Response to Reviewer #1

We sincerely appreciate the valuable comments provided by the reviewer, which greatly contributed to enhancing the quality of the paper. Detailed responses are shown below. The reviewer's comments are in regular font, and the author's responses are in red. The changes made to the text are highlighted in blue. The corresponding contents have been updated in the manuscript.

Reviewer 1:

Summary:

Naphthalene (Nap) and its derivatives are important intermediate volatile organic compounds (IVOCs) contributing to the formation of secondary organic carbon (SOC). This manuscript uses the CMAQ model to investigate the impacts of Nap and methylnaphthalene (MN) on the formation of SOC and other secondary pollutants in the YRD region. Overall, the manuscript is well-written and easy to follow. The results are interesting and meaningful. I recommend accepting this manuscript after some minor revisions.

Response: Thank you so much for taking the time to thoroughly review our manuscript. Your valuable comments are greatly appreciated and helpful in improving the quality of the manuscript. We have carefully considered your comments and made revisions accordingly. To enhance the clarity, we have changed 'base1' to 'base_zeroNapMN'. Additionally, we conducted a new scenario (base_zeroMN) where the emissions of 1-methylnaphthalene (1-MN) and 2-methylnaphthalene (2-MN) in case-1product were set to zero to quantify the individual impacts of naphthalene (Nap) and methylnaphthalene (MN). We found a mistake in calculating the emissions of Nap and MN from transportation and residential sources in the MEIC inventory for the surrounding area of YRD. Thus, we re-simulated the model using corrected emissions and updated the entire manuscript accordingly. These corrections only have minor influences on the results, and the conclusions remain unchanged. Detailed point-to-point responses are shown below.

Minor comments:

1. Lines 29-30: Does the 3.1% contribution refer to total methylnaphthalene?

Response: Yes, the total contribution from 1-MN and 2-MN was 3.1%. After corrections to the naphthalene and methylnaphthalene emissions from transportation and residential sources in MEIC, the total contribution is 2.4%. We have revised the text as follows:

Lines 29-31: "The concentrations of 1-MN and 2-MN were relatively low, averaging at 2 ppt and 5 ppt. Together, they accounted for only 2.4% of the aromatic-derived SOC."

2. Lines 154-156: Why only the anthropogenic emissions of Nap and MN were scaled

in the emis-adjust case? According to Figure S3, the Nap and MN emissions in the YRD regions are much lower than those in other regions. Could you please show the difference between the MEIC and the YRD emission inventories, and add a brief discussion about such uncertainty?

Response: Thank you for pointing this out. We apologize for the mistake in calculating the Nap and MN emissions from transportation and residential sources in the MEIC inventory, which resulted in an overestimation of the emissions in the surrounding area of YRD. We have corrected this error and updated regional distributions of Nap and MN emissions as shown in Figure R1 (Figure S2 in the revised Supplement). It is worth noting that there are no significant differences between the emissions of YRD and its surrounding regions (MEIC). Since Nap and MN primarily originate from anthropogenic sources (86.7% for Nap and 76.0% for MN), we have only adjusted their anthropogenic emissions in the emis-adjust case.

The model has been re-run with corrected emissions, and the results as well as figures and tables have been updated. Additionally, the text has been revised and a brief discussion about the uncertainty has been added as follows:

Lines 25-27: "Constrained by the observations, anthropogenic emissions of Nap and MN in the entire region were multiplied by 5 and 7, respectively, to better capture the evolution of pollutants."

Lines 158-160: "Considering their predominantly anthropogenic origin, their anthropogenic emissions in the entire region from emis-orig were multiplied by 5 and 7 respectively in the emis-adjust case."

Lines 168-170: "It should be noted that uncertainties associated with the emission inventory and source profiles, which are based on sector-specific mass ratios presented in Table S2, may potentially affect both the distribution and source contributions of Nap and MN."

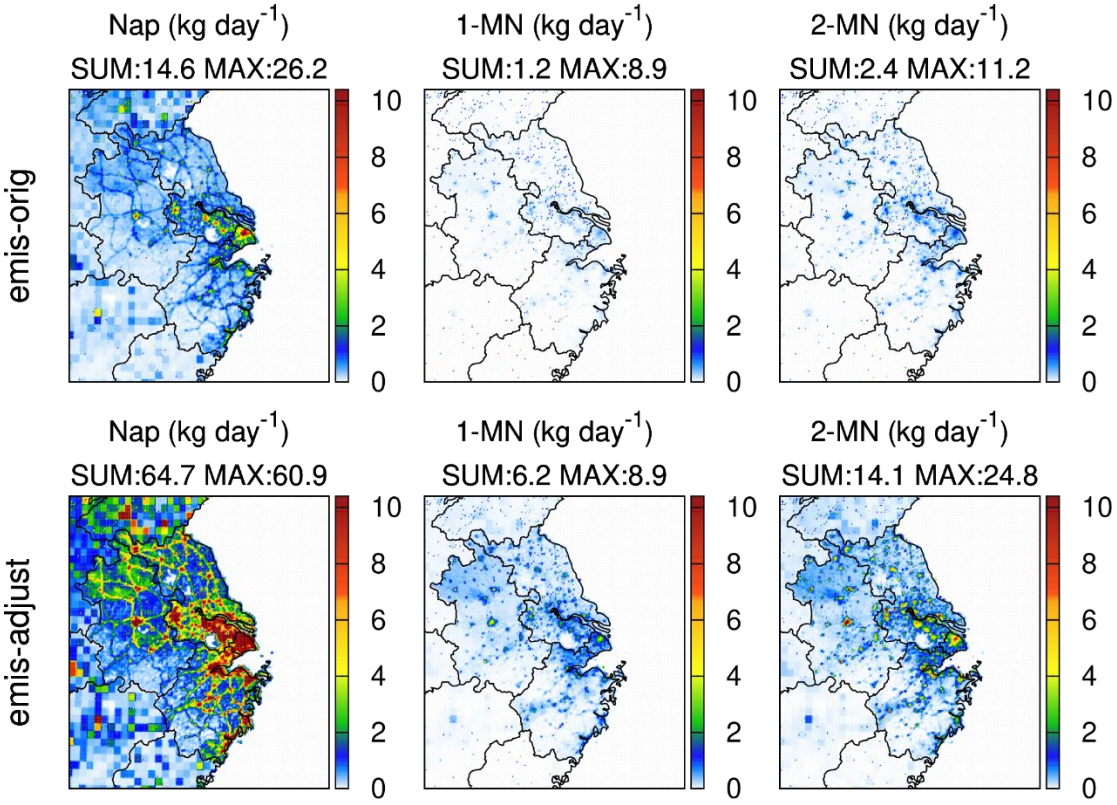

Figure R1. Regional distributions of Nap, 1-MN, and 2-MN emissions in emis-orig and emis-adjust. SUM represents the total emission rate (tons day^{-1}) over the YRD region. MAX represents the maximum emission rate (kg day^{-1}) in the grids of the YRD.

3. Lines 160-161: Is the total Nap and MN emission rate over the YRD region 0.9 kg day-1? Please verify the numbers.

Response: Thank you for pointing this out. 0.9 kg day^{-1} refers to the average emission rate of each grid over the YRD region. To enhance clarity, we have calculated the total emission rate of the YRD region and denoted it as 'SUM' in Figure R1 (Figure S2 in the revised Supplement) as above. The text has also been modified as follows:

Lines 162-165: "After adjustments, the total emission rate of Nap and MN in the YRD region in emis-adjust (85.0 tons day^{-1}) was approximately 4 times higher than that in emis-orig (18.2 tons day^{-1}). The total MN emission rate in the YRD region in emis-adjust was 20.3 tons day^{-1}, lower than that of Nap."

4. Lines 195-196: Could you clarify the meaning of "the original settings" mentioned here?

Response: The term "the original settings" refers to the results of case-1product-orig and case-2product-orig simulated with emis-orig that Nap emissions in the YRD were based on the 2017 YRD inventory, while Nap emissions in the rest of the domain and MN emissions of the entire domain were calculated with sector-specific mass ratios and total emissions of non-methane volatile organic compounds. To make this clear, we

have revised the text and Table 1 (Table R1) as follows:

Lines 205-207: “The concentrations of Nap in case-1product-orig and case-2products-orig were significantly underestimated by 79% compared to the observations.”

Table R1 Settings of the scenarios.

Case	Emission setting	SOA parameterization for MN
case-1product-orig	Nap emissions in the YRD were based on the 2017 YRD inventory; Nap emissions in the rest of the domain and MN emissions in the entire domain were calculated using sector-specific mass ratios and total emissions of non-methane volatile organic compounds (emis-orig)	one-product method
case-2products-orig		two-product method
case-1product	The anthropogenic emissions of Nap and MN in the entire domain from emis-orig were multiplied by 5 and 7, respectively (emis-adjust)	one-product method
case-2products		two-product method
base_zeroNapMN	Emissions of Nap and MN were set to zero based on emis-adjust	one-product method
base_zeroMN	Emissions of MN were set to zero based on emis-adjust	one-product method

5. Lines 199-200: It is recommended to change the units for 1.40E-2 ppb and 1.50E-2 ppb to ppt.

Response: Thank you for the advice. We have revised the text and replotted Figure 2 (Figure R2) and Figure S3 (Figure R3) based on the updated results as follows:

Lines 209-211: “The modeled concentration of MN by emis-adjust (14.0 ppt) was also comparable to the observed value (15.0 ppt) and showed a good correlation between the two (r=0.59).”

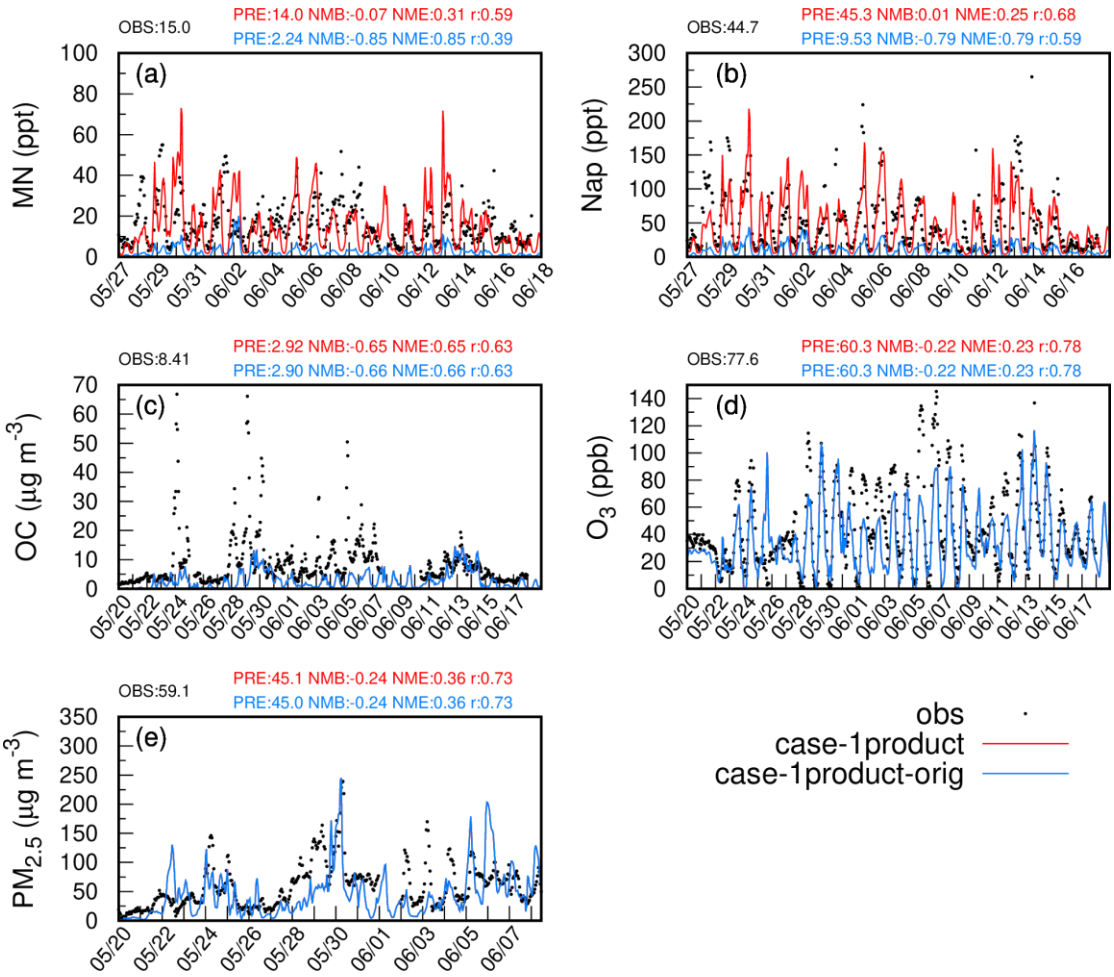

Figure R2. Observed and simulated hourly concentrations of MN, Nap, OC, $PM_{2.5}$, and O_3 based on emis-adjust (red) and emis-orig (blue) at the Taizhou site. Model performances for daily MN, Nap, OC, $PM_{2.5}$, and MDA8 O_3 are shown in blue for case-1product-orig and in red for case-1product. OBS and PRE represent the average of observations and predictions, respectively. Note that the red and blue lines overlap in (c)-(e).

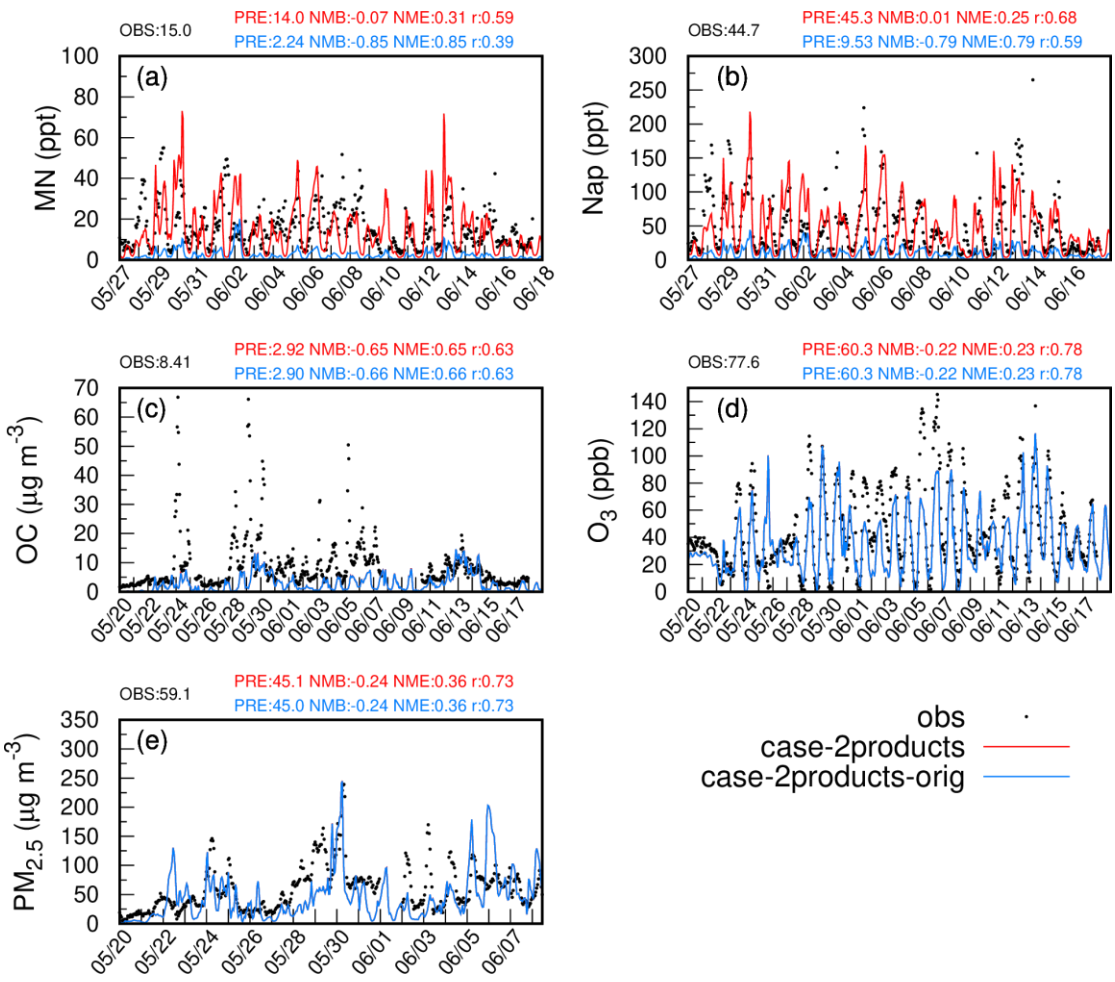

Figure R3. Observed and simulated hourly concentrations of MN, Nap, OC, $PM_{2.5}$, and O_3 based on emis-adjust (red) and emis-orig (blue) at the Taizhou site. Model performances for daily MN, Nap, OC, $PM_{2.5}$, and MDA8 O_3 are shown in blue for case-2products-orig and in red for case-2products. OBS and PRE represent the average of observations and predictions, respectively. Note that the red and blue lines overlap in (c)-(e).

6. Lines 201-202: According to Figure 1, the simulated concentrations of OC and $PM_{2.5}$ were nearly identical for both cases. Therefore, the term "improved" may not be appropriate here.

Response: Thank you for your advice. We have modified the text as follows:

Lines 211-213: "For other species, the concentrations of OC and $PM_{2.5}$ were slightly increased in emis-adjust compared to that of emis-orig, although they were underestimated in both scenarios."

7. Lines 269-272: There is no significant difference in the simulated OC concentrations between case-1product and case-2products.

Response: Thank you for pointing this out. We have modified the corresponding descriptions in the main text as follows:

Lines 284-287: "In general, the concentrations of SOC produced by the three PAHs in case-1product were higher than that in case-2products, exhibiting similar spatial distribution patterns in both cases. We will focus on the results from case-1product in the subsequent analysis."

8. Lines 294-302: I'm curious about the diurnal variations in O_3 and radicals. Could you provide more details?

Response: Thank you for the advice. The diurnal variations in O_3, radicals, and SOC at the two sites have been included in the Supplement as Figure S12 (Figure R4). A brief description of the diurnal changes has been added in the revised text as follows:

Lines 315-318: "It was found that both OH· and HO_2· displayed bimodal variations at the two sites, with the most pronounced changes of 0.7–1.0% and 1.6–2.2% occurring in the morning, respectively (Fig. S12). The concentrations of SOC and O_3 were elevated in the daytime, reaching peak increments of 2.1–2.3% and 0.4–0.5% at noon."

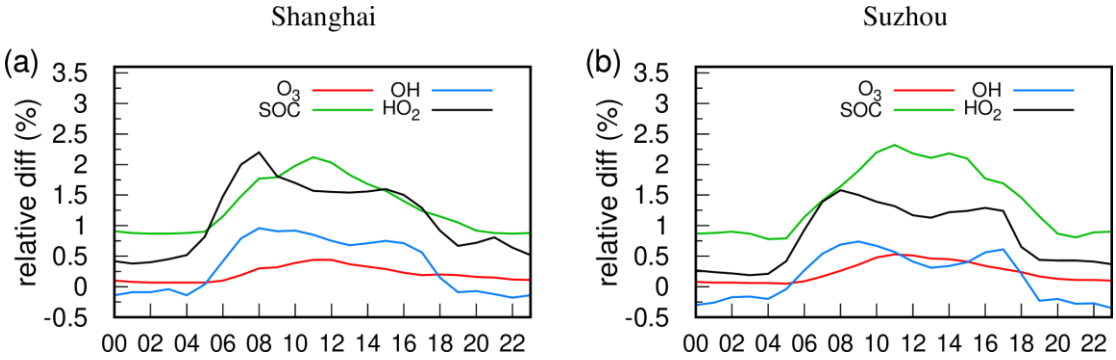

Figure R4. Diurnal relative changes in case-1product compared to base_zeroNapMN in (a) Shanghai and (b) Suzhou.

9. Figure 4: Please check the line length in the color bar ticks.

Response: Thank you for the reminder. We have updated Figure 4 (Figure R5) accordingly, which is now presented as Figure 5 in the revised manuscript.

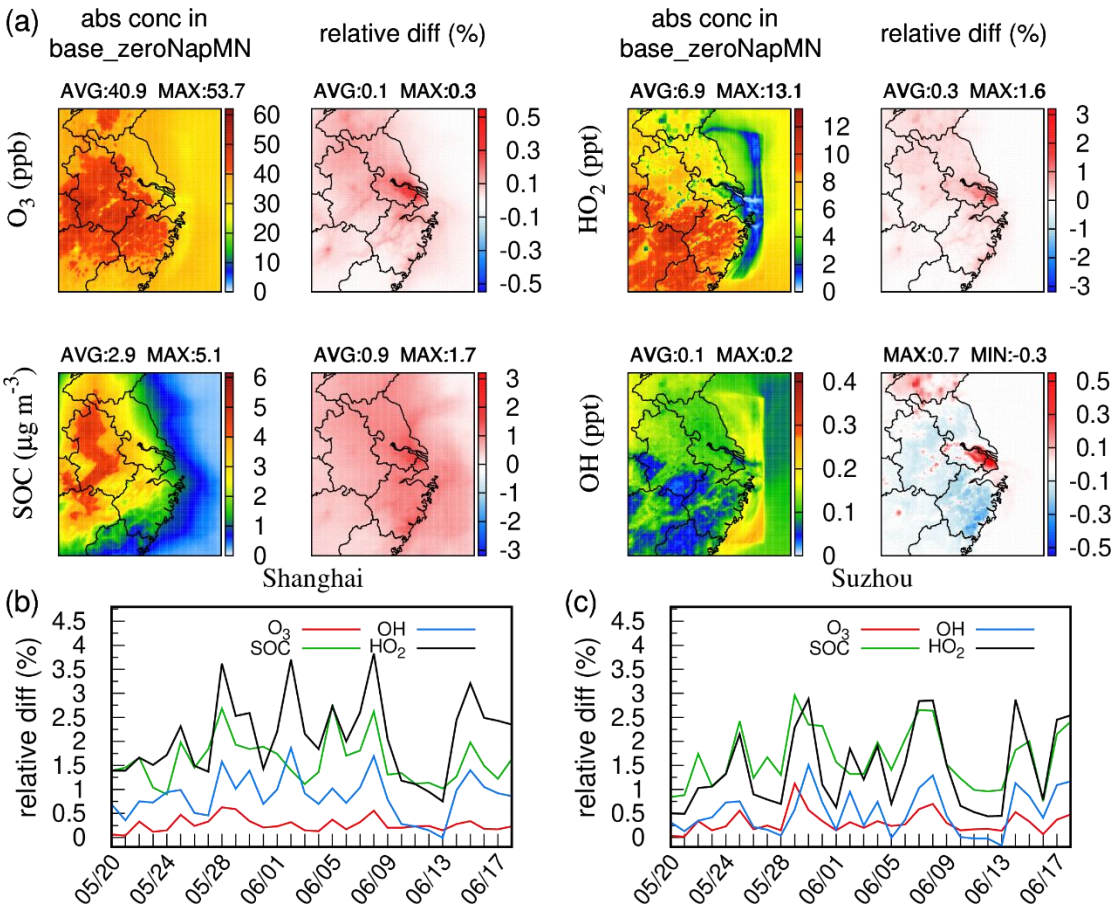

Figure R5. (a) Average concentrations of SOC, O_3, OH·, and HO_2· in base_zeroNapMN and changes in case-1product relative to base_zeroNapMN. Daily relative changes in case-1product compared to base_zeroNapMN in (b) Shanghai and (c) Suzhou.