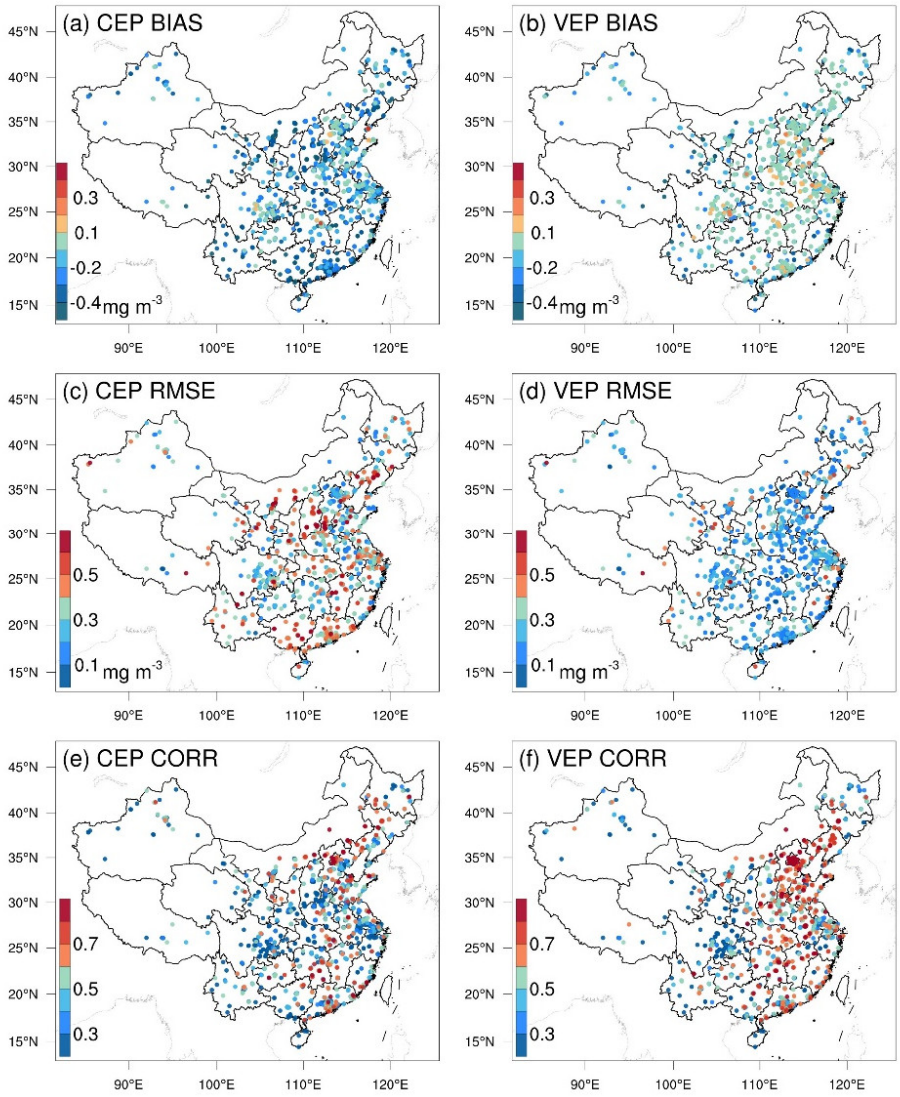

Figure R1. Spatial distribution of mean bias (BIAS, a and b), root mean square error (RMSE, c and d), and correlation coefficient (CORR, e and f) for simulated CO using prior (left, CEP) and posterior (right, VEP) emissions, respectively, against observations.

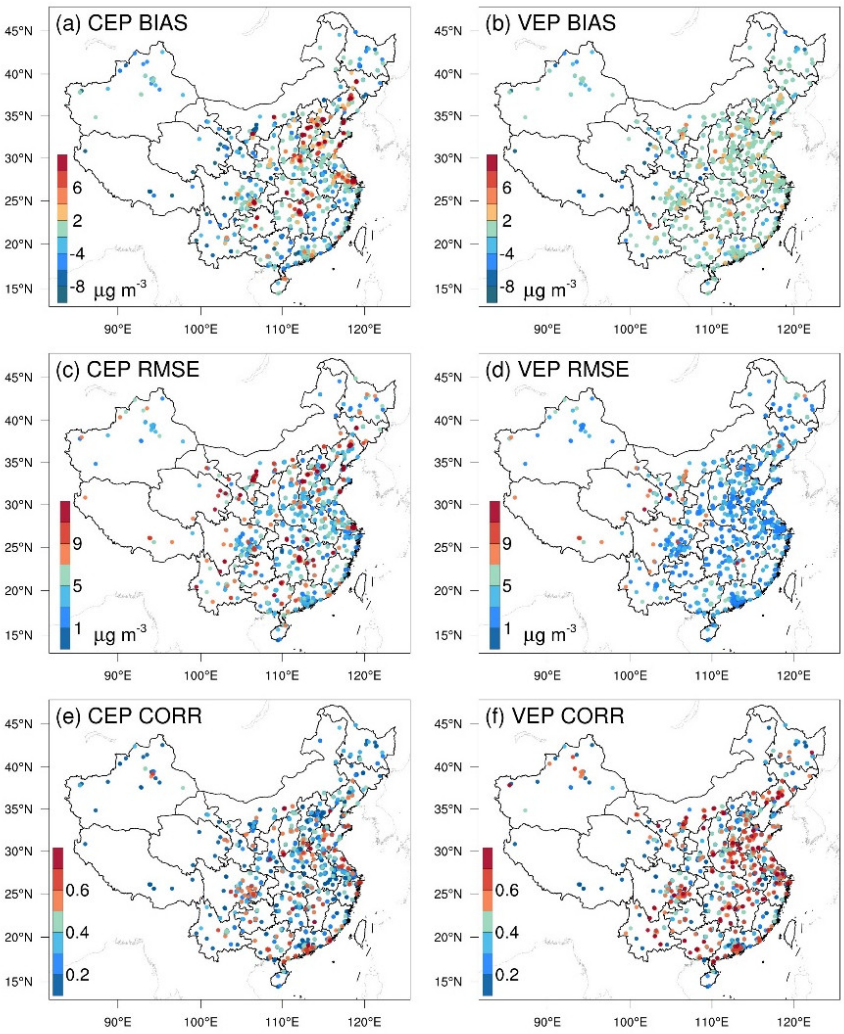

Figure R2. Same as Figure R1, but for SO_2.

2. Figure 4a and Figure 6a seem not consistent, the difference in south China in Figure 6a looks not as significant as the north. Also the observations look no significant spatial variation on Figure 6a, and MDA8 O_3 in August is most in blue-green color (~110 ug/m3).

Response: Thanks for your comments. Figure 4a shows the BIAS of simulated O_3 throughout the entire phase in the CEP1, while Figure 6a shows a comparison between simulated and observed values for MDA8. Figure R3 shows the BIAS of MDA8 simulated in the CEP1 experiment, i.e., simulated minus observed in Figure 6a. A spatial and magnitude resemblance can be observed between Figure R3 and Figure 4a.

The original Figure 6a indeed did not show significant spatial variations. We have readjusted the legend (Figure R4), please refer to Figure 6 in the revised manuscript.

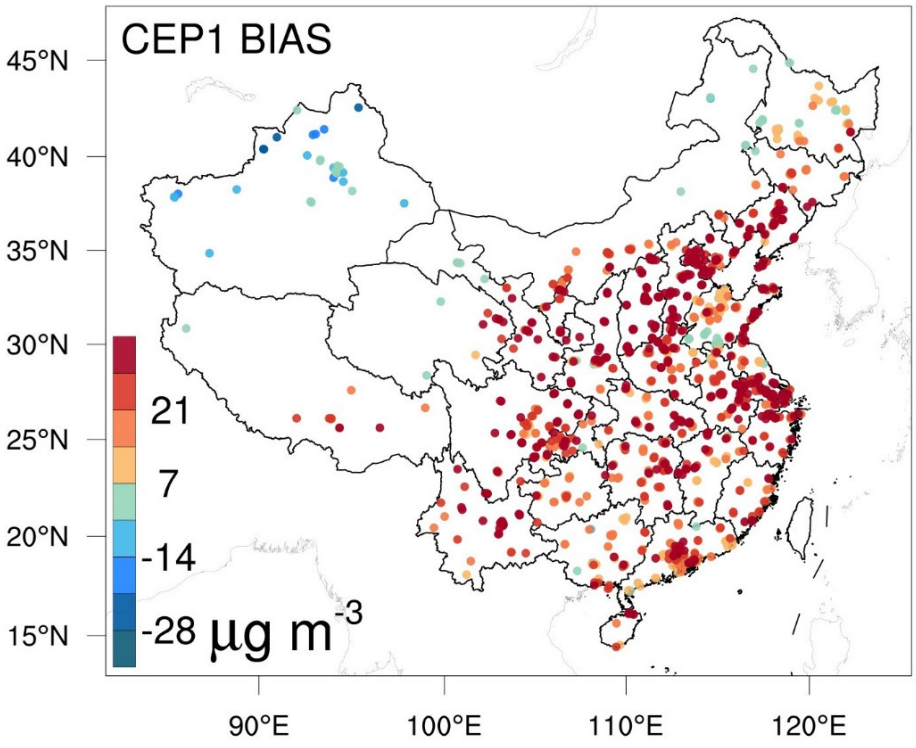

Figure R3. Spatial distribution of BIAS for simulated maximum daily 8-hour average (MDA8) O_3 concentrations in the CEP1 experiment.

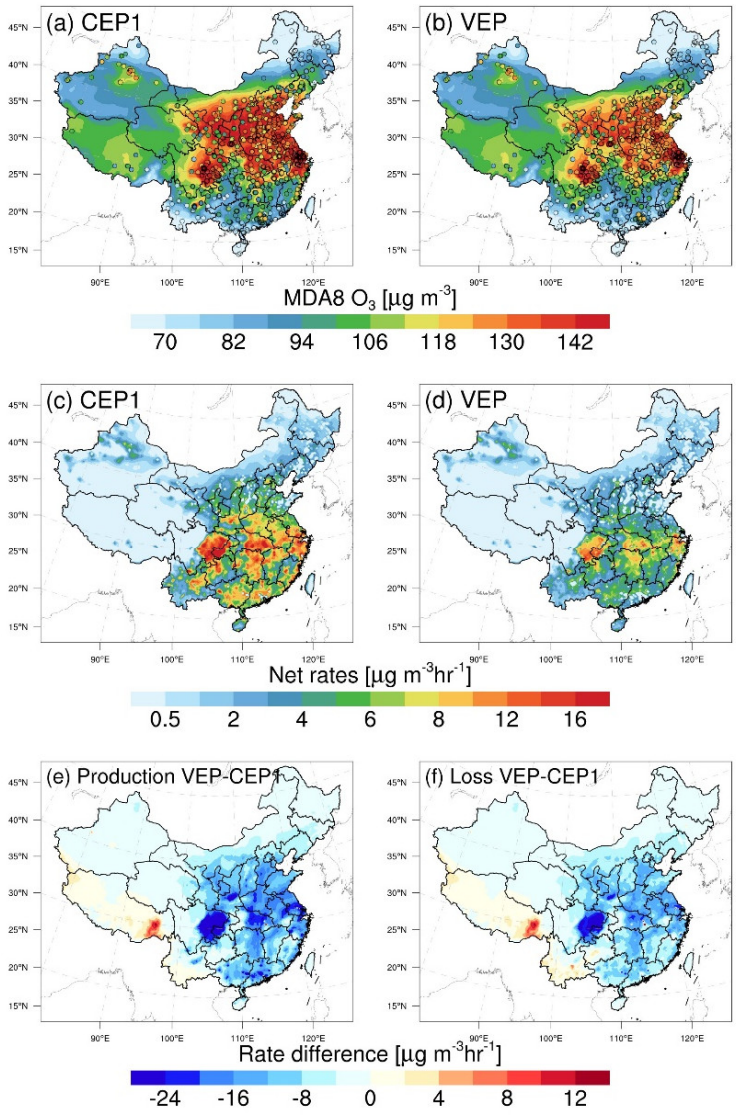

Figure R4. Comparisons of (a, b) simulated maximum daily 8-hour average (MDA8) O_3 concentrations, (c, d) net reaction rates, (e, f) and differences in production and loss rates between CEP1 and VEP experiments at the surface. Surface MDA8 O_3 values (circles) from the national control air quality stations were overlaid (Figure 6 in the revised manuscript)

3. What do different symbols/colors in Figure 1 mean?

Response: Thanks for this comment. In Figure 1, black squares denote surface meteorological measurement sites; navy triangles indicate sounding sites, and red and blue dots represent air pollution measurement sites, where red dots are used for

assimilation and blue dots for independent evaluation.

We have added a legend in Figure 1, and supplemented the explanation in the caption. See lines 286-287, page 11.

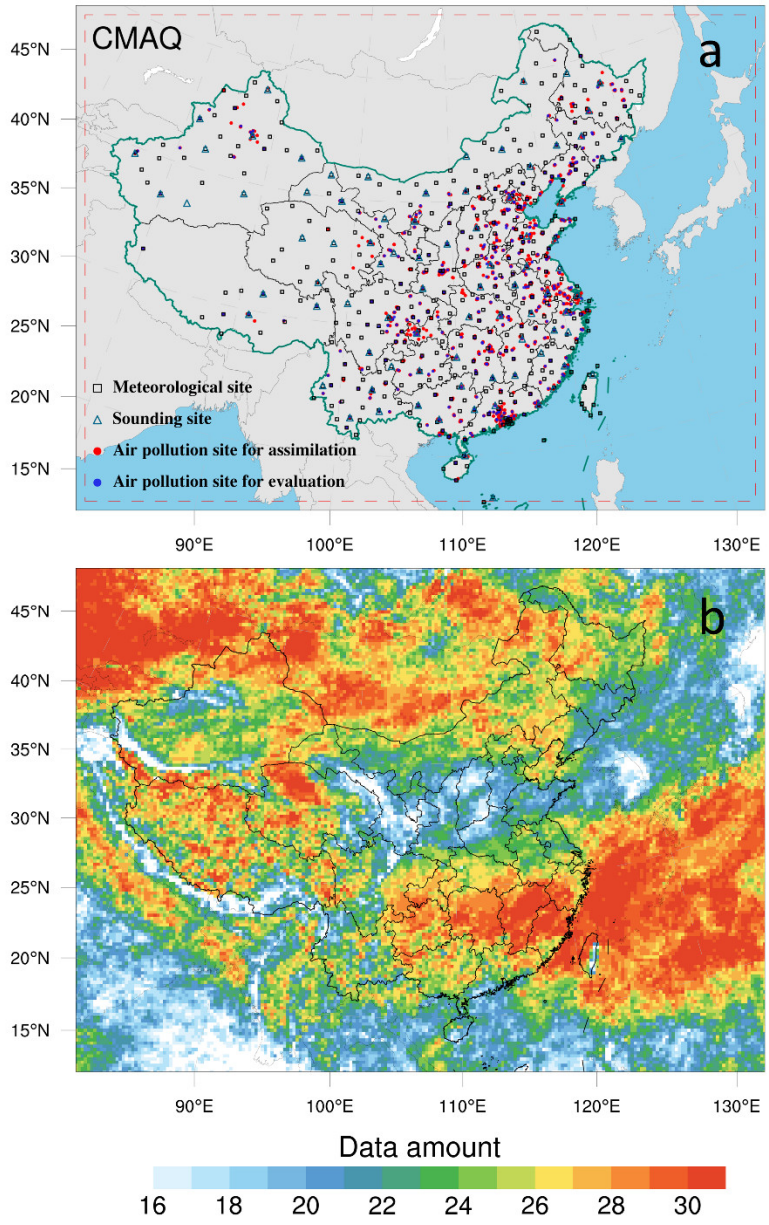

Figure 1. Model domain and observation network (a) and data amount of TROPOMI HCHO retrievals during August 2022 in each grid (b). The red dashed frame delineates the CMAQ computational domain; black squares denote surface meteorological measurement sites; navy triangles indicate sounding sites (Text S1), and red and blue dots represent air pollution measurement sites, where red dots are used for assimilation and blue dots for independent evaluation.

4. Why not choose 2020 as the study year if you have 2020 MEIC emissions?

Response: Thanks for this comment. Yes, we chose 2020 MEIC inventory as the prior emission, and selected August 2022 as the research stage. On one hand, the publicly available MEIC inventory has a lag, currently updated only until 2020. However, our system adopts a 'two-step' inversion strategy, allowing for the timely correction of residual errors from the previous assimilation window in the current window, thus ensuring that the RAPAS system has a relatively low dependence on prior emissions, which has been proven in (Feng et al., 2023). On the other hand, in the summer of 2022, the eastern part of China experienced the strongest and longest-lasting heatwave since 1961 (Wang et al., 2023). High temperatures and drought significantly affect vegetation growth and NMVOC emissions, which also impact O_3 production. Such a complex weather system provides a good test for the assimilation capability of our system.

We have added following discussions in the revised manuscript. See lines 321-325, page 12.

"During the summer of 2022, southern China experienced severe heatwave conditions. The combination of high temperatures and drought had a pronounced effect on vegetation growth and NMVOC emissions, thereby influencing O_3 production (Wang et al., 2023). Consequently, we opted to focus on August 2022, as it presented an ideal period for testing the assimilation capabilities of our system. Before implementing the emission inversion,… …"

5. BVOCs is greatly overestimation by MEGAN (over 50%). Have any other studies reported similar findings with MEGAN in any regions? If no, please explain why such problem occurs in China? Why previous modeling studies in China with CMAQ have not encountered such problems (also the O_3 overprediction problem)?

Response: Thanks for this comment. The estimation of BVOC emissions by MEGAN is not consistently overestimated. For example, Bauwens et al. (2016) optimized global BVOC emissions using source inversion of OMI HCHO observations and found that

MEGAN tends to overestimate BVOCs in low-latitude regions while underestimating them in high-latitude areas. Research on the inverse estimation of BVOC emissions in China remains limited. For China, Bauwens et al. (2016) showed that the greatest overestimation of BVOC emissions occurred in southern China, up to 45%. The overestimation of BVOC emissions simulated by MEGAN in China is further validated by model evaluation studies (Kim et al., 2017; Kim et al., 2024). In other regions, Marais et al. (2014) found that MEGAN's isoprene emissions were 5-10 times higher than the canopy-scale flux measurements obtained from African field campaigns in equatorial forest and woody savannas. Warneke et al. (2010) found that MEGAN overestimated BVOC emissions by up to a factor of two when compared to estimates based on airborne measurements over Texas. Millet et al. (2008) compared isoprene emissions derived using satellite-observed HCHO columns with MEGAN emissions for North America, noting an average overestimation of BVOC emissions by a factor of 2, reaching up to a factor of 5 in certain locations. Similarly, Wang et al. (2017) observed a significant overestimation of BVOC emissions by MEGAN, averaging a factor of 3 in the United States. Kaiser et al. (2018) applied an adjoint algorithm to estimate isoprene emission over the southeast US, revealing an average overestimation of MEGAN-derived BVOC emissions by 40%, slightly lower than the 50% overestimations reported by Bauwens et al. (2016). Additionally, Chaliyakunnel et al. (2019) found the modeled BVOC emissions using MEGAN were overestimated by approximately 30–60% for most locations and seasons. Therefore, there is indeed a possibility of significant uncertainty in MEGAN.

The significant decrease in BVOC emissions observed in this study may also be influenced by other factors. Apart from inaccuracies in the distribution of plant functional types, empirical parameterization, especially concerning responses to temperature and drought stress, can introduce substantial uncertainties (Angot et al., 2020; Jiang et al., 2018). Zhang et al. (2021) highlighted that the temperature-dependent activity factor increases evidently with rising temperatures in the MEGAN model. Additionally, Wang et al. (2021) pointed out that the missing of a drought scheme is

one of the factors causing the isoprene overestimation in the MEGAN model. Wang et al. (2022) applied new drought stress algorithms to simulate the impact of drought on isoprene emission and found that drought can decrease isoprene emission globally by 11%. During the summer of 2022, southern China experienced severe heatwave conditions. The MEGAN model may not effectively capture the impacts of high temperatures and drought on vegetation, leading to significant uncertainties in BVOC emissions.

It has been widely observed in existing studies that there is a trend of overestimation of O_3 in China, which is similar to the overestimation found in this study. Please refer to Comment 1 for further details.

We have added the following discussions. See lines 374-390, page 15.

"Additionally, uncertainties in MEGAN parameterization have significant implications for NMVOC emission estimations, particularly concerning the responses of vegetation in MEGAN to temperature and drought stress (Angot et al., 2020; Jiang et al., 2018). Zhang et al. (2021) highlighted that the temperature-dependent activity factor noticeably increases with rising temperatures in MEGAN. Wang et al. (2021b) pointed out that the missing of a drought scheme is one of the factors causing the overestimation of isoprene emissions in MEGAN. Opacka et al. (2022) optimized the empirical parameter in the MEGANv2.1 soil moisture stress algorithm, resulting in significant reductions in isoprene emissions and providing better agreement between modelled and observed HCHO temporal variability in the central U.S. During the study period, China experienced severe heatwave conditions, which may further hinder the MEGAN's ability to effectively capture the impacts of high temperatures and drought on vegetation, thus resulting in significant overestimation in NMVOC emissions (Wang et al., 2022). Nevertheless, the large magnitude of emission reductions of 50.2% in our inversion is comparable to studies in southern China (Bauwens et al., 2016; Zhou et al., 2023), southeastern US (Kaiser et al., 2018), Africa (Marais et al., 2014), India (Chaliyakunnel et al., 2019), Amazonia (Bauwens et al., 2016), and parts of Europe (Curci et al., 2010), but opposite to the large-scale emission increase over China in Cao et al. (2018). For NO_x (Figure S4), … …"

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