Response to the Comments of Referees

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Title: How meteorological conditions influence aerosol-cloud interactions under different pollution

regimes

Author(s): Jianqi Zhao, Xiaoyan Ma, Johannes Quaas, and Tong Yang

We thank the reviewers and the editor for providing helpful comments to improve the manuscript. We have revised the manuscript according to the comments and suggestions of the referees.

The referee's comments are reproduced (black) along with our replies (blue). All the authors have read the revised manuscript and agreed with the submission in its revised form.

Anonymous Referee #1

Using WRF-Chem-SBM, the authors conduct a realistic short-term regional simulation and a counterfactual one (same as the realistic one but without continental aerosols). They briefly showcase the model is valid by comparing the control (realistic) simulation with some observations. Then they use the pair of simulations conducted to study ACI. Ultimately, the results are mixed and show that ACI is complex and regime-dependent. I think this is a fine manuscript overall: it is interesting, relevant, and seems logical. I do have some concerns before publication (which I will ultimately support) — for the recored, I concur with Reviewer 1's concerns, though I tried not to repeat them.

We sincerely appreciate your encouraging and constructive comments and suggestions, which have greatly helped us improve the quality of this study. All of your comments have been carefully addressed and the corresponding revisions have been incorporated into the manuscript. Our detailed point-by-point responses are provided below (in blue).

General comments:

An interesting detail in the experiment setup is disabling the radiative effects of aerosols and clouds. That's smart, but how much does it really impact the simulations here? Could this decision be contextualized with prior studies and potentially by showing results where these effects are not disabled? How about other confounding effects — of course, you didn't choose to modify other processes (the entropy of the system is different in the simulations). I would like the authors to discuss this decision a little more, and maybe opine on what else could be done to get rid of confounding factors/feedbacks

This approach (disabling the radiative effects of aerosols and clouds) has been commonly used for separating ACI/ARI signals in modeling studies. For example, Liu et al. (2020) isolated the significant enhancement effect of ACI on the extreme heavy rainfall event in Guangzhou in 2023 by turning ARI on/off in simulations. Wang et al. (2020) used similar ARI on/off sensitivity experiments to analyze the impact of dust aerosols on radiative energy budget during a dust storm

and examined their role in the accumulation of primary pollutants over land and the formation of secondary pollutants over the ocean. Zhao et al. (2025), by comparing sensitivity experiments with ACI/ARI turned off against a baseline simulation, disentangled the contributions and mechanisms of both ACI and ARI during a complex pollution episode...

In order to examine how this approach (disabling the radiative effects of aerosols and clouds) impact the simulations, we have conducted the simulations tests (all tests use settings consistent with the Control experiment except for radiation settings) in which cloud radiative effects (Rad CLD), aerosol radiative effects (Rad AER), and both aerosol and cloud (Rad ALL), are turned on. The ECO regional average results are shown in Fig. R1. The radiative effects of aerosols and clouds show minimal influence on the temporal trends of meteorology, aerosols, and clouds (Figs. R1a-e). It is acknowledged that during specific periods, the radiative effects might alter N_a through influencing transport and deposition processes and modify cloud properties by affecting meteorological and aerosol conditions. Their impact on the regional averages of Na, Nd, and CLWP exceeds 20% at certain times. In terms of ACI signals (Figs. R1f-h), the cloud radiative effect accelerates the increase of N_d with N_a and the increase of CLWP with RH. In contrast, the aerosol radiative effect opposes the radiative effect of clouds. The mutual offset between these effects results in the Rad ALL test exhibiting the closest agreement with the Control experiment for these two trends. Compared to these, the relationship between clouds and LTS exhibits weaker regularity. The perturbation from ARI further increases the uncertainty, posing a significant challenge to elucidating the influence of LTS on ACI. Overall, from the perspective of ACI analysis, the quantitative deviations and enhanced qualitative uncertainty caused by ARI necessitate turning off ARI in ACI simulations. We added this description to Section 3.3 of the manuscript.

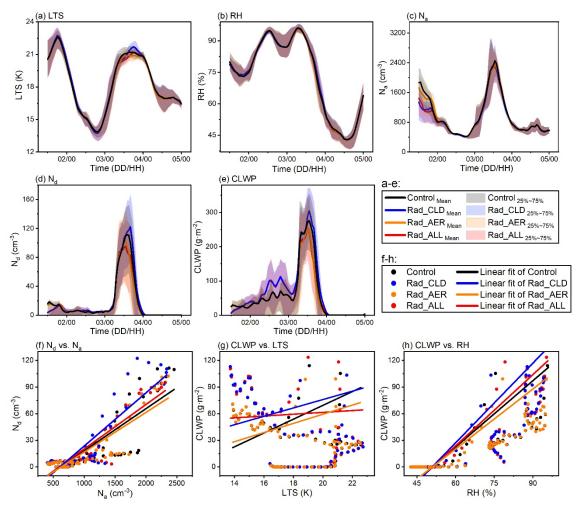


Figure R1. LTS (a), RH (b), N_a (c), N_d (d), CLWP (e), N_d versus N_a (f), as well as CLWP versus LTS (g) and RH (h) for the Control experiment and each radiative sensitivity experiment (Rad_CLD: cloud radiative effects enabled; Rad_AER: aerosol radiative effects enabled; Rad_ALL: both cloud and aerosol radiative effects enabled), showing data averaged over ECO (RH is the column mean from the surface to 1300 m, and N_d and N_a are in-cloud means).

This study kind of left me hopeless about the state of ACI research... While I was reading, I was hoping for something in the conclusion section to offer guidance for future research and/or deeper reflection on what all this means for the community at large. I only found something about higher resolution and bigger domains in the final paragraph, but it seems that's not going to really help, right? Is this just a difficult problem that we are not going to solve well enough?

We regret bringing this feeling to you and are sorry for the misleading statement. In the revised manuscript we have included more discussions and hope to make this clearer. Higher resolution and bigger domains might help, but our point in this manuscript is that to call more attention from the community to assess ACI under different meteorological conditions, our goal is to understand the sensitivity of ACI to meteorology under different conditions and to analyze how ACI varies with aerosols from a meteorology-centered perspective.

Numerous observational and modeling studies have investigated the mechanisms and impacts of

ACI (Saleeby et al., 2010; Bennartz et al., 2011; Ma et al., 2018; Jia et al., 2019, 2022; Guo et al., 2022; Haghighatnasab et al., 2022), as well as their dependence on meteorological conditions (Salma et al., 2021; Zheng et al., 2022; Liu et al., 2024) and aerosol properties (Reutter et al., 2009; Hudson and Noble, 2014). While these efforts have substantially advanced our understanding of ACI, several limitations remain. For instance, (1) bulk microphysical parameterizations may distort ACI signals, quantitatively and even qualitatively; (2) quantitative and qualitative conclusions derived from case studies may not hold under different conditions; and (3) analyses of ACI sensitivity to individual factors are often confounded by co-varying influences, leading to uncertainties in signal separation.

This study addresses these issues by: (1) use of high-resolution spectral bin microphysics modeling to better represent realistic ACI processes; (2) selection of a case exhibiting significant co-variations in meteorology, aerosols, and clouds to support a relatively comprehensive analysis; and (3) examination of ACI responses to the co-variation of different meteorological factors and of meteorological factors and aerosols, enhancement of ACI signal detectability through large N_a differences between Control and Clean experiments, and improvement of meteorological consistency and signal robustness via four-dimensional data assimilation and the disabling of ARI. This study helps to clarify the mechanisms behind the nonlinear variation of ACI with environmental conditions and reduces the associated uncertainties.

Regarding SBM specifically, are you not willing to release the code? Has the code not been released before? Beyond the code, I invite the authors to reflect on whether SBM is the right tool here (this is similar to Reviewers 1's first general comment, but specifically about SBM). It is not readily clear to me if using SBM is better or worse than using a bulk scheme (MG2, P3, etc.)

SBM is currently incorporated in the WRF model but remains uncoupled with online aerosols, and WRF-Chem's online aerosol—cloud module only supports bulk microphysics schemes. Dr. Jiwen Fan's team has developed a coupled version that integrates SBM with online aerosols, which is referred to as the WRF-Chem-SBM model. Researchers should contact Dr. Jiwen Fan for the code.

The differences between bin and bulk treatments of cloud microphysics lead to large discrepancies in their simulations of ACI under the same dynamic and thermodynamic conditions (Fan et al., 2016). Both methods have their own advantages and limitations. The main advantage of bulk is its low computational cost, which makes it suitable for large domains and long-term simulations. However, due to (1) difficulties in accurately handling the CCN budget (Fan et al., 2012), (2) reduced sensitivity to aerosols caused by the adoption of the saturation adjustment approach, and (3) simplified treatment of the conversion from cloud water to rainwater and of hydrometeor fall velocities, bulk cannot guarantee the accuracy of ACI signals (Khain et al., 2015; Fan et al., 2016). The bin method also has some limitations, such as (1) being computationally expensive, usually applicable only to relatively small domains for short time periods, and (2) its accuracy being constrained by our theoretical understanding of cloud microphysics. Nevertheless, bin is

physically more realistic, and relevant evaluations have shown that bin outperforms bulk in reproducing cloud–rain structures and resolving ACI (Khain et al., 2015). This is also the main reason why we chose SBM. Additionally, our subsequent study will step forward on the effects of aerosol size distribution and chemical composition on ACI under different meteorological conditions, the bin-based treatment of CCN size and composition in SBM would be essential. We have added this clarification in the third paragraph of the Introduction.

Comments I wrote while reading the manuscript:

L10: you can remove "quite" here (or you can keep it)

Removed.

L14: What's being driven? The clouds' existence or some specific property thereof?

Cold advection and updraft variations drive the qualitative effects of aerosols on cloud properties including N_d, CER, CLWP, and cloud lifetime. The description has been updated to: "under winter monsoon background, the qualitative influence of aerosols on cloud properties is modulated by variations in updrafts and cold advection (characterized by LTS) driven by atmospheric circulation that co-varies with aerosol concentrations ".

F2: I would personally plot na and CLWP on log scale and I would ensure the same axis is used (so that the reader can see how much lower Na and CLWP will be in the clean case)

We have redrawn Figure 2, with each subplot presenting a comparison between two experiments for a single variable.

T1: You might as well also list the microphysics you're using (SBM)...

We have added.

L189: I would say this more precisely — you're trying to avoid feedbacks into the states, right? See my first general comment

Yes, we have revised it to the more precise expression "To avoid interference from aerosol and cloud radiative feedback on ACI signals, aerosol and cloud radiative effects are disabled in the simulations" Additionally, as noted in our response to your first general comment, we have provided a more detailed

justification for this in Section 3.3 regarding the robustness of the simulated signals.

L196: Can you say more about this assimilation method?

We have added the description of the assimilation method in the final paragraph of Section 2.2.

L208: I don't really see the "positive impact" — in general, what do you mean by "positive" here? Like improvement compared to observations? Second, I don't see any significant movement in F3. The results are pretty good anyway, so it seems the assimilation made little difference and it wasn't needed... But either way, I think discussing the assimilation is confusing because the results look pretty decent without it (so there's no reason for it)

Sorry for misleading, here we wish to express that assimilation generally improves simulated meteorological fields. We have revised the descriptions in the revised manuscript.

Figure 3 in the revised manuscript shows that assimilation enhances simulations of some meteorological elements but also widens the gap between simulations and observations in many vertical layers. Based on this, the statement of the overall "positive" effect lacks conviction. We revised this paragraph to clarify that, from the perspective of vertical profiles, assimilation both increases and decreases the observation-simulation differences in meteorological elements, and we used the clear improvement in the observation-simulation correlation shown in Figure S3 to demonstrate its overall enhancing effect.

This statement is required for evaluating meteorological field simulations and elucidating assimilation effects. Additionally, the role of assimilation in ensuring consistency of meteorological conditions across different experiments and improving the robustness of simulated ACI signals is also briefly noted here.

L217: Define "Control_NoDA" somewhere before you use it; I assume you mean control without data assimilation, right?

Yes. We have added the definition of "Control_NoDA" in the first paragraph of Section 3.2 and in the figure caption of Figure 3.

L252: That's good, I like the fact that you did this. Question though: did you also do the experiment with the aerosol/cloud radiation effects enabled? Did you see anything interesting? I guess I am asking if you could tell us precisely what you got out of disabling these effects ...

Thank you for this comment. We conducted sensitivity tests with cloud (Rad_CLD), aerosol (Rad_AER), and both (Rad_ALL) radiation effects enabled to quantify their impacts on ACI, as

detailed in our response to your first general comment and illustrated in Figure R1.

Overall, radiation effects exert minor influence on the trends of ECO meteorological-aerosol-cloud variations, but they do impact short-term changes during some periods. During these periods, the regional average changes in N_a , N_d , and CLWP induced by radiation effects can exceed 20%. On the ACI signal, in quantitative terms, the cloud radiation effect accelerates the increase of N_d with N_a and the increase of CLWP with RH, whereas the aerosol radiation effect acts in opposition. Qualitatively, ARI increases the uncertainty in the relationship between LTS and ACI.

By disabling aerosol and cloud radiation effects in our simulations, we avoid the quantitative shifts and heightened qualitative uncertainty in ACI signals caused by ARI, thereby enabling a more accurate analysis of how meteorological conditions influence ACI across different environments.

L264: replace "is the containing of continental aerosols" with "is using continental aerosols" Replaced.

F5: good job on this figure :) just remind the reader the sampling frequency/averaging of the data in the caption

Thank you. We have reviewed and revised all figure captions to ensure that the data processing methods for each figure are appropriately described.

F9: I am not sure what this figure is showing exactly. Can you explain it more in the text? What are you trying to show us?

Figure 9 presents the synergistic variation of cloud properties under two pollution regimes in response to meteorological and aerosol conditions.

Previous analyses of cloud property variations with meteorological conditions, based on nearly 2000-fold differences in N_a between the two pollution regimes, demonstrated how ACI varies with dynamic and atmospheric humidity conditions. In Section 4.2, we further analyze the role of meteorological conditions in ACI under different environments, primarily based on Figure 9. Specifically, we focus on three issues: (1) what role do meteorological conditions play in ACI variations with aerosols? (2) How much impact can changes in meteorological conditions exert on ACI under given aerosol conditions? (3) How does the sensitivity of ACI to meteorological and aerosol conditions change with environmental variations?

We have revised both the text and paragraph structure of Section 4.2 to provide a clearer discussion centered around Figure 9.

F10: As Reviewer 1 indicated, be careful with how you define the units.

Thanks for the reminder. We have corrected the data in Figure 10 and changed the figure to size distribution plots with dN/dlogD (cm⁻³) as the vertical axis.

F11: Consider reworking this figure so that the majority of it is not white space (same for F9)

We have redrawn Figures 9 and 11, adjusting the axes to minimize excessive blank space.

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