

Dear Reviewer,

We appreciate your careful consideration of our manuscript. We have carefully responded to all of your **point-by-point** comments and issues and have revised the manuscript accordingly. These revisions are described in detail below.

This manuscript uses a chemical box model approach to calculate VOC and NO_x loss rates, radical chemistry and ozone production rates when constrained with pollutant concentrations in a suburb of Beijing, China. Overall, I recommend revisions of the modelling approach. Below are my specific comments.

Response: Thank you for your positive comments and good suggestions. We will respond your comments point-by-point.

1. Line 43. Reference for Seinfeld is missing Pandis author.

Response: Thank you for your suggestion. We have updated the missing author in the corresponding position. In addition, we have double checked and updated all references in the revised manuscript.

2. Line 125. To compare the model vs measurements, it is advised to look at slope, intercept and not just correlation coefficient.

Response: Thank you for your good suggestion. In the revised SI, we updated the Figure S2 with adding the slope and intercept. We have also updated in the revised manuscript “The correlation coefficient is 0.9 (with a slope of 0.7), indicating that the concentrations of NMHCs are comparable using these two measurement techniques” in [lines 124-126](#).

It should be noted that in line 108, we are discussing the performance between the two instruments for VOC measurements rather than that between the model and measurements. When we comparing the performance of the model with measurements, the slope was also added in addition to the correlation coefficient in the revised manuscript ([line 203](#)).

3. Line 147. The hydrocarbon ratio photochemical clock concept works well when you have an isolated source that co-emits the hydrocarbons and a receptor site with no emissions in between. If there are different sources in between and also mixing of airmasses into the plume with different photochemical ages then it complicates the concept and rationale. The use of an early morning time to define the aromatic ratio is also not explained well. The early morning measurements likely reflect the concentration of local emissions under a shallow inversion layer. If the emissions are uniformly distributed across region, then this might be the best time to estimate the emission ratio for xylene to ethylbenzene. The assumption that the emission source remains constant over the region is questionable. It might be helpful to look at gridded regional air quality emissions over the region and plot the emission ratio to see if it is relatively uniform spatially.

Response: Thank you for good comments and suggestion. We agree with you that the uniform distribution and constant emissions of VOCs sources are critical for calculating chemical loss of VOCs using hydrocarbon ratio photochemical clock. At least, the emission pattern of VOCs should be constant during the whole day when compared with that in the early morning. Previous studies have found that the ambient ratios of VOCs can reflect their relative emission rates from sources (Golden et al., 2000; Jobson et al., 2004). To verify the rationality of this assumption, Shao et al (2011) had tested relative emission rates from sources by testing different ambient ratios of four pairs of hydrocarbons, i.e., benzene vs acetylene, trans-2-butene vs cis-2-butene, ethene vs toluene and n-hexane vs toluene, that having similar k_{OH} values in each pair. We also further choose the benzene vs acetylene and n-hexane vs toluene to check whether VOCs emissions are constant during our observations. The results are showed in Figure R1 (same as Figure S5). The linear correlation coefficients (R^2) were higher than 0.7 and were equal to those reported by Shao et al (2011). This indicates that the assumption of a constant VOCs emission should be reasonable during our observations.

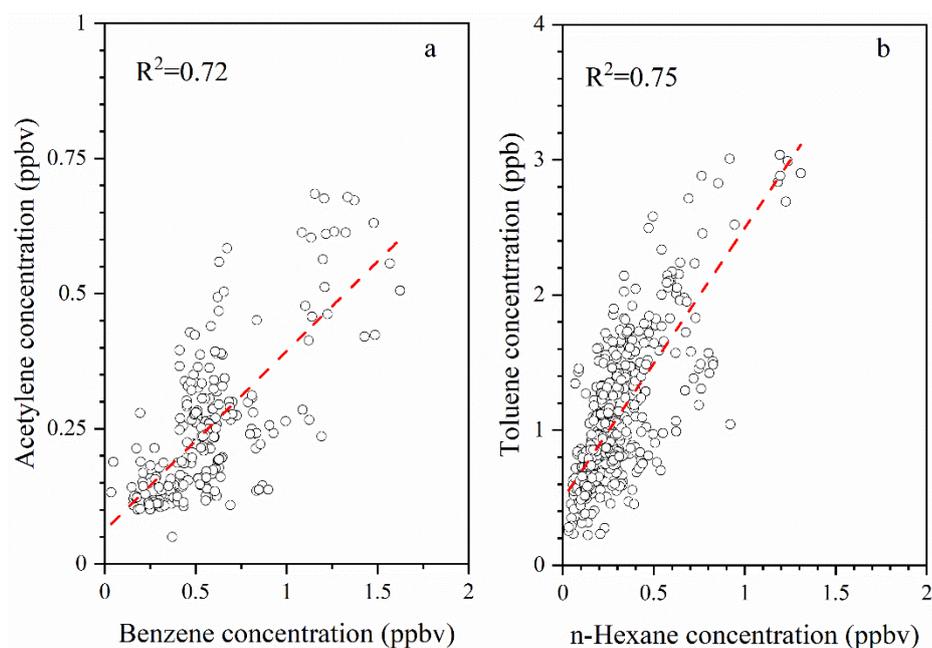


Figure R1. The relationship between the concentration of toluene vs acetylene and n-hexane vs toluene.

As for the assumption that the emission source remains constant over the region, it is better to look at gridded regional air quality emissions over the region. Unfortunately, such regional gridded data are unavailable at the present time. Alternatively, we performed spatial distribution analysis using a source-receptor model (potential source contribution function, PSCF). In Figure R2 (same as Figure S6), the emissions of VOCs are not spatially uniform in Beijing, i.e., strong emissions are in the south or southeast directions. However, the PSCF patterns in the daytime are highly similar to that in the early morning. This means that the emissions of VOCs should be constant during the daytime.

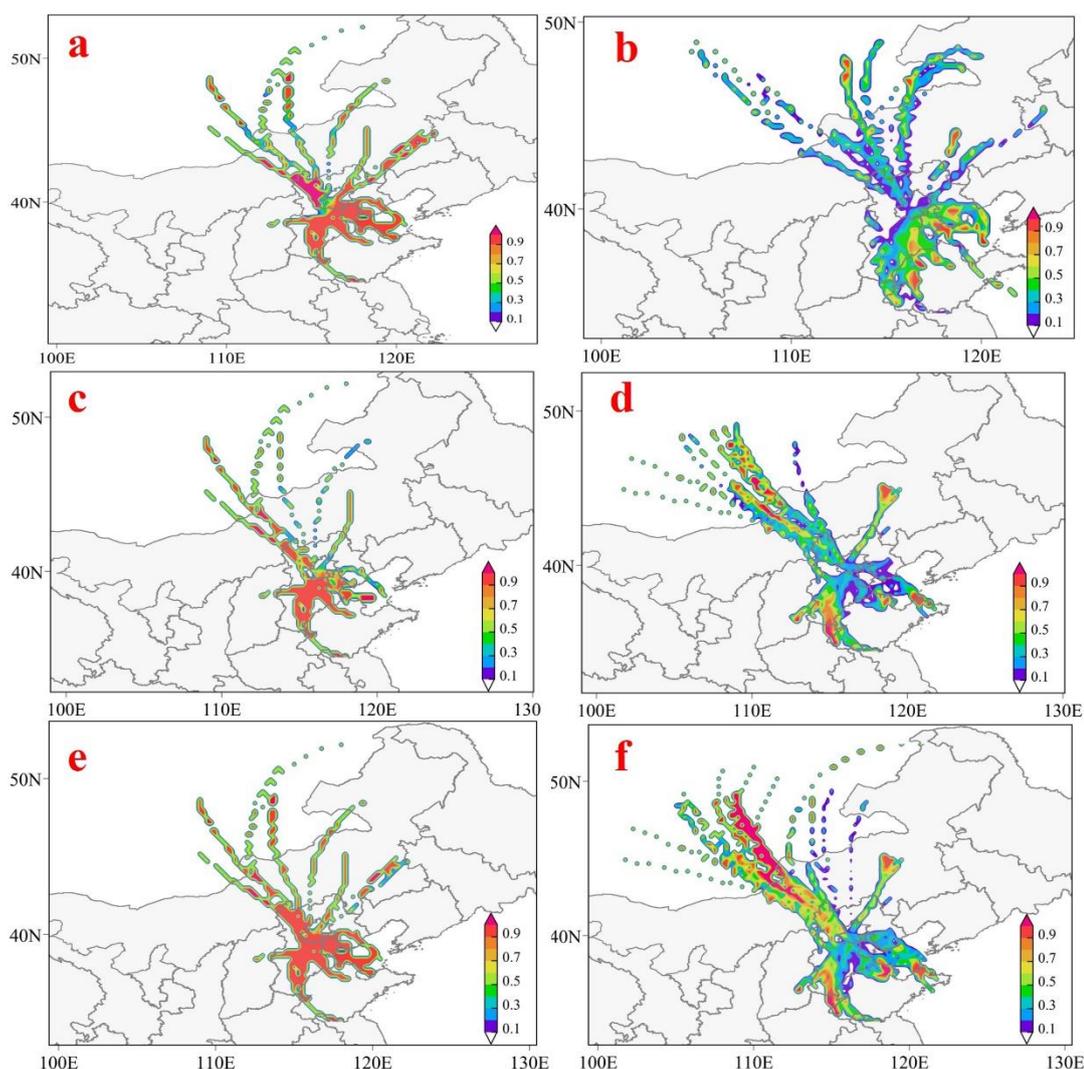


Figure R2. The potential source contribution function (PSCF) maps for the ratio of xylene to ethylbenzene (a and b), ethylbenzene (c and d), and xylene (e and f) arriving in the observation site. The figures of a, c and e are the results of 05:00 and 06:00, and the figures of b, d and f are the results of daytime (07:00-19:00).

In the revised manuscript, we added the sentences “. In previous work (Shao et al., 2011; Zhan et al., 2021), the selection of ethylbenzene and xylene as tracers was justified for calculating ambient OH exposure under the following conditions: 1) the concentrations of xylene and ethylbenzene were well correlated (Figure S4), which indicated that they were simultaneously emitted; 2) they had different degradation rates in the atmosphere; and 3) the calculated PICs were in good agreement with those calculated using other tracers, such as i-butene/propene (Zhan et al., 2021). To test the

relative constant emission ratio from different sources, we chose benzene vs. acetylene and n-hexane vs. toluene as references, and the result is shown in Figure S5. These ambient ratios could directly reflect their relative emission rates from sources (Goldan et al., 2000; Jobson et al., 2004). The linear correlation coefficients (R^2) were generally higher than 0.7, which were equal to that reported by Shao et al. (2011). To further test the assumption that the emissions of xylene and ethylbenzene were constant throughout the day, their potential sources were calculated using a source-receptor model (the potential source contribution function, PSCF). As shown in Figure S6, xylene and ethylbenzene showed similar distributions. In addition, the ratio of ethylbenzene/xylene at 5:00 and 6:00 was similar to that during the daytime. These results indicated that the emissions of xylene and ethylbenzene were constant throughout the day.” in lines 160-175. In future, we will try to combine OBM and the regional air quality model to better understand the influence of photochemical initial VOCs.

4. Line 178-182. The mean daytime OH used for the calculation was $4.3E6 \text{ molec/cm}^3$. Please state that the measured aromatic ratio was also a mean value for the same daytime period used to calculate the OH.

Response: Thank you for good comments. Indeed, here we are discussing the daytime OH exposure ($\text{molecules cm}^{-3} \text{ h}$) but not OH concentration (molecules cm^{-3}). We calculated hourly OH exposure from 07:00 to 19:00 using the ratio of xylene to ethylbenzene according to Eqs.1 and 2 in the manuscript. Then, we obtained the mean daytime OH exposure $4.3 \pm 1.9 \text{ molecules cm}^{-3} \text{ h}$. The mean daytime OH concentration ($4.3 \pm 1.9 \times 10^6 \text{ molecules cm}^{-3}$) was calculated based on the JO1D using the method reported in our previous work (Liu et al., 2020b, Liu et al., 2020c). In lines 178-182 in the revised manuscript, we rephrased the sentences to make it clearer “The OH exposure was from 0.82 to $8.1 \times 10^6 \text{ molecule cm}^{-3} \text{ h}$, with a mean daytime value of $4.3 \pm 1.9 \times 10^6 \text{ molecules cm}^{-3} \text{ h}$. Accordingly, the mean photochemical ages were $1.7 \pm 0.9 \text{ h}$ using the mean daytime (8:00-17:00 LT) OH concentrations ($4.3 \pm 3.1 \times 10^6 \text{ molecules cm}^{-3}$) calculated based on JO1D using the method reported in our previous work (Liu

et al., 2020b; Liu et al., 2020c).”.

5. Line 185. I agree that the biogenic isoprene emission is not co-located with the aromatic emission sources and this complicates the ratio method. Can the authors look at regional model emissions for isoprene to see if the isoprene sources are local near the site or whether they are closer to the aromatic sources? A possible assumption is that the production and loss of isoprene balance along the transport and concentration of isoprene remain constant in the trajectory from the aromatic source region to the site. Again, regional air quality model results would be able to show what the distribution of isoprene looks like around the site.

Response: Thank you for good suggestion. We think that isoprene is mainly from biogenic emissions in summer but not co-located with the aromatic emission sources. This is evidenced by the diurnal variations of isoprene, xylene and ethylbenzene (Figure R3, same as Figure S14) although the regional model emissions for isoprene are unavailable at the present time. As shown in Figure R3, isoprene and aromatics (xylene and ethylbenzene) showed totally different diurnal variations. This means isoprene sources are from biogenic emissions.

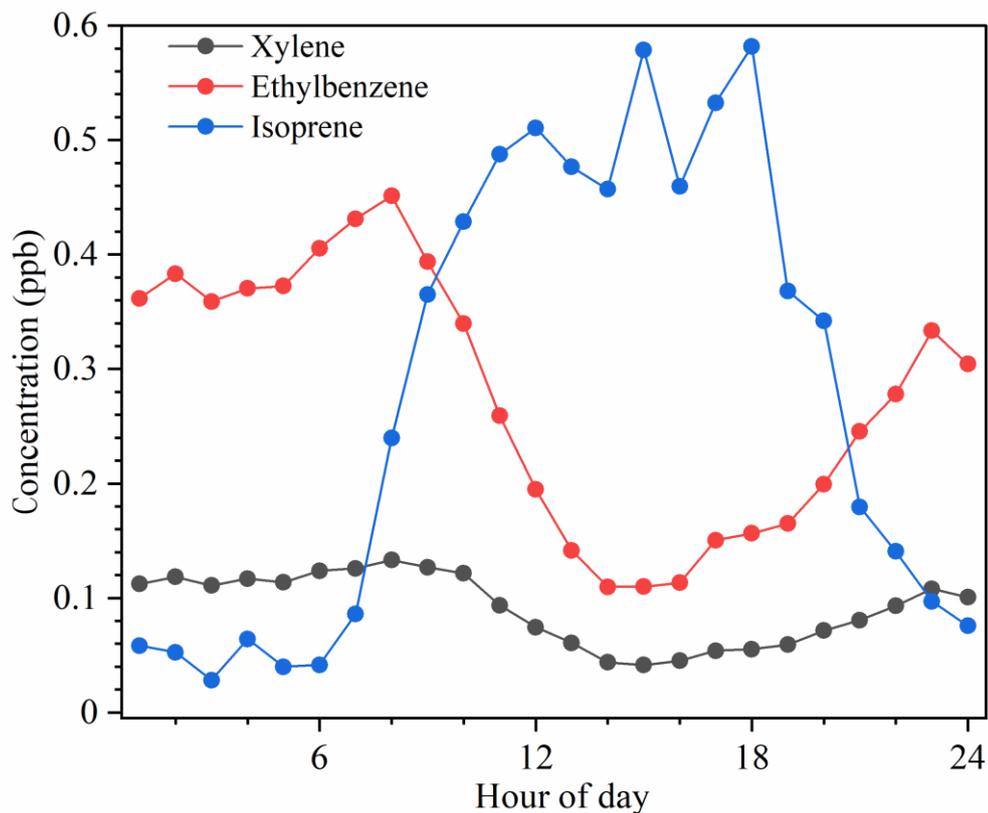


Figure R3. The mean diurnal curves of xylene, ethylbenzene and isoprene.

On the other hand, like xylene and ethylbenzene, we also carried out PSCF analysis (Figure R4, same as Figure S15) for isoprene to check whether the production and loss of isoprene is balanced along the transport and concentration of isoprene remain constant in the trajectory from the aromatic source region to the site. The results showed that the spatial pattern of isoprene is even during our observations. This indirectly indicated that it could be considered as the balance on production and loss of isoprene along the transport, and the concentration of isoprene remain constant in the trajectory from the aromatic source region to the site.

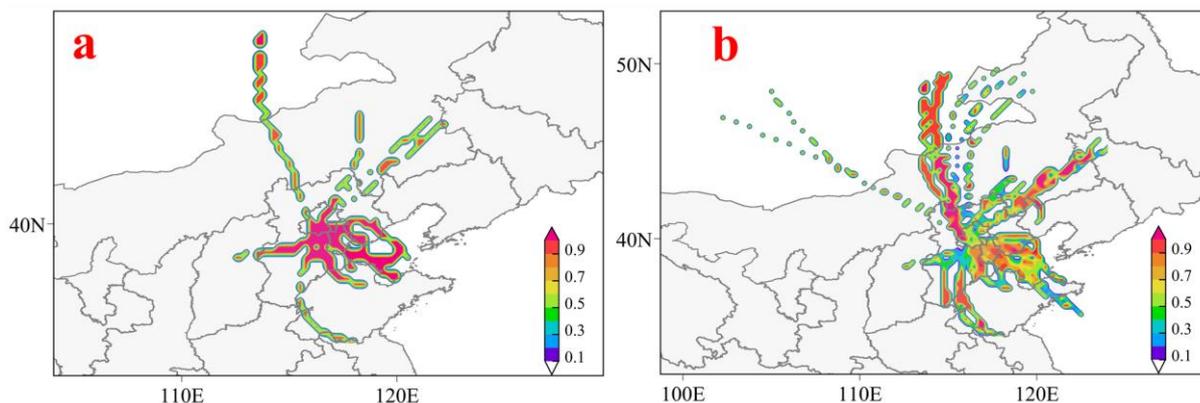


Figure R4. The potential source contribution function (PSCF) maps for the isoprene arriving in the observation site. The figures of a and b are the results of 05:00 and 06:00, and the daytime (07:00-19:00), respectively.

In lines 124-130 in the revised SI, we added the sentences “Isoprene is mainly from biogenic emissions but not co-located with the aromatic emission sources in summer, which is evidenced by the diurnal variations of isoprene, xylene and ethylbenzene (Figure S14) although the regional model emissions for isoprene are unavailable at the present time. The results of PSCF analysis showed that the spatial pattern of isoprene is even during our observations in S15, which indirectly indicated that it could be considered as the balance on production and loss of isoprene along the transport, and the concentration of isoprene remain constant in the trajectory from the aromatic source region to the site.”.

6. Line 233. I would recommend “grouped into lumped species” instead of parcels.

Response: Thank you for good suggestion. We have updated “the observed VOCs are grouped into different lumped species according to the classification of RACM2” in the lines 233-235 in the revised manuscript.

7. Line 238. Five minutes is enough time for the radicals to reach steady-state but not the NO_x and OVOCs. For example, PAN has a long lifetime and would not reach steady state in 5 min. Given that the intent is to “correct” the VOCs to an initial condition and you expect an ~ 2hr photochemical time then an equivalent time to run the model might be best approach and would give time for OVOCs to spin up to more reasonable mixing ratios.

Response: Thank you for comments and suggestion. The time interval for the outputs is usually set to 60 min (Lu et al., 2013; Lu et al., 2017; Tan et al., 2019) determined by the time resolution of observation data. However, the time interval of 60 min is too coarse to run the model because the sun can move a lot within one hour. Therefore, a higher time resolution (5 min) is usually taken along with interpolation of

the inputs in OBM simulations to reduce the influence of great distortion of meteorological parameters in a long-time interval (Tan et al., 2017). The time interval (60 min) in this study is similar with that used by Lu et al (2013) and Lu et al (2017), and higher than that (30 min) reported by Tan et al (2018), which indicated that the photochemical time setting was acceptable. We agree with you that OVOCs and PAN have a long lifetime compared with the radicals (OH, HO₂ and RO₂). Although the long lifetime species would not reach steady state in 5 min, the model outputted their concentrations with a time resolution of 60 min. This indeed accounted for the time for OVOCs and PAN to spin up to more reasonable mixing ratios. In addition, the chemical cycle of radicals is critical for ozone formation. The simulation is more reliable once the radical sources, sinks and propagation of radicals can be well simulated at this time interval.

In the revised manuscript, we have updated the sentences “**The chemical model simulated photochemical reactions with input species for a time interval of 60 minutes, which was enough for NO_x, OH, HO₂, and RO₂ to reach a steady state because the typical relaxation time of the chemical system is 5-10 minutes in summer (Tan et al., 2018). However, all the species and parameters were input at a 5 min interval by data interpolation to reduce simulation inconsistencies and large distortions of meteorological parameters at longer time intervals (Tan et al., 2018)**” in lines 235-240.

8. One major concern that I have in the photochemical initiation of VOCs is that the authors are not considering the correction needed for NO_x as it also reacts in the trajectory reaching the site. The VOCs and NO_x can be emitted by common combustion processes, particularly in an urban area. The authors also note they follow the same diurnal profile as VOCs. The lifetime of NO_x is comparable to some VOCs so why not correct the NO_x as well? Other studies have done this and in fact the NO_x/NO_y is an alternative ratio in the photochemical clock method (Hayes et al., 2013).

Response: Thank you for good comments and suggestion. We agree with you that photochemical conversion of NO_x should be corrected. Because the NO_y concentrations were unavailable during our observations, we simply corrected the

concentration of NO and NO₂ using the OH exposure and assuming a gas phase oxidation by OH radical ($k_{\text{NO-OH}}=3.0\times 10^{-11}$ cm³ molecules s⁻¹ and $k_{\text{NO}_2\text{-OH}}=1.2\times 10^{-11}$ cm³ molecules s⁻¹, Atkinson et al., 2004). The corrected data are showed in Figure R5. The average concentrations of NO, NO₂, and NO_x were underestimated 0.6±0.6 ppb, 1.5±1.4 ppb, and 2.1±2.0 ppb, respectively, lower than the underestimated VOCs (6.9±0.5, Figure 1), which indicated that the role of the photochemical loss of NO_x is less important than VOCs when evaluating the ozone formation process and mechanism. In the early morning, the PICs of NO_x were almost the same as the observed values. On the other hand, the previous studies (Li et al., 2020; Tan et al., 2019; Sun et al., 2011) have showed that the Beijing belonged to VOCs-limited regime, which means that the NO_x is sufficient during the nonlinear relationship of NO_x-VOCs-O₃. In other word, the chemical loss of NO_x compared with photochemical loss of VOCs was less important in Beijing. Therefore, we focus on the contribution of photochemical loss of VOCs to ozone formation mechanism in this study. And in future, we will compare the ratio of NO_x to NO_y (alternative calculation) with the ratio method of xylene to ethylbenzene.

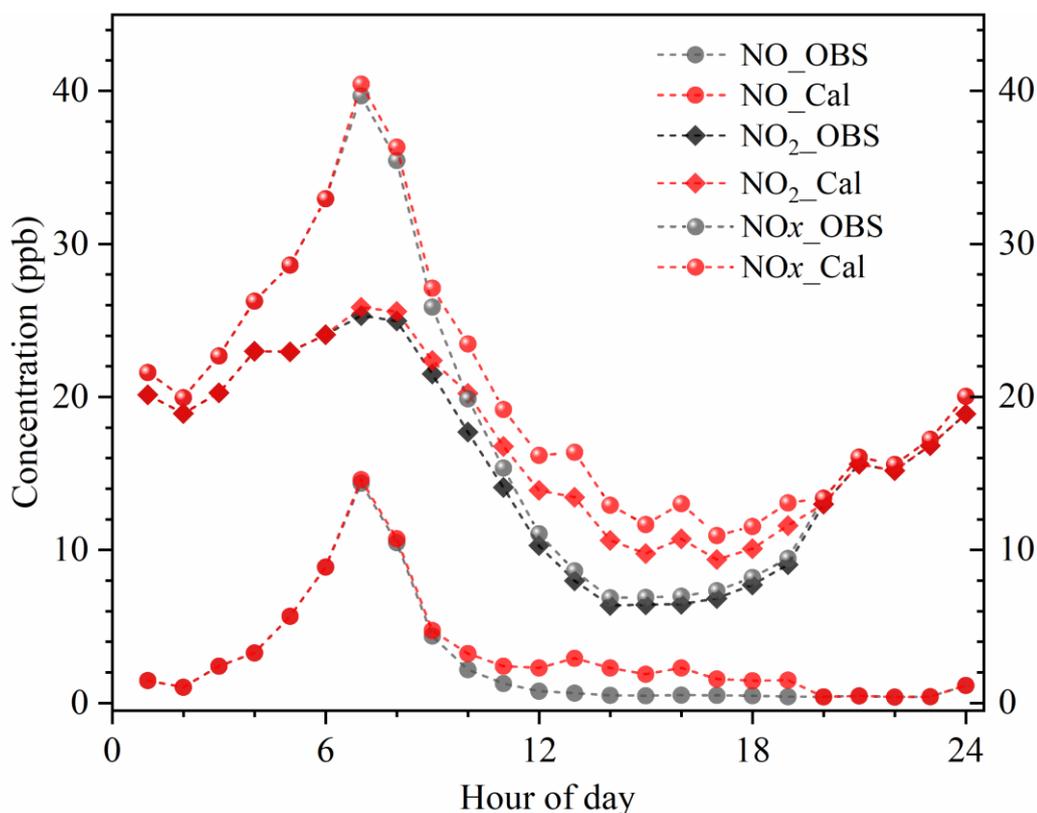


Figure R5. The mean diurnal curves of NO, NO₂ and NO_x. The corrected concentration of NO_x was calculated with the OH exposure based on the ratio method.

9. Line 248. If I understand this section, the observed ozone max was 119 ppbv and the modeled max was 70 ppbv. This seems like a large difference. If this can be explained by the mixing and transport of ozone from regional scale then maybe it might be best to constrain the ozone to the observed value each hour. For diagnosing the chemistry pathways (Figure 3,4), this ensures the ozone is at a reasonable level for calculating the alkene ozonolysis and O₁D+H₂O reaction rate.

Response: Thanks for your good comments and suggestion. The mean ozone concentration during the observation period was 44.8 ± 27.2 ppb with a maximum of 119.1 ppb (Figure S5). As shown in Figure S5, the modeled O₃ concentrations actually were comparable with the observed values. It is worth to note that we used the average data of the campaign as input to simulate the EKMA curve in this study, and then we set up 30×30 matrixes by reducing or increasing the measured VOCs and NO_x concentrations to the model input according to the base case. The modeled max value you mentioned (70 ppb in Figure 2) is comparable with the maximal mean ozone concentrations during our observations (Figure 1). When we diagnosing the chemistry pathways (Figure 3 and 4), we have constrained O₃ concentrations. Thus, this ensures the ozone is at a reasonable level for calculating the reaction rates between alkene and ozonolysis, and between O₁D and H₂O.

In the revised manuscript, we have added a sentence “**It is worth mentioning that the average survey data were selected as the baseline scenario in simulating the EKMA curve in this study.**” in lines 241-242.

10. Line 313. The daytime average P(O₃) is calculated at 3 ppbv/hr higher than with using measured VOCs. The authors appear to extrapolate to a 24-hr average by multiplying by 24 hr to get 36 ppbv/hr higher than with the measured VOCs. The nighttime P(O₃) averages could be different than daytime averages, so it would be preferred to state what the daytime average difference is between corrected and

measured VOC approaches.

Response: Thanks for your good comments. The OBM in this study was just used to simulated the daytime photochemical process (07:00~19:00 LT). Thus, the daytime average of $F(O_3)$ is 3.0 ± 2.1 ppb h^{-1} higher than with using measured VOCs. And the value of ~ 36 ppb day^{-1} is just calculated from 07:00 to 19:00 based on the average $F(O_3)$, not including the nighttime.

In the revised manuscript, we updated the sentences “The average daytime $P(O_3)$ from 07:00 to 19:00 based on the initial concentrations of VOCs was 4.0 ± 3.1 ppb h^{-1} higher than that based on the measured VOCs concentrations (Figure 3b). At the same time, the $F(O_3)$ from 07:00 to 19:00 based on the initial concentrations of VOCs was also 3.0 ± 2.1 ppb h^{-1} higher than the measured counterpart (Figure S11). Thus, the net O_3 production could be **accumulatively underestimated** by ~ 36 ppb day^{-1} from 07:00 to 19:00 if the consumption of VOCs was not considered.” in lines 310-315.

11. Line 432. I think the discussion of the diurnal profile and the different chemical and transport processes should include the mixing of the stable nocturnal surface layer in the mid-morning. As the sun heats the surface, there is significant mixing of surface layer with air above in the residual atmospheric layer which is likely composed air from the prior day mixed boundary layer and then transported to the site overnight. This residual air likely contains hydrocarbons and ozone from different sources and at different photochemical ages. The ozone increase around 9-10am is often associated with this vertical mixing. The ozone mixing down to surface is photochemically produced but from a different region from previous days (unless the region is influenced by a lake/land breeze where recirculation of the same air mass can occur.

Response: Thanks for your good comments. We agree with you that the diurnal profile of ozone will be affected by the mixing of the stable nocturnal surface layer in the mid-morning. It should mention that $R(O_3)$ is the physical transportation including horizontal and vertical mixing, and we attributed the variation of ozone in the morning all to the transportation. In fact, the upper layer is known as the residual layer (RL) in the morning, which is isolated from the surface due to inversion at night (Tan et al.,

2021). RL usually contains the air mass with higher O₃ concentration than that of nocturnal boundary layer, and when the boundary layer is gradually uplifted, the vertical transport was important due to the fast entrainment.

In the revised manuscript, we have added the sentences “**This was mainly because the residual layer (RL) that formed at night was unfavourable for the inversion of airmass in the early morning (Tan et al., 2021). The RL usually contains an airmass with a higher concentration of O₃ than that in the nocturnal boundary layer. Vertical transport becomes prominent due to the fast entrainment when the boundary layer is gradually uplifted.**” in lines 433-437.

12. Line 388. The authors state that “The radical budget analysis illustrated that the O₃ formation processes between the observed and photochemical initial VOCs showed no significant difference.” The title of the manuscript implies that the initial photochemical loss of VOCs does have an impact on the ozone formation mechanism. It seems that the ozone production rates are sensitive to the corrected VOC loss (and likely corrected NO_x loss as well). Maybe an improved title could be “Influence of Photochemical Loss of VOCs and NO_x on Ozone Formation Rates and Diagnosed Ozone Production Sensitivity in Beijing, China”

Response: Thank you for comments and suggestion. As above, the chemical loss of NO_x compared with photochemical loss of VOCs was less important in Beijing, and we focus on the contribution of photochemical loss of VOCs to ozone formation mechanism in this study. Actually, the current title “Influence of Photochemical Loss of VOCs on Understanding Ozone Formation Mechanism” might also reflect what you mean. However, we still prefer the original one to make it more concise.

13. Figure S9 shows some days with large model over-predictions. Can the authors explain what factors are contributing to the model over-predictions?

Response: Thank you for good comments. The OBM is zero-dimensional atmospheric modeling coupled with just gas-phase reaction, which means that it cannot

consider the influence of meteorological process (vertical diffusion and horizontal transportation). The dilution caused by wind or boundary layer was not well represented during the simulation, and this may lead overestimation or underestimation to some extent. The similar over-predictions were also reported by Zong et al (2018) and Zhang et al (2020), but it cannot affect the simulation of ozone formation process and mechanism because we have the constrained ozone concentration during simulations. In the revised manuscript, we added the sentences “It is worth mentioning that the results of model simulation can sometimes be overestimated or underestimated to some extent, which has also been reported by previous studies (Zong et al., 2018; Zhang et al., 2020), but this did not affect our simulations of the ozone formation process and mechanisms because we constrained the ozone concentration during our simulations.” in lines 204-208.

14. Overall, I really like Figures 3, 4 and 5. My recommendation would be to include a NO_x correction, as well as the VOC correction, and to consider a longer model time so that the OVOCs reach closer to their typical ambient concentrations. Of particular interest would be the aromatic and monoterpene oxidation products as the precursor aromatic and monoterpene have an intermediate lifetime (several hours) and their OVOCs are not typically measured. The figures are of very good quality. The paper needs some improvements for English language.

Response: Thank you for your positive comments and good suggestions. We have carefully responded to all of your point-by-point comments and issues above and have revised the manuscript and SI accordingly. We also carefully corrected language errors by a native speaker (as shown in Figure R6). The corrections have been marked in blue in the revised manuscript.

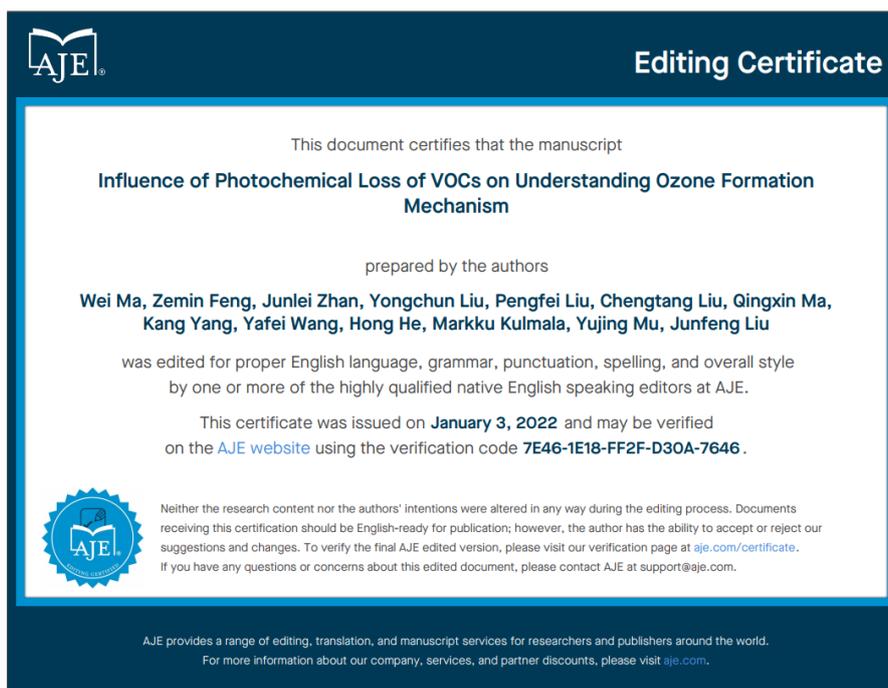


Figure R6. The editing certificate by the highly native English speaking editors at AJE.

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