1 Dear editor,

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

6

7 Review 1 #

8 Thank you to the authors for carefully considering my questions in the first round and 9 thoroughly revising their manuscript. I have a few minor suggestions to clarify the 10 added content for readers, otherwise this paper is ready for publication.

11 **Response:** Thank you for your positive comments and kind help.

12

In the expanded literature review in the introduction (around l. 87) or the next
 paragraph, it may help the reader if the authors enumerated how the MIR method differs
 from the method used here.

Response: Thank you for your good suggestions. The MIR of VOCs is determined
according to,

18
$$MIR = \lim_{\Delta VOC \to 0} \left[\frac{O_3(VOCs_{MIR} + \Delta VOCs) - O_3(VOCs_{MIR})}{\Delta VOCs} \right]$$
 (Eq. R1)

where, $O_3(VOCs_{MIR})$ and $O_3(VOCs_{MIR}+\Delta VOCs)$ are the simulated maximal O_3 19 20 concentrations with the VOCs concentration same as that under the MIR conditions and with an increased quantity of VOCs ($\Delta VOCs$), respectively. Therefore, the first step is 21 to determine the MIR conditions including NO concentration and other input 22 parameters of the base scenarios in a specific region. Figure R1 shows the schematic of 23 24 the VOCs' incremental reactivity (IR) as a function of NO concentration. Thus, the NO 25 concentration under MIR conditions is the value that corresponds to the maximal IR. Other input parameters include the concentrations of VOCs, O₃, SO₂, CO, and HONO, 26 and the meteorological parameters in the base scenarios are usually selected according 27 to their median or mean values in ozone pollution events. Therefore, the MIR values of 28 VOCs depend on the meteorological conditions, the components of VOCs, and the 29

30 concentrations of other pollutants even if the O₃ formation is VOCs-sensitive as 31 required by the MIR method. In this work, we performed OBM simulations when 32 discussing the influence of photochemical loss of VOCs on ozone formation. Thus, it 33 reflected the real atmospheric conditions during our observations.



34

35

36

37

Figure R1. Schematic of VOCs' incremental reactivity as a function of NO concentration.

We added the sentence "In addition, the MIR values of VOC species for a specific 38 region are calculated with the base scenario, in which NO concentration and other 39 parameters are the values that correspond to the maximal incremental reactivity (IR). 40 The fixed MIR values of different VOCs can neither reflect the non-linear relationship 41 42 between ozone and VOCs, involving in the complicated radical recycling (OH-RO₂-RO-HO₂-OH) related to the production of ozone, nor be used for analyzing the radical 43 budget of the initial VOCs concentration. Thus, a quantitative analysis is necessary to 44 explicitly understand the influence of photochemical loss of VOCs on ozone formation 45 and its mechanisms based on OBM studies, in which the dynamic atmospheric and 46 meteorological conditions is accounted for." in lines 93-102 in the revised manuscript. 47 48

49 2. I didn't see Fig. R3 from the response in the SI. I thought this figure was useful both

as a check on the usability of the xylene/ethylbenzene clock and the uncertainty due to
the choice of clock, and recommend the authors include it in the supplement.

52 **Response:** Thank you for your good suggestions. We have added this figure (Figure 53 R2) as Figure S5 in the revised SI and updated the sentences "3) the calculated PICs 54 were in good agreement with those calculated using other tracers, such as i-55 butene/propene (Figure S5) (Zhan et al., 2021)." in lines 172-174 in the revised 56 manuscript.



57

58

Figure R2. Comparison of PICs calculated for xylene/ethylbenzene and i-

- Butene/Propene (Zhan et al., 2021). Error bars are standard deviations.
- 60

59

3. For the comparison between the NMS and OS sites, was there a reason why the comparison could only be done for 1 day? If you could use a larger subset of the campaign, you could get a better statistical distribution of the error on this method. If there are clear reasons why this is the only viable day (e.g. the winds never placed the NMS site downwind of the OS site on other days), then it would be good to state that. **Response:** Thank you for your comments. We agree with you that a larger subset will better present the statistical distribution of the error on this method. However, after we 68 checked the wind trajectory of the whole campaign, no other days could match the two

69 sites well. This requires longer observations in the future.

70

4. Following #3, I recommend adding this comparison to the conclusion, as that is an
important piece of information for readers to take away.

Response: Thank you for your good suggestions. According to your suggestion, we added the sentence in lines 466-469 "And the mean ozone concentration of downwind site was 27.6 ppb day⁻¹ higher than the observation site, slightly lower than the difference (~36 ppb day-1) between PIC-VOCs and observed VOCs, which indirectly supported the accuracy of the above results." in the revised manuscript.

5. In lines 230-231, what is meant by "second reaction rate"? Does this mean the second
of two possibilities (e.g. NO + RO2 -> NO2 + RO vs. NO + RO2 -> RONO2) or second
order reactions (i.e. reactions with two reactants)? If the former, I recommend a
different way to describe the reactions, as the order in which reactions are listed isn't
consistent in every source.

Response: Thank you for your good suggestions. The "second reaction rate" is the second-order reaction rate constant. We have corrected this to "the second-order reaction rate constants" in lines 230-231 in the revised manuscript. We also checked the entire text.

97 Review 2 #

98 The manuscript has been much improved with the revisions. I feel the reviewer 99 comments were thoughtfully considered and addressed where possible. The areas of 100 uncertainty remaining have been discussed adequately.

101 **Response:** Thank you for your positive comments and kind help.

102

I would recommend rephrasing the revised lines 441-446 with the help of a native 103 104 English speaker. Maybe, these sentences could be considered. "This was mainly because, under stable conditions, the nighttime residual layer (RL) is isolated from 105 mixing with the nighttime surface layer. The RL layer usually contains an air mass with 106 a higher ozone mixing ratio than in the surface layer. In the morning, surface heating 107 causes mixing upward in the surface layer until the temperature inversion is eroded 108 away and rapid mixing of pollutants throughout the surface and boundary layer occurs." 109 Response: Thank you for your good comments and suggestions. We have rephrased 110 the sentences according to your suggestion in lines 441-446 in the revised manuscript 111 "This was mainly because, under stable conditions, the nighttime residual layer (RL) is 112 isolated from mixing with the nighttime surface layer. (Tan et al., 2021). The RL layer 113 usually contains an air mass with a higher ozone mixing ratio than in the surface layer. 114 In the morning, surface heating causes mixing upward in the surface layer until the 115 temperature inversion is eroded away and rapid mixing of pollutants throughout the 116 surface and boundary layer occurs.". 117

118