Response to Anonymous Referee #2

MS No.: egusphere-2025-4017

Title: Decadal Evolution of Aerosol-Mediated Ozone Responses in Eastern China under Clean Air Actions and Carbon Neutrality Policies

This manuscript presents a comprehensive modeling study that evaluates the driving factors controlling aerosol impacts on surface ozone (O_3) response, across two seasons (winter vs summer), to two emission reduction phases that have strategic policy shifts. By separating aerosol effects into aerosol-radiative interactions (ARI) and heterogeneous chemistry (HET), the authors show that summertime O_3 responses are primarily HET- driven, while wintertime responses are mainly driven by ARI. The study also demonstrates how meteorological variability contributes to summertime O_3 responses and projects how these processes may behave under air-quality control strategies. The topic is timely and of clear scientific and societal significance: it advances understanding of the nonlinear nature of photochemical O_3 production and multi-pathway effects of aerosol on this process. It is also of societal significant as the conclusion is informative and understanding the driving factors will help guide emission reduction policy to be more effective and comprehensive. The modeling approach is generally appropriate and carefully implemented. However, the manuscript would benefit from major revisions to improve clarity and to remove ambiguous or potentially misleading wording. I recommend major revision; my detailed comments follow.

Response: We sincerely appreciate the reviewer's thorough evaluation and constructive comments. We have thoroughly revised the manuscript in accordance with these suggestions, which have substantially improved the quality and clarity of the work. Detailed responses to each comment are provided below, with all page and line numbers referring to the clean revised version of the manuscript.

Major comments:

1. The manuscript alternates between two different pairwise comparisons - (a) anthropogenic emissions vs. meteorological variability, and (b) aerosol-radiative interactions (ARI) vs. heterogeneous chemistry (HET) - without clearly stating how these four factors relate to each other. This creates a sense of disconnection in the abstract lines 19-24, the reader sees that "anthropogenic emissions and meteorology dominate winter and summer O_3, respectively" immediately followed by a discussion of ARI vs HET. Please clarify and explicitly state the conceptual framework that links the four factors.

Response: We sincerely thank the reviewer for this insightful and constructive comment. We agree that the manuscript required a clearer articulation of the conceptual framework linking the four factors—anthropogenic emissions, meteorological variability, ARI, and HET. Following the reviewer's suggestions, we have revised the

Abstract, Introduction, and the structure of the Results section to explicitly clarify their hierarchical relationships. Below we summarize our revisions for each sub-point.

(1) In the Abstract, add a short sentence that explains the two comparisons used, like "we separate changes in O_3 into those driven directly by emissions/meteorology and those mediated by aerosol processes", or after the sentence "anthropogenic emissions and meteorological variability respectively dominated winter and summer O_3 increases" (line 19), follow immediately with a short clarifying sentence linking that conclusion to the ARI/HET result.

Response: We thank the reviewer for this constructive suggestion. As suggested, we added a concise clarifying sentence to explicitly link the two dimensions of comparison.

Abstract changes (Page 2, lines 19-20):

"We separate O_3 changes into those driven directly by anthropogenic emissions and meteorological variability, and those mediated by aerosol processes through ARI and HET."

(2) In the Introduction, define the four factors and their roles: anthropogenic emissions and meteorological variability are external drivers that change precursor concentrations and transport; ARI and HET are aerosol-mediated mechanisms that modify photochemistry and how these mechanisms mediate O_3 response to precursor (NO_x) decrease or meteorological variabilities.

Response: We thank the reviewer for this constructive suggestion. In the Introduction, we added a new paragraph that defines the four factors and clarifies their hierarchical roles.

Manuscript changes (Section 1, Page 4, lines 72-77):

"Anthropogenic emissions and meteorological variability act as external drivers that directly regulate precursor concentrations, atmospheric chemical regimes, and transport processes. In contrast, ARI and HET represent aerosol-mediated mechanisms that reshape the photochemical environment by altering photolysis rates and radical budgets. These aerosol-driven mechanisms determine the extent to which surface O_3 responds to precursor (particularly NO_x) reductions or meteorological perturbations. This conceptual framework underpins our separation of O_3 changes into externally driven components and aerosol-modulated components in this study."

(3) In the Results or discussion sections, organize the presentation so that readers first see the partitioning of O_3 responses into contributions from emission reduction vs meteorology variability, and then – for the emission-driven portion – show how aerosol processes (ARI and HET) modulate the response. Could add a schematic to make the logic explicit.

Response: We thank the reviewer for this constructive suggestion. The manuscript already follows this structure: Section 3.1 quantifies the contributions of anthropogenic emission reductions and meteorological variability to O_3 changes, and Sections 3.2

evaluate how ARI and HET further modulate these externally driven O_3 responses. To make this hierarchical relationship clearer, we have added explicit transition sentences at the end of Section 3.1 and at the beginning of Sections 3.2, emphasizing that ARI and HET act as aerosol-mediated modifiers of the emission-driven O_3 changes.

Manuscript changes:

(Section 3.1, Page 11, lines 259-260): “These externally driven O_3 changes provide the foundation for evaluating how aerosol-mediated processes further modulate the emission-reduction-driven portion of the O_3 response.”

(Section 3.2, Page 12, lines 266-267): “Building on the external drivers identified in Section 3.1, we next examined how ARI and HET modified the emission-reduction-driven O_3 response.”

In addition, we have included a simple schematic in the supplement and a description in Results section illustrating the conceptual framework linking external drivers and aerosol-mediated processes, which helps clarify the logic of the analysis. These revisions collectively clarify the conceptual structure and improve readability.

Manuscript changes (Section 3.2, Page 18, lines 368-371):

“Figure S8 illustrated the hierarchical relationships among the four factors analyzed in this section. Emission reductions and meteorological variability constituted the external drivers of O_3 changes, whereas ARI and HET acted as aerosol-mediated modulators that adjust the emission-reduction-driven O_3 responses. This framework motivated our presentation sequence, where external drivers were examined first, followed by the modulation effects of ARI and HET.”

Newly added figure in supplement:

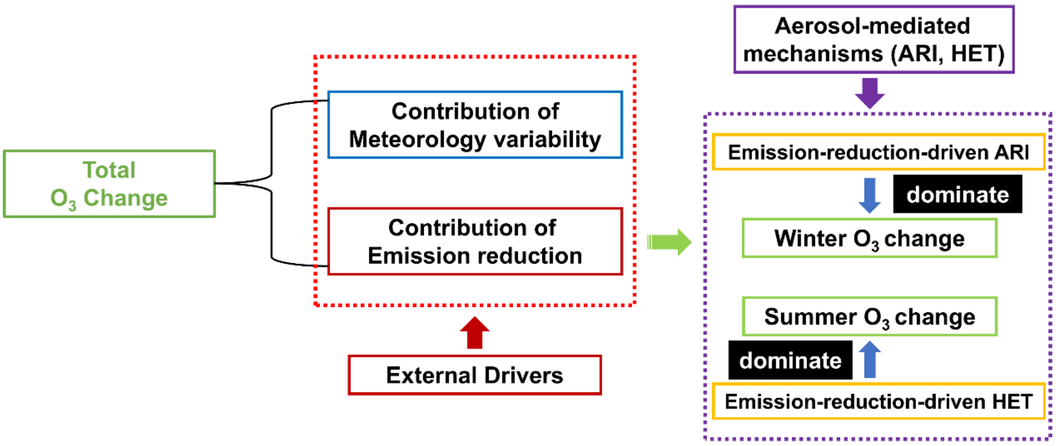

Figure S8. Schematic overview of the analytical framework separating externally driven O_3 changes (from emissions and meteorology) from the aerosol-mediated modulation by ARI and HET, which jointly determine the seasonal and phase-dependent O_3 responses.

2. The Abstract's wording (Lines 22–24) that frames the Phase I–Phase II change in terms of "radical scavenging" is misleading and risks oversimplifying heterogeneous chemistry (HET). Radical uptake by aerosol (i.e., HO_2 scavenging) is a loss pathway for radicals: a reduction in aerosol mass or aerosol liquid water will generally reduce this loss and therefore tends to promote ozone formation. Thus, the statement that the "weakening of this effect during Phase II reduced O_3" is unclear: if the radical-scavenging loss decreases further in Phase II, that would not by itself explain a reduction in O_3. Instead, the reversal in the net HET effect between Phase I and Phase II likely reflects changes in the net balance of multiple heterogeneous pathways (for example, reduced radical uptake and changes in aerosol-mediated production or recycling of reactive nitrogen species such as HONO or $ClNO_2$), together with changes in aerosol liquid water content and the magnitude of aerosol reductions.

Response: We thank the reviewer for this important comment. We agree that describing the Phase I → Phase II change of the HET effect solely in terms of radical scavenging oversimplifies the heterogeneous chemistry represented in WRF-Chem. To address this, we have revised both the Abstract and Section 3.2.

(1) Reword the Abstract lines 22-24 to avoid implying that radical scavenging alone explains the Phase I → Phase II sign change.

Response: We thank the reviewer for this important comment. We agree that the previous Abstract wording unintentionally overemphasized radical scavenging and did not adequately reflect the multicomponent nature of heterogeneous chemistry. To address this concern, we revised the Abstract to clarify that the Phase I → Phase II sign reversal of the HET effect arises from the combined influence of several heterogeneous pathways—not radical uptake alone.

Abstract changes (Page 2, lines 23-26):

"Summer O_3 was more sensitive to HET. In Phase I, aerosol decreases weakened heterogeneous radical uptake, enhancing O_3 formation (+1.62 ppb). In Phase II, however, the net HET effect reversed sign (–2.86 ppb), driven by shifts in multiple heterogeneous pathways—including changes in radical uptake, HONO and N_2O_5 chemistry, and aerosol liquid water—rather than radical scavenging alone."

(2) In the Result section 3.2, when discussing HET roles, include discussion that separates HET into its component effects: radical scavenging, heterogeneous production of reactive nitrogen like HONO and $ClNO_2$, or at least a discussion of the chemical mechanisms used in the model parameterization of heterogeneous chemistry. In addition, a chemical diagnostics for the ozone production/loss terms during phase I and Phase II could also be useful as this allows readers to see which HET component could explain the change of sign of HET impact between phases.

Response: We greatly appreciate the reviewer's valuable suggestions. In response, both Section 2.2 and Section 3.2 have been substantially revised to provide a clearer and more mechanism-based presentation of heterogeneous chemistry (HET) in WRF-Chem.

Section 2.2 has been expanded to introduce, for the first time in this study, the full set of heterogeneous reaction pathways and the updated HET module implemented in our model (detailed descriptions of all HET pathways and parameterizations are provided in Table S1 (response to comment 3)). The revised manuscript now explicitly describes the major heterogeneous pathways represented in the model, including: (1) radical uptake (HO_2, OH, NO_3), (2) NO_2 heterogeneous conversion to HONO and HNO_3, (3) N_2O_5 hydrolysis regulating nighttime NOx partitioning, (4) SO_2 and H_2SO_4 heterogeneous oxidation, and (5) direct O_3 uptake on dust and black carbon surfaces.

Section 3.2 then builds on this framework to explain how the relative contributions of these pathways differ between Phase I and Phase II and how they lead to opposite O_3 responses. Although a full integrated process rate (IPR) analysis was not available for this study, the added diagnostics of HO_2, HONO, and N_2O_5 provide direct and independent evidence supporting the heterogeneous pathways responsible for the sign reversal. The revised discussion therefore offers a mechanistic, process-consistent, and evidence-supported explanation for the observed transition from a positive HET effect in Phase I to a negative effect in Phase II. In subsequent studies, we plan to incorporate a full IPR framework, employ more explicit radical and reactive-nitrogen diagnostics, and further refine the representation of aerosol liquid water and heterogeneous reaction parameterizations—all of which will help strengthen the process attribution and deepen the mechanistic understanding of aerosol-mediated O_3 responses.

Manuscript changes:

(Section 2.2, Page 6, lines 127-134):

"Heterogeneous chemistry exerts complex influences on O_3 formation by altering radical budgets, modifying reactive nitrogen cycling, and changing aerosol-phase reaction rates. In the enhanced WRF-Chem, HET is represented through multiple pathways on dust and black carbon surfaces, including (1) heterogeneous uptake of HO_2, OH, NO_2, and NO_3; (2) nighttime N_2O_5 hydrolysis to $2HNO_3$; (3) heterogeneous formation of HONO from NO_2 uptake on carbonaceous aerosols; (4) SO_2 and H_2SO_4 heterogeneous oxidation; and (5) direct O_3 uptake on dust and black carbon surfaces. These processes collectively modify photolysis-driven radical initiation and NOx partitioning. Therefore, the net HET effect reflects the balance among several aerosol-mediated pathways rather than a single mechanism. The specific heterogeneous reactions and their corresponding uptake coefficients (γ) used in this study are listed in Table S1."

(Section 3.2, Pages 15-16, lines 313-339):

"During Phase I, the substantial reductions in aerosol mass and surface area primarily weakened HO_2 heterogeneous uptake, as indicated by elevated HO_2 (Figure 7d). This reduction in radical loss increased the availability of HO_2 and OH, leading to an enhancement in the photochemical ozone production term $P(O_3)$ (Dyson et al., 2023). In parallel, N_2O_5 also increased during Phase I (Figure S5a), consistent with suppressed heterogeneous hydrolysis under reduced aerosol liquid water (ALW) and diminished aerosol surface area (Brown and Stutz, 2012). The weakened N_2O_5 hydrolysis further limited nighttime conversion of reactive nitrogen to HNO_3, maintaining NOx in more photochemically active forms (Ma et al., 2023b). Meanwhile, heterogeneous NO_2 uptake—an important HONO source—was significantly reduced, consistent with the simulated decrease in HONO (Figure S5d). The reduction in HONO slightly weakened early-morning radical initiation (Yu et al., 2022), but this influence was outweighed by the strong enhancement in HO_2 and the limited conversion of NOx into HNO_3. As a result, HET exerted a net positive contribution to O_3 (+1.62 ppb) in Phase I.

In contrast, Phase II exhibited a fundamentally different chemical response. Although aerosol loadings continued to decrease, the relative importance of heterogeneous pathways shifted substantially. HO_2 declined during Phase II (Figure 7d), indicating a reduced radical pool and weaker propagation of daytime photochemical production. At the same time, N_2O_5 decreased markedly (Figure S5b), suggesting that nighttime NO_3/N_2O_5 chemistry became less effective at sustaining reactive nitrogen cycling under even lower aerosol surface area and ALW. Rather than promoting efficient nighttime NOx recycling, this suppression favored a net loss of reactive nitrogen through terminal sinks (e.g., HNO_3), shifting NOx partitioning toward less photochemically active forms and weakening daytime P(O_3). Conversely, HONO concentrations rebounded during Phase II (Figure S5e). This increase reflects the altered balance between NO_2 uptake and nighttime NOx partitioning under reduced N_2O_5 hydrolysis. However, despite this HONO increase, its positive effect on radical initiation could not compensate for the combined decline in HO_2, weakened N_2O_5 hydrolysis, and enhanced HNO_3 formation (George et al., 2015). The joint effect was a net reduction in the morning radical pool and diminished photochemical O_3 production (-2.86 ppb). This multi-pathway adjustment explains the observed sign reversal of HET's effect on O_3 between the two phases and underscores the importance of considering the full suite of heterogeneous processes—rather than radical uptake alone—when interpreting aerosol-mediated O_3 responses. In future work, we plan to apply integrated process rate (IPR) diagnostics to more directly evaluate how individual heterogeneous pathways—such as HO_2 uptake, HONO formation, and N_2O_5 hydrolysis—shape the resulting O_3 responses. Coupled with continued improvements in heterogeneous chemistry parameterizations and more comprehensive constraints on radical, reactive nitrogen, and aerosol liquid water fields, this will enable a more detailed and process-resolved understanding of phase-dependent O_3 changes."

Newly added figure in supplement:

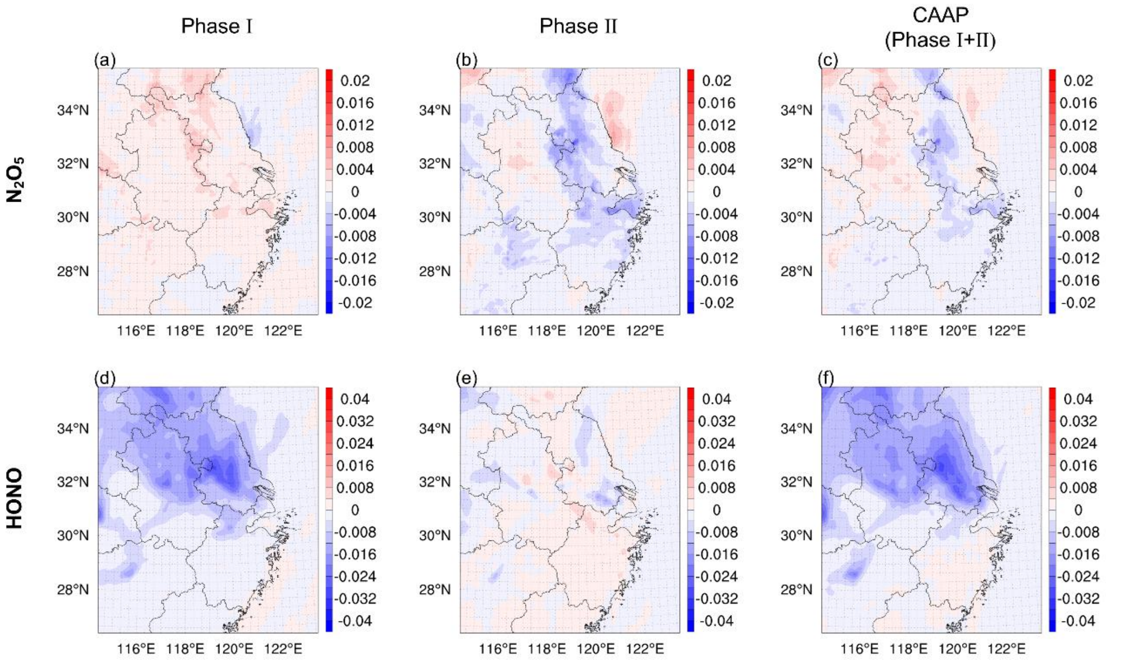

Figure S5 Spatial distributions of N_2O_5 (a-c) and HONO (d-f) concentration (ppb) changes induced by aerosol heterogeneous chemistry (HET) in summer during two phases of the Clean Air Action in the Yangtze River Delta (YRD).

References:

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3. The manuscript correctly notes that uncertainties in heterogeneous chemistry parameterizations could influence the results, but the current treatment is not stated and does not make clear how robust the paper's conclusions are to variations in those parameterizations. Again, HET processes directly modulate reactive-nitrogen recycling (e.g., HONO formation, N_2O_5 hydrolysis), radical budgets (HO_2 uptake), and hence O_3 production regimes; therefore, more explicit discussion and, where possible, quantification of the uncertainty introduced by HET assumptions is essential. By adding discussion of HET impact with more details, it would help.

Response: We sincerely thank the reviewer for raising this important point. We agree that uncertainties in the parameterization of heterogeneous chemistry (HET)—including the uptake coefficients and their dependencies on aerosol liquid water content, acidity, and particle composition—may influence the simulated O_3 responses. We acknowledge that this constitutes an important limitation of the current study and appreciate the opportunity to clarify it more explicitly.

First, although we recognize the scientific value of quantifying these uncertainties through targeted sensitivity simulations, the present work was designed as a multi–phase, multi–season modeling framework that prioritizes consistency across scenarios. Systematically perturbing heterogeneous uptake coefficients would require rerunning the entire experimental suite under multiple alternative chemical configurations, which falls outside the methodological scope and computational design of this study. We have now explicitly acknowledged this limitation in the revised manuscript and clarified why such experiments, while valuable, were not included in the current modeling framework.

Second, in line with the reviewer's helpful suggestion, we have expanded the Supplementary Material (Table S1) to provide a more detailed and transparent description of the heterogeneous pathways newly implemented in our WRF-Chem configuration, particularly those occurring on dust and black carbon surfaces. These additions include the corresponding parameterizations and supporting references, thereby improving clarity and reproducibility.

Third, Section 3.5 (Discussion) has been substantially revised to include a more explicit examination of uncertainties associated with HO_2, NO_2, and N_2O_5 uptake coefficients. We further discuss how variations in these parameters could modify radical budgets, reactive nitrogen cycling, and consequently the magnitude of O_3 responses. While the absolute perturbations may vary under different plausible γ values, the phase-dependent sign reversal of the HET effect is unlikely to change, as it arises from a structural shift in the relative importance of multiple heterogeneous pathways rather than the sensitivity of any single reaction. This robustness is also supported by previous studies (e.g., Shao et al., 2021; Li et al., 2019), which show that different HO_2 uptake coefficients alter the magnitude but not the direction of the O_3 response.

Finally, we fully agree with the reviewer on the importance of more rigorous characterization of HET-related uncertainties. In future work, we plan to incorporate dedicated heterogeneous-chemistry sensitivity simulations together with integrated process rate (IPR) diagnostics to quantitatively evaluate the contribution and robustness of individual HET pathways. These improvements will help further reduce uncertainty and strengthen the mechanistic interpretation of aerosol–O_3 interactions under evolving emission scenarios.

We sincerely appreciate the reviewer's constructive comments, which have allowed us to provide clearer documentation of the heterogeneous chemistry processes included in this study and a more comprehensive evaluation of their associated uncertainties.

Newly added table in supplement:

Table S1. The heterogeneous reactions and the uptake coefficients are considered in our study.

Reaction	Uptake Coefficient γ	Reference
(a) dust		
O_3 (g) → O_3 (ads)	1×10^{-4}	(Bauer et al., 2004)
OH (g) → OH (ads)	$\frac{0.18}{1+(RH\times100)^{0.36}}$	(Bedjanian et al., 2013)
HO_2 (g) → HO_2 (ads)	0.1	(Phadnis and Carmichael, 2000)
H_2O_2 (g) → H_2O_2 (ads)	3.33×10^{-4} (RH<0.15) $3.33\times10^{-4}+\frac{(RH-0.15)\times2.7\times10^{-4}}{0.55}$ (0.15<RH<0.7) 6.03×10^{-4} (RH>0.7)	(Pradhan et al., 2010)
NO_2 (g) → HNO_3 (ads)	5.0×10^{-5}	(Li et al., 2019c)
NO_3 (g) → HNO_3 (ads)	3.0×10^{-3}	(Li et al., 2019c)
HNO3 (g) → HNO_3 (ads)	1.0×10^{-2}	(Liu et al., 2008)
N_2O_5 (g) →2HNO_3 (ads)	3.0×10^{-3} (RH<0.3) $0.0425\times RH-0.00975$ (0.15<RH<0.7) 2.0×10^{-2} (RH>0.7)	(Bauer et al., 2004)
SO_2 (g) →SO_4^{2-} (ads)	1.0×10^{-4} (RH < 0.5) $1.0\times10^{-4}+\frac{RH-0.5}{(1-0.5)\times2\times10^{-4}}$ (RH>0.5)	(Zheng et al., 2015)
H_2SO_4(g) → SO_4^{2-} (ads)	5.0×10^{-2} (RH<0.5) 0.1 (RH>0.5)	(Huang et al., 2014)
(b) Black carbon		

O_3 (g) → O_3 (ads)	$1.8\times10^{-4}\times e^{-\frac{1000}{T}}$	(Tie et al., 2005)
OH (g) → OH (ads)	5.0×10^{-2}	(Slade and Knopf, 2013)
HO_2 (g) → HO_2 (ads)	1.0×10^{-2}	(Saathoff et al., 2001)
NO_2(g) → 0.5HONO + 0.5HNO_3	5.0×10^{-4}	(Lei et al., 2004)
NO_3 (g) → HNO_3 (ads)	3.0×10^{-4} (RH<0.5)	(Saathoff et al., 2001)
	1.0×10^{-4} (RH>0.5)	
N_2O_5 (g) → 2HNO_3 (ads)	4.0×10^{-5} (RH<0.5)	(Saathoff et al., 2001)
	2.0×10^{-4} (RH>0.5)	
HNO_3 (g) → HNO_3 (ads)	1.0×10^{-3}	(Rogaski et al., 1997)

References:

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Manuscript changes:

(Section 2.2, Page 6, lines 127-134):

"Heterogeneous chemistry exerts complex influences on O_3 formation by altering radical budgets, modifying reactive nitrogen cycling, and changing aerosol-phase reaction rates. In the enhanced WRF-Chem, HET is represented through multiple pathways on dust and black carbon surfaces, including (1) heterogeneous uptake of HO_2, OH, NO_2, and NO_3; (2) nighttime N_2O_5 hydrolysis to $2HNO_3$; (3) heterogeneous formation of HONO from NO_2 uptake on carbonaceous aerosols; (4) SO_2 and H_2SO_4 heterogeneous oxidation; and (5) direct O_3 uptake on dust and black carbon surfaces. These processes collectively modify photolysis-driven radical initiation and NOx partitioning. Therefore, the net HET effect reflects the balance among several aerosol-mediated pathways rather than a single mechanism. The specific heterogeneous reactions and their corresponding uptake coefficients (γ) used in this study are listed in Table S1."

(Section 3.5, Pages 24-25, lines 510-530):

"Uncertainties in HET parameterizations also introduce potential variability into the estimated O_3 responses. The uptake coefficients (γ) for HO_2, NO_2, and N_2O_5 depend on aerosol liquid water content, acidity, ionic strength, and particle composition (Jacob, 2000), yet these dependencies remain imperfectly constrained in current atmospheric models. As a result, uncertainties in these parameters may alter the magnitude of individual heterogeneous pathways simulated in this study. For example, higher assumed HO_2 uptake would strengthen radical loss and could reduce the positive HET contribution during Phase I, whereas larger N_2O_5 hydrolysis rates would enhance nighttime conversion of NOx to HNO_3 and potentially intensify the negative HET influence in Phase II. Likewise, uncertainties in NO_2 uptake and HONO yields could modulate early-morning radical initiation and shift the balance between radical propagation and reactive nitrogen recycling.

Importantly, while such uncertainties may influence the absolute magnitude of HET-induced O_3 perturbations, they are unlikely to overturn the direction of the response. Prior modeling studies provide support for this robustness. For instance, Shao et al. (2021) showed that varying γ_{HO2} between 0.2 and 0.08 altered the magnitude of the O_3 increase driven by reduced HO_2 heterogeneous uptake—from approximately 6% (consistent with the ~7% reported by Li et al., 2019a) to about 2.5% during 2013–2016—yet the effect remained positive in all cases. These findings indicate that although heterogeneous uptake assumptions can change the amplitude of the response, the sign of the O_3 change is preserved because the underlying chemical mechanism (reduced radical loss leading to enhanced photochemical production) remains the same. By analogy, the phase-dependent sign reversal identified in our study reflects a structural shift in the competition among HO_2 uptake, N_2O_5 hydrolysis, and HONO formation pathways, and is therefore unlikely to be reversed by plausible uncertainties in individual uptake coefficients. Our future studies will incorporate dedicated sensitivity simulations and integrated process rate (IPR) diagnostics to more systematically quantify how uncertainties in heterogeneous chemistry parameterizations propagate into O_3 simulations. Improvements in observational constraints on aerosol acidity, liquid water content, and heterogeneous reaction rates will further strengthen mechanistic understanding and reduce uncertainty in model-based assessments of aerosol–O_3 interactions under evolving emission pathways."

References:

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4. Choice of O_3 metric (daily mean vs MDA8): The authors justify using daily mean O_3 on the grounds that MDA8 "may underestimate full-day aerosol effects." I disagree that daily mean is a superior diagnostic for separating daytime vs nighttime processes: opposite-signed changes during day and night can cancel in the 24-h mean, obscuring

mechanism interpretation. Therefore, it would help if the authors provided mean diurnal cycles of O_3 (and key chemical drivers such as $P(O_3)/L(O_3)$) for baseline and each phase. These plots will (i) show whether daytime and nighttime responses compensate, (ii) allow comparison with observations for model evaluation, and (iii) improve mechanistic attribution.

Response: We sincerely thank the reviewer for this valuable and constructive comment. We agree that relying solely on daily mean O_3 may mask potential compensating effects between daytime photochemical production and nighttime deposition or titration. In light of this concern, we have removed the original statement claiming that MDA8 may underestimate full-day aerosol effects, as it could be misleading without supporting diurnal diagnostics.

To directly address the reviewer's concern, we have added diurnal-cycle analyses for baseline O_3 and the aerosol-mediated O_3 changes (HET, ARI, and AEs) for Phase I, Phase II, and the overall Clean Air Action period in both winter and summer (Figures S6–S7). These results show that O_3 perturbations across all phases and both seasons are overwhelmingly dominated by daytime changes near the photochemical peak (14–16 LT), whereas nighttime variations are much smaller in magnitude and share the same sign as the daytime responses. Therefore, nighttime effects do not offset or compensate daytime changes. This indicates that the daily-mean O_3 responses presented in the main text reflect genuine daytime-dominant adjustments rather than artifacts of day–night cancellation. A new paragraph summarizing these findings has been added to Section 3.2.

Regarding the reviewer's suggestion to include diurnal variations in $P(O_3)/L(O_3)$, we greatly appreciate this recommendation. In the current modeling framework, however, the integrated process rate (IPR) module was not activated, as the experimental design focused on maintaining a consistent chemical configuration across a large suite of multi-phase and multi-season simulations. Enabling IPR would require rerunning the full experimental set under an alternative chemistry configuration, which falls outside the methodological scope of the present study. We have clarified this constraint in the revised manuscript. As an alternative, we provide diagnostics of key radical and reactive nitrogen species (HO_2, HONO, and N_2O_5), which capture the fundamental processes controlling photochemical production and nighttime nitrogen cycling, and thus offer mechanistic insight comparable to $P(O_3)/L(O_3)$. Further details on these diagnostics and their interpretation are provided in our response to Comment 2.

We sincerely thank the reviewer again for this insightful comment. The additions and clarifications prompted by this suggestion have substantially improved our manuscript by providing stronger mechanistic support and clearer justification of the chosen O_3 metric.

Newly added figure in supplement:

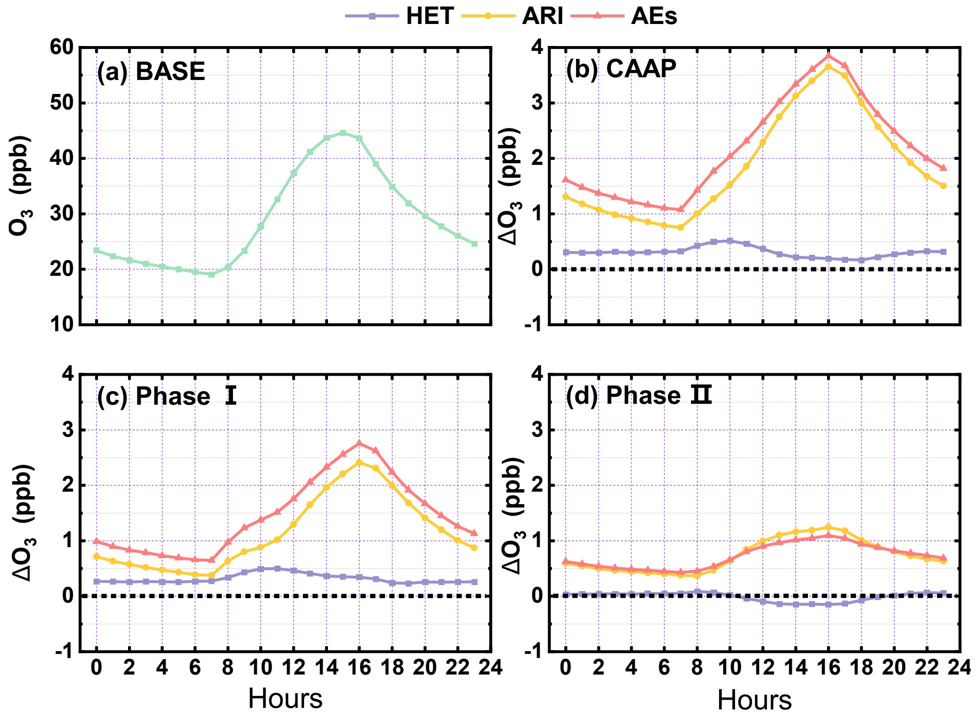

Figure S6. Diurnal variations of (a) baseline O_3 concentrations (ID: 20E20M_AEs) and (b–d) aerosol-induced O_3 changes (ΔO_3) in winter. Panels show the impacts during the overall Clean Air Action period (CAAP), Phase I, and Phase II, each decomposed into heterogeneous chemistry (HET), aerosol–radiation interactions (ARI), and their combined effects (AEs).

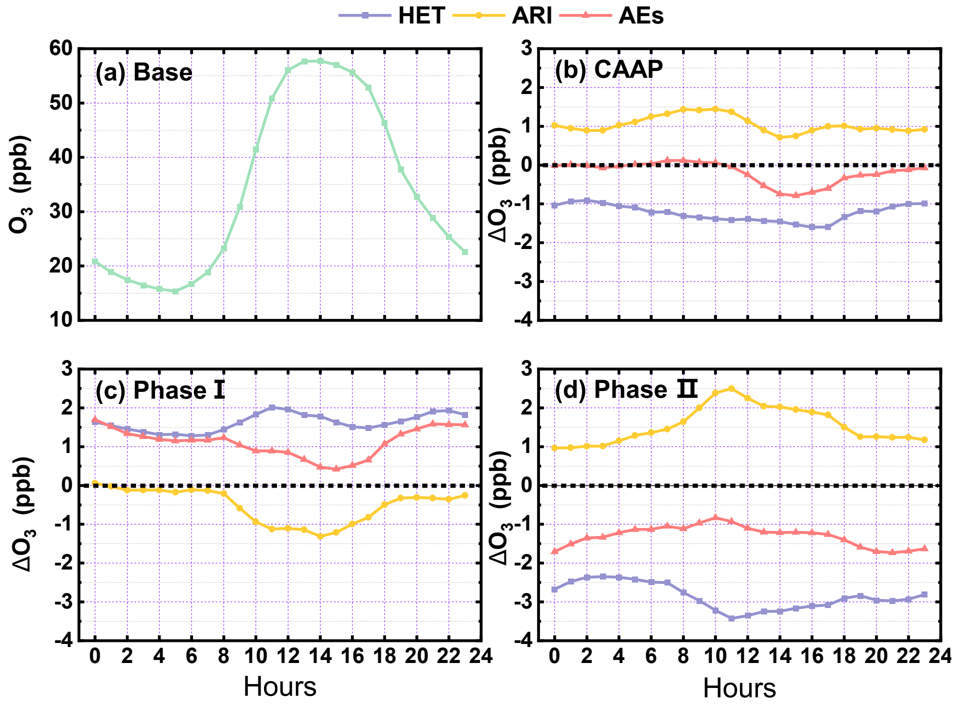

Figure S7. Diurnal variations of (a) baseline O_3 concentrations (ID: 20E20MI_AEs) and (b–d) aerosol-induced O_3 changes (ΔO_3) in summer. Panels show the impacts during the overall Clean Air Action period (CAAP), Phase I, and Phase II, each decomposed into heterogeneous chemistry (HET), aerosol–radiation interactions (ARI), and their combined effects (AEs).

Manuscript changes (Section 3.2, Page 17, lines 343-356):

"To further evaluate whether daytime and nighttime O_3 responses compensate within the daily mean metric, we examined the diurnal cycles of baseline O_3 concentration and the aerosol-mediated impacts (HET, ARI, and AEs) during Phase I, Phase II, and the overall CAAP period for both winter (Figure S6) and summer (Figure S7). Across all phases and both seasons, the dominant O_3 perturbations occur during daytime hours, coinciding with the photochemical peak at 14–16 LT. In winter, Phase I exhibits a pronounced daytime enhancement driven by ARI (up to ~2.41 ppb), whereas HET induces a consistently positive but comparatively weaker increase (up to ~0.49 ppb). In Phase II, the ARI-induced enhancement weakens notably (peaking at ~1.24 ppb), and HET-induced changes remain minor. In summer, the diurnal behavior more clearly reflects a daytime - dominated response. During Phase I, HET produces a marked midday O_3 enhancement (up to ~2.01 ppb), while ARI imposes a weaker yet persistent negative contribution. In contrast, Phase II is characterized by a strong HET-driven daytime O_3 decrease (maximum ~3.43 ppb), overwhelming the comparatively modest positive ARI effect. For all cases, nighttime O_3 changes share the same direction as daytime responses but remain substantially smaller in magnitude, insufficient to offset the daytime signals dominated by photochemistry. These diurnal patterns confirm that the phase-dependent O_3 responses to aerosol effects are not artifacts of day–night compensation in daily mean metrics, but instead arise from robust, daytime-dominant photochemical adjustments."

Minor comments:

5. Figure 3 caption – panel references need correction. The panels currently cite (b) and (c) which doesn't match the description, please correct.

Response: We thank the reviewer for pointing out this oversight. The panel references in the caption of Figure 3 have now been carefully checked and corrected to ensure full consistency with the figure layout and the corresponding descriptions in the text. The revised caption accurately refers to the correct subpanels and their contents **(Page 12, lines 262-264)**.

6. Figures S7-S8: the y-axis is labeled "O_3", but plotted quantity is the change in O3, please relabel to change of O_3 (ΔO_3).

Response: We thank the reviewer for catching this labeling error. In the revised Supplementary Information, the original Figures S7–S8 have been renumbered as Figures S11–S12. In these figures, the plotted variable represents the change in O_3 rather than the absolute concentration. We have therefore corrected the y-axis label to "ΔO_3 (ppb)" in both figures to accurately reflect the displayed quantity. This correction is purely a labeling issue and does not affect the interpretation or conclusions of the results.

7. The manuscript contains several sentences that are unclear and would benefit from careful English editing. For example, the sentence: "Therefore, the commonly used

MDA8 O_3 may underestimate full-day aerosol effects." is ambiguous. If the intended meaning is that using only MDA8 can miss aerosol impacts that occur outside the daytime 8-hour window, especially at night, please reword.

Response: We thank the reviewer for this valuable comment. We agree that the original sentence ("Therefore, the commonly used MDA8 O_3 may underestimate full-day aerosol effects.") was ambiguous and could cause confusion. Our intended meaning was that reliance on MDA8 O_3, which represents only the maximum 8-hour daytime average, may fail to capture aerosol-related influences occurring outside this window, including early morning and nighttime periods. To avoid misinterpretation, we have removed this sentence from the revised manuscript. In addition, we have carefully reviewed and edited the surrounding text, as well as other passages identified as unclear, with particular attention to the **Methods and Results** sections, where technical descriptions and interpretations have been further refined and clarified. These revisions have improved the overall clarity, precision, and readability of the manuscript.

8. For figure 7, which presents changes in HO_2 concentrations, it'd be clearer to express HO_2 in molecules/cm^3 or ppt, as these are the standard units used for radical species. Using these units would also avoid displaying values with multiple leading zeros (as in ppb) and help readers to better assess the relative magnitude and atmospheric significance of the simulated HO_2 changes.

Response: We thank the reviewer for this helpful suggestion. We have updated Figure 7 by converting HO_2 concentrations from ppb to ppt. The revised figure now presents HO_2 in ppt, which improves readability and facilitates comparison with previous modeling and observational studies. The corresponding figure caption and text description in Section 3.2 have also been updated accordingly.

Revised Figure 7 (Page 17, lines 340-342):

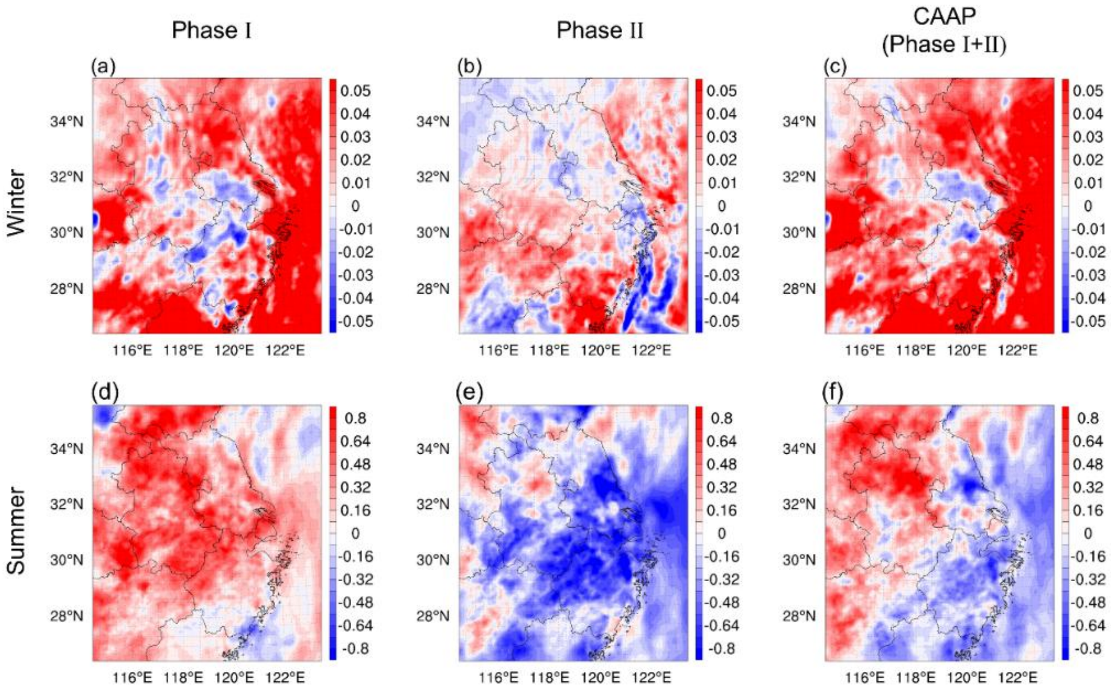

Figure 7 Spatial distributions of HO_2 concentration (ppt) changes induced by aerosol heterogeneous chemistry (HET) in winter (a-c) and summer (d-f) during two phases of the Clean Air Action in the Yangtze River Delta (YRD).

9. Lines 14 – 29: the font size of Abstract does not seem consistent; lines 14-19 font size seems smaller than those of lines 20-29.

Response: We thank the reviewer for pointing out the formatting issue in the Abstract. The inconsistency in font size between lines 14–19 and lines 20–29 resulted from a formatting artifact during manuscript preparation. We have now corrected the font settings so that the entire Abstract is presented in a uniform and journal-compliant font size in the revised version.

We would like to once again express our sincere gratitude to the reviewers for their thoughtful and constructive comments. Their insights have been invaluable and have greatly enhanced the clarity, rigor, and overall quality of our manuscript.