

Point-by-point responses to the comments & suggestions from the editor and reviewers

Manuscript ID: EGUSPHERE-2026-1101

Title: “Direct thermal enhancement dominates over emission-mediated pathways in heatwave-induced O₃ and SOA increases across China”

We appreciate the comments from the editor and reviewers on this manuscript. We addressed the comments and suggestions carefully and made necessary changes as shown in the marked revision of the manuscript. The detailed point-by-point responses are listed below in blue and modifications in the manuscript are in red.

Reviewer 1

The manuscript addresses an important and timely question: how heatwaves enhance O₃ and SOA through direct meteorological effects and emission-mediated pathways. The topic fits well within the journal scope. The WRF-CMAQ sensitivity design is generally appropriate, and the main conclusion that direct high-temperature meteorological effects dominate the national O₃/SOA increases is interesting and policy relevant. The regional finding that BVOC-related effects are particularly important for O₃ in the VOC-limited North China Plain is also valuable. I recommend publication after minor revisions.

Response: We sincerely thank the reviewer for the positive evaluation and constructive feedback. We have carefully considered all comments and revised the manuscript to clarify our attribution methods, acknowledge observational and methodological limitations, and improve figure readability. Detailed point by point responses and corresponding manuscript modifications are provided below.

1. The attribution method should be explained more clearly. The authors state that nonlinear interactions lead to a residual term and that percentage contributions are normalized against the sum of absolute changes from separated scenarios. This treatment may influence the reported “over 80%” meteorological contribution, so a clearer explanation or sensitivity discussion is needed. See p. 4, lines 101–105, and p.

1, lines 18–21.

Response: We thank the reviewer for this comment. In the CMAQ model, secondary pollution formation is highly nonlinear. When high temperatures and elevated biogenic emissions occur simultaneously in the BASE22 scenario, they create synergistic chemical interactions. Consequently, the sum of the absolute concentration changes from the three isolated sensitivity scenarios (MET22, BVOC22, SNO22) does not exactly equal the total interannual difference. This mathematical gap is the residual term, which represents the overlapping synergistic effects and typically accounts for a minor fraction of 3%-5%.

To accurately evaluate the relative weight of each pathway within a closed 100% framework, we normalized the individual contributions against the sum of their absolute changes. This specific treatment ensures that the overlapping synergistic residual is distributed proportionally among the drivers rather than arbitrarily assigned to one. Figure 3 in the manuscript confirms that the unnormalized increments driven by meteorology (the MET scenario) are inherently and vastly larger than those from isolated emission variations (the BVOC and SNO scenarios) across most region. We have updated section 2.3 to include this explanation and revised the abstract to state that the over 80% contribution is a normalized value.

Changes in manuscript (lines 18-19): “Results show that high temperatures drive over 80% (normalized) of the O₃ and SOA increases nationally, primarily through favorable weather conditions and enhanced atmospheric oxidation capacity.”

(lines 104-110): “Due to the nonlinearity of secondary pollution formation, summing these isolated contributions leaves a residual term when compared to the total difference, which typically accounts for 3% to 5% of the total change across most regions in this study. This term reflects the overlapping synergistic interactions between the high temperatures and elevated emissions concurrently present in the BASE22 scenario. To accurately clarify the calculation sequence, we first determined the raw contribution of each sensitivity case by dividing its isolated concentration change by the total

interannual difference. To eliminate the arithmetic overlap from the residual and establish a closed framework, these individual raw percentages were then normalized against the sum of their absolute values, ensuring the final adjusted contributions of all drivers sum to 100%.”

2. The use of summer 2021 as the baseline for the 2022 heatwave should be discussed as a limitation. Interannual differences may reflect not only heatwave effects, but also circulation variability, emissions, and boundary conditions. A short discussion, or comparison with a multi-year baseline if available, would strengthen the attribution. See p. 4, lines 90–99.

Response: We thank the reviewer for this constructive suggestion. The summer of 2021 was selected as the baseline for two major reasons. First, anthropogenic emissions in China were highly comparable and remained stable between 2021 and 2022 due to continuous emission control policies, which minimizes the interference of emission fluctuations in our attribution system. Second, although 2021 experienced notable warmth, the 2022 heatwave was an unprecedented, record-breaking extreme event across China (Wang et al., 2024; Sun et al., 2022). Compared to the extraordinary severity of 2022, the meteorological conditions in 2021 provide a relatively moderate control case to isolate the impacts of this rare extreme heatwave. We recognize that using a single year baseline includes interannual weather variability, and we have added a brief discussion in section 4 to state this limitation.

Changes in manuscript (lines 316-322): “While scenario analysis successfully isolates the key drivers of the 2022 anomaly, using the summer of 2021 as the sole baseline cannot completely separate the heatwave from background interannual weather variability. Nevertheless, this selection is justified by the highly comparable anthropogenic emissions between 2021 and 2022 resulting from continuous national emission control policies, which minimizes the interference of emission fluctuations. Furthermore, although 2021 also experienced high temperatures, the 2022 heatwave remains an unprecedented extreme event across China. The atmospheric conditions in

2021 therefore provide a relatively moderate control case to highlight the impacts of this rare extremity. Future studies should use a multi-year baseline to better separate localized extreme impacts from background climate variability.”

3. The uncertainty in MEGAN-based BVOC and SNO emissions should be discussed more explicitly, since these emissions are central to the indirect pathway analysis. This is especially important for regional conclusions regarding NCP BVOC sensitivity and SNO effects. See p. 3, lines 75–82, and p. 5, lines 106–120.

Response: We thank the reviewer for this constructive comment. Previous studies show that while MEGAN generally overestimates regional BVOC emissions, it frequently underestimates them in urban areas due to coarse vegetation data that fail to capture high-emitting urban greening (Zhang et al., 2025). Besides, MEGAN typically underestimates SNO emissions because it oversimplifies agricultural fertilizer inputs (Oikawa et al., 2015; Huang et al., 2023). Considering the geographical landscape of the NCP, which features dense urban clusters intertwined with agricultural lands, it is plausible that both urban BVOC and agricultural SNO emissions during the heatwave were higher than simulated. We have expanded section 3.5 to suggest that, indirect pathways in the NCP likely play a larger role than our estimates.

Changes in manuscript (234-238): “These estimated impacts of biogenic emissions in the NCP likely represent a conservative estimate. Specifically, coarse vegetation data and simplified fertilizer parameterizations lead the MEGAN model to underestimate urban BVOCs and SNO emissions, respectively (Zhang et al., 2025) (Oikawa et al., 2015; Huang et al., 2023). As the NCP features dense urban clusters interspersed with intensive agricultural lands, the actual emissions of both precursors and their subsequent contributions to secondary pollution during the heatwave are expected to exceed the current simulations.”

4. The model evaluation is acceptable, but the SOA-related conclusions would benefit from clearer validation or caveats. The current evaluation focuses mainly on

meteorology, O₃, PM_{2.5}, and NO₂, while direct observational constraints on SOA are not clearly shown. Please clarify whether OA/SOA observations are available, or explicitly state this limitation. See p. 5–6, lines 121–135.

Response: We thank the reviewer for this constructive suggestion regarding SOA validation. The evaluation is restricted to criteria pollutants because a systematic and continuous nationwide monitoring network for OA or SOA components is currently unavailable in China. Consequently, direct observational constraints on the simulated SOA concentrations are unfeasible for this study. As suggested, we have explicitly acknowledged this observational limitation and the associated uncertainties of SOA simulations in the revised manuscript.

Changes in manuscript (lines 140-144): “However, a direct evaluation of SOA is currently unfeasible due to the absence of a nationwide monitoring network in China. Furthermore, previous studies reported that air quality models generally underestimate SOA concentrations because of missing precursor emissions, such as semivolatile/intermediate VOCs, and unrepresented multigenerational oxidation pathways (Zhao et al., 2022; Li et al., 2026). Consequently, the actual amplification of SOA caused by high temperatures is likely more severe than the model estimates.”

5. The nighttime O₃ mechanism is interesting but could be clarified. The explanation involving NO₃ chemistry, reduced NO titration, and boundary-layer stability should better distinguish chemical effects from vertical mixing or transport effects. Additional diagnostics such as nighttime PBL height, NO_x, NO₃, or vertical diffusion changes would be helpful if available. See p. 10–11, lines 220–225.

Response: We sincerely appreciate this constructive feedback. We have added a new figure (see **Fig.1** below) showing the diurnal variations of PBL height, NO₃ radical, and NO_x to illustrate the O₃ diurnal changes. Figure 1 shows a nocturnal decrease in PBL height, which confirms that vertical mixing is suppressed and traps pollutants near the surface. Within this stable layer, chemical processes determine the O₃ trends. Figure 4b in the manuscript shows that weather driven biogenic emissions (BVOC22 case) cause

a net reduction in nighttime NO_3 radical concentrations, which indicates the chemical consumption of NO_3 radicals by BVOCs. This process changes the nocturnal nitrogen cycle and reduces NO for O_3 titration. Therefore, as shown by the positive biogenic contribution in Figure 4a, the reduced titration preserves residual O_3 and leads to its nighttime accumulation.

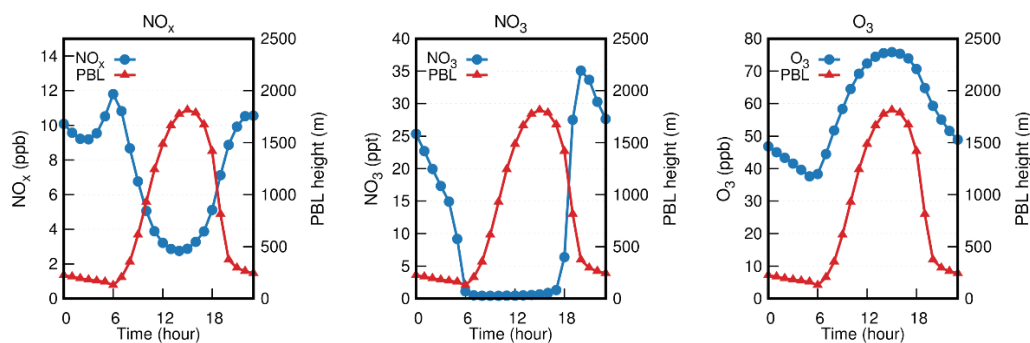


Figure 1. Diurnal variations of key chemical species and PBL height in the NCP under the BVOC22 scenario.

Changes in manuscript (lines 228-234): “Moreover, the impact of the BVOC22 on NCP O_3 exhibits a diurnal reversal, from negative in the daytime to positive at night (**Fig. 4**). During the day, an increase in BVOCs enhanced the oxidation of NO to NO_2 , and further facilitated the formation of O_3 . However, the concurrently increased NO from NO_2 photolysis also intensifies the O_3 loss via titration ($\text{O}_3 + \text{NO}$). When the titration process became slightly dominant, this could lead to a net negative daytime contribution to O_3 . Conversely, at night, the suppressed PBL height limits vertical mixing and traps pollutants (**Fig. S6**). Within this stable layer, increased BVOCs promoted NO_3 radical chemistry, which is verified by the net decrease of nighttime NO_3 concentrations (**Fig. 4b**). This process consumed NO and thus reduced the nocturnal O_3 titration, facilitating residual O_3 accumulation.”

6. Some figures contain dense information and relatively small labels. Improving figure readability and keeping scenario names consistent would make the manuscript easier to follow, especially for Figs. 3–6.

Response: We thank the reviewer for the suggestions on figure quality. Figures 3-6

have been updated to increase the font sizes of all necessary labels, legends, and axis titles. We have also verified that the scenario abbreviations (B for BASE, M for MET, V for BVOC, and N for SNO) are used consistently across all panels, labels, and captions.

Overall, this is a useful and timely study. After addressing the above minor issues, the manuscript should be suitable for publication.

Response: We sincerely thank the reviewer for the positive overall evaluation and the highly constructive feedback. We have carefully addressed all the raised issues in the revised manuscript, which we believe has significantly improved the quality of our study.

Reviewer 2

This manuscript presents a well-designed modeling study on the drivers of O₃ and SOA increases during heatwaves in China. The experimental design is appropriate, and the findings regarding the dominant role of direct temperature effects and the heightened VOC sensitivity in the North China Plain are valuable for understanding climate air quality interactions. The paper is generally well written and organized. However, several minor issues remain that should be addressed before publication. Overall, the manuscript is suitable for acceptance by ACP after minor revision.

Response: We sincerely thank the reviewer for the positive evaluation of our work. We have carefully addressed all of your comments and incorporated the suggested changes into the revised manuscript. Detailed point by point responses are provided below.

1. In the BVOC22 sensitivity simulation, the authors state that isoprene, terpenes, and sesquiterpenes are selected as the dominant BVOC species. Please provide the relative emission proportions of these three groups under the baseline (2021) and heatwave (2022) conditions. This information would help readers assess which biogenic precursors are most responsible for the observed O₃ and SOA changes.

Response: We thank the reviewer for this insightful suggestion. Based on the model outputs, the relative emission proportions of BVOCs remained highly consistent between the 2021 baseline and the 2022 heatwave conditions (see Fig. 2 below). Under the 2021 conditions, isoprene, terpenes, and sesquiterpenes accounted for approximately 81.8%, 15.5%, and 2.6% of the total emissions, respectively. Under the 2022 conditions, these proportions shifted only slightly to 82.8%, 14.7%, and 2.5%. This indicates that while the absolute emission amounts increased during the heatwave, the primary precursor composition driving the O₃ and SOA changes remained stable. We have revised section 2.3 to help readers better assess the biogenic precursors.

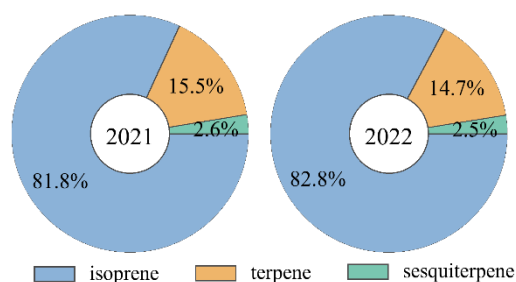


Figure 2. Components of biogenic terpenoid emissions. Contribution of isoprene, terpene and sesquiterpene to total biogenic terpenoids emissions from 2021 to 2022 in China.

Changes in manuscript (lines 81-83): “Based on the MEGAN estimates, the relative proportions of isoprene, terpenes, and sesquiterpenes are highly consistent between the 2021 baseline and 2022 heatwave conditions (**Fig. S1**). They account for 81.8%, 15.5%, and 2.6% of the total BVOC emissions in 2021, and 82.8%, 14.7%, and 2.5% in 2022, respectively.”

2. Section 3.6 reports that the PRD region has the highest O₃ temperature sensitivity, yet Figure 1 shows that summer temperatures in the PRD slightly decreased from 2021 to 2022. Please clarify how a sensitivity slope can be derived under a net cooling condition. Is the sensitivity estimate driven by spatial variability rather than interannual temperature change? A brief explanation would help readers interpret this result correctly.

Response: According to our methodology, the sensitivity in our study is defined as the linear regression slope calculated across all grid cells within a region, which represents spatial variability rather than an interannual trend. Therefore, although the regional average temperature in the PRD slightly decreased in 2022, the spatial linear regression across the individual PRD grids still yields a highly positive sensitivity slope.

Changes in manuscript (lines 259-261): “As defined in section 2.4, this positive slope represents the spatial linear regression across PRD grids rather than interannual temperature changes. This spatial variability heightens local risks and necessitates targeted mitigation.”

3. Section 3.4 reports that the direct meteorological contribution (MET22) to O₃ in the North China Plain is more pronounced at night than during the day, attributed to temperature inversion trapping surface O₃. However, nocturnal O₃ is typically depleted by NO titration. Please explain why the trapped O₃ is not rapidly titrated by NO under the stagnant, high-pressure conditions described.

Response: We thank the reviewer for this question. The trapped O₃ is not rapidly titrated by NO because the lowered PBL height restricts the vertical diffusion of O₃ and precursors. Within the PBL, these precursors accelerate NO₃ radical chemistry. This pathway consumes NO, which competes with direct O₃ titration (O₃ + NO) and facilitate O₃ to accumulate. We have revised section 3.4 to clarify this mechanism.

Changes in manuscript (lines 228-234): “Moreover, the impact of the BVOC22 on NCP O₃ exhibits a diurnal reversal, from negative in the daytime to positive at night (Fig. 4). During the day, an increase in BVOCs enhanced the oxidation of NO to NO₂, and further facilitated the formation of O₃. However, the concurrently increased NO from NO₂ photolysis also intensifies the O₃ loss via titration (O₃ + NO). When the titration process became slightly dominant, this could lead to a net negative daytime contribution to O₃. Conversely, at night, the suppressed PBL height limits vertical mixing and traps pollutants (Fig. S6). Within this stable layer, increased BVOCs promoted NO₃ radical chemistry, which is verified by the net decrease of nighttime NO₃ concentrations (Fig. 4b). This process consumed NO and thus reduced the nocturnal O₃ titration, facilitating residual O₃ accumulation.”

4. In Figure 2 caption, the difference term is written as “BASE22-BASE2”. It should be “BASE22-BASE21”. Please add the missing digit “1”.

Response: We thank the reviewer for pointing out this typographical error. We have corrected the text in the Figure 2 caption.

Changes in manuscript (lines 172-173): “Figure 2. (a) The differences in simulated BVOC emission rates between BASE22 and BASE21 (BASE22–BASE21, units: mole s⁻¹.”

5. Section 3.4 defines daytime as 8:00 to 20:00 and nighttime as 20:00 to 8:00. Please clarify whether these times refer to local standard time or Beijing Time (UTC+8). Additionally, please confirm whether the model simulations accounted for time zone differences across the study domain (e.g., western China versus eastern China) when aggregating diurnal patterns.

Response: We thank the reviewer for this question. The daytime and nighttime definitions refer to Beijing Time (UTC +8). The model uses the longitude of each grid cell to calculate local solar radiation and photochemistry. For regional aggregation, we used Beijing Time as the unified standard. Since the four selected urban clusters are located in eastern and central China, they geographically align with the Beijing Time zone, minimizing solar time discrepancies. We have explicitly specified this in the Figure 4 caption.

Changes in manuscript (lines 215): “Daytime is defined as 8:00 to 20:00 and nighttime as 20:00 to 8:00 Beijing Time.”

References

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