Response to the Comments of Referees

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Title: Exploring aerosol-cloud interactions over eastern China and its adjacent ocean using the WRF-SBM-MOSAIC model
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We thank the reviewers and 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 #2

This paper investigated the interactions of aerosols and clouds over eastern China (EC) and its adjacent ocean region (ECO) during wintertime based on WRF-Chem with the SBM scheme by coupling the online aerosol module. The results show that the cloud variables are simulated more precisely compared to the bulk model and the default SBM scheme. Besides, the use of the four-dimensional data assimilation is evaluated using multiple observations and shows a positive effect on the simulation results. Upon all these improved models and methods, the authors analyze the differences in aerosol-cloud interactions over EC and ECO owing to the distinct aerosol physical and chemical properties and the meteorological conditions and examine the variations of cloud droplet number concentration with the increase of aerosol number concentration. Moreover, the rapid adjustments for precipitation clouds and non-precipitation clouds are discussed with the variations of cloud liquid water content and cloud effective radius over EC and ECO. This seems like tremendous work exploring aerosol-cloud interactions.

We thank the reviewer for taking the time to assess the manuscript. The reviewer's comments provide great help to improve our research and address deficiencies. We have revised the manuscript carefully according to the reviewer's comments. Please see the following detailed point-by-point responses.

Major comments:

1. One concern is about the evaluation of aerosol simulation. This study only evaluated the simulation of near-surface $PM_{2.5}$, which is not sufficient considering that the aerosol effects on clouds act at a certain altitude. An evaluation of aerosol vertical profile or at least aerosol optical depth should be added.

   Thanks for the suggestion. We have added the evaluation of simulated aerosol optical depth in the revised manuscript (Figure 4d-f and lines 298-302).

2. The authors selected the liquid-only clouds from MODIS data based on some criteria, and the cloud top is detected by MODIS. To compare with MODIS, how to pick the liquid-only clouds from the model output? How is the cloud top of model output defined to evaluate COT, CLWP, CER, and N_d?

   In our submitted manuscript, liquid-phase clouds are defined when the clouds with liquid cloud water content and cloud fraction above 0. Considering that the differences in satellite retrievals and model parameterization calculations, many previous studies defined the liquid-phase clouds in the models based on certain thresholds when comparing with satellite-retrieved data, for example, Roh et al. (2020, https://doi.org/10.1175/JAS-D-19-0273.1) classified the clouds with CLWC > 1 mg m^{-3} and cloud ice water content (CIWC) < 1 mg m^{-3} as liquid-phase clouds. Therefore, we redefined liquid-phase cloud in the revised manuscript, based on the selection of column COT ≥ 5 that matched with MODIS filtering, the vertical layers (48 layers in total) with cloud optical thickness for water (COTW) > 0.1 and cloud optical thickness for ice (COTI) < 0.01 at each grid point and each time are selected as liquid-phase cloud layers, and the highest layer meeting this condition is the simulated cloud top (this filtering of simulated data is only used for comparison with MODIS data, and the analysis of aerosol-cloud interactions in liquid-phase clouds in this study is strictly limited to CLWC > 0 and CIWC = 0). We add this statement in lines 269-276.

3. For the precipitation clouds investigated in this study, the simulated precipitation should be compared with observational data.

   Thanks for the reminder. We have added the evaluation of simulated precipitation in the revised manuscript (Figure 4a-c and lines 296-298).

4. Do the samples in Figure 11-12 contain only liquid water? Could the samples contain the liquid part of the mixed clouds? How to exclude the influence of other types of clouds considering the mechanism of cloud formation and development varies with the cloud type?

   We are grateful for this suggestion, it is a point we had not considered before. In the revised manuscript, we have imposed a strict restriction on cloud phase, selecting only the grids with CLWC > 0 and CIWC = 0 (CIWC is the sum of ice, snow, graupel and hail water contents) as liquid-phase clouds, and clarified this restriction in lines 275-276. In addition, we clarified in the manuscript title and content that this study targets liquid-phase cloud.

5. As for the N_d variations in each N_{aero} interval in Figure 11, at the second stage, the authors stated that N_d in EC still increases swiftly with N_{aero} due to the relatively strong updraft and surface radiative cooling. Please explain why the N_d in ECO does not increase like N_d in EC.

   In ECO, due to the inability to rely on the effects of surface like EC to reach supersaturation, the dominance of water vapor variation on aerosol activation is more pronounced, and the supersaturation shows a steady decreasing trend with increasing N_d. After N_{aero} exceeds 10000 cm^{-3} (average N_d exceeds 500 cm^{-3}), the increase in small aerosols and the decrease in supersaturation prevents its N_d from continuing to increase and N_d starts to show a decreasing trend. We add this explanation in lines 458-461 and add a detailed analysis of the difference between EC and ECO atmospheric supersaturation pathway in lines 410-429.

6. Many studies using satellite data to explore aerosol-cloud interactions view AOD or aerosol index (AI) as an indicator of aerosol concentration due to the limit of observations. I wonder if the simulated AOD is improved after the assimilation. It would be great if the authors could plot the variations of the simulated CLWP with AOD in EC and ECO.

   Thanks for the suggestion. We have added the evaluation of the effect of assimilation on AOD simulation in the revised manuscript (Figure 4d-f and lines 298-302). We have also plotted the variations of the simulated CLWP with AOD, and added an analysis of the variations of CLWP and its relevant influences with AOD in Figure 15 and lines 522-537.

7. In section 3.4, when exploring aerosol-cloud interactions, meteorological fields may affect both aerosols and clouds, resulting in covariance between the two, such that changes in clouds cannot be attributed to aerosols. The authors need to further discuss the role of the meteorological field in this study and exclude its effects.

   Thanks for the suggestion. We have discussed the variations of N_d and CLWC under different meteorological (U-wind, V-wind, W-wind, temperature, water vapor content, temperature variation and water vapor variation) and aerosol conditions in the revised manuscript (Fig. 13 and Fig.16 as well as lines 475-491 and lines 546-551).

Minor comments:

1. Line 9: Delete "of". "Coupling a spectral-bin cloud…" is suitable.

   Corrected .

2. Line 22: Delete ", which".

   Corrected.

3. Line 22: It should be "large-scale"

   Corrected.

4. Line 28: Aerosols show indirect effects as CCN and IN.

   Corrected.

5. Line 29: "Remain" should be changed to "remains".

   Corrected.

6. Line 51: It should be "depends on" instead of "depends".

   Corrected.

7. Line 86: It should be "Benefiting from advances in computational science".

   Corrected.

8. Line 115: It should be "is" before "consistent".

   Corrected.

9. Line 130: "Thus greatly promoting" is more suitable. So is "optimizing" in Line 270.

   Corrected.

10. Line 260: Replace "This" with "These".

    Corrected.

11. Line 355: Replace "comes" with "come" and delete "are" at the end of this line.

Corrected.

12. Line 370: Is there any direct evidence to prove that radiative cooling makes a large number of cloud droplets distributed near the surface?

    Since surface radiative cooling occurs mainly from night to early morning, we analyze the variations of temperature profile and near-surface supersaturation in EC during daytime (7:00 to 18:00 in Beijing time) and nighttime (19:00 to 06:00 of the next day in Beijing time), and the results are shown in Fig. S2b and c. It can be seen that the presence of nighttime near-surface thermal inversion makes the nighttime supersaturation generally higher than the daytime during the simulation period, which effectively boosts the aerosol activation and indicates the important influence of surface longwave radiative cooling on atmospheric supersaturation and aerosol activation. We add this analysis to lines 419-422 of the revised manuscript.

13. Figure 11 shows the variation of N_d with aerosol and other related factors based on the statistics of the model grids with CF greater than 0 at each time. The method to calculate CF in this study is that CF equals 1 when the sum of cloud water and cloud ice mixing ratios is greater than 10^{-6} $kg \cdot kg^{-1}$, otherwise, the CF equals 0. Why does the calculation of CF use cloud ice mixing ratio since this study focuses on liquid clouds?

    Thanks for the comment. The CF in the original manuscript is calculated from the model parameterization (the threshold method mentioned in this comment). In the original manuscript we sampled the grids with CF and CLWC greater than 0. In the revised manuscript, we have imposed a strict restriction to exclude the ice phase, i.e. only the grids with CLWC>0 and CIWC=0 are selected.

14. Figure 6-8: Have you done a significance test for the correlation coefficient?

    We calculated spatial correlation based on Pearson product-moment method, which is not accompanied by a significance test. Due to the spatial and temporal discontinuity of MODIS data and our data filtering, only some of spatial coordinates have relatively continuous time series, and many coordinates have only one or a few valid values, which makes it meaningless to do significance tests for each coordinate point, so we did not perform significance test for the spatial correlation. In addition, we performed significance tests on the $PM_{2.5}$ data with good continuity, and we used RMSE to auxiliary spatial correlation coefficients.

15. How do the authors define the liquid cloud? I just wonder why the liquid clouds appear higher than 4 km in Figure 10b. Besides, should the role of sea salt acting as the ice nuclei be considered in the discussion?

    In the submitted manuscript, we did not strictly select the grids containing only liquid-phase clouds for the analysis of cloud droplet distributions in Fig. 10. Because aerosol activation can occur at any place where supersaturation and aerosol conditions are met, cloud droplets can appear above 4 km. In the revised manuscript, we only analyze the cloud droplets in the liquid-phase clouds, and the new N_d distribution can be seen in Fig. 11c-d, where almost no liquid-phase clouds appear above 4 km. The coupling of SBM and MOSAIC by us and most related researchers focus mainly on the activation of aerosols into cloud droplets without modifications to the SBM ice phase nucleation, so the role of sea salt acting as ice nuclei cannot be directly resolved. In addition, we make a further statement in the title and content of the article to explore only aerosol-cloud interactions in liquid-phase clouds, so the role of sea salt acting as an ice nuclei is not discussed here.

16. What do the downdrafts in Figure 10d imply?

Thanks for the comment. The figure presents the spatial distribution of the atmospheric vertical velocity, where both updrafts and downdrafts appear over the whole spatial scale.

17. The manuscript mentioned that the near-surface areas around 29°N and 31°N (Fig. 10b) exhibit high atmospheric supersaturation due to the effect of topographic uplift (Fig. 10c). Why the cloud number concentration is rather low in the south of the topographic uplift?

Thanks for the reminder. This is because Fig. 10 in our previous manuscript was based on oblique profiles (along the yellow lines in Fig. R1a), which did not provide enough complete information. We modify it by using the average value of the corresponding latitude instead of the oblique profile to make the figure present more reasonable information.

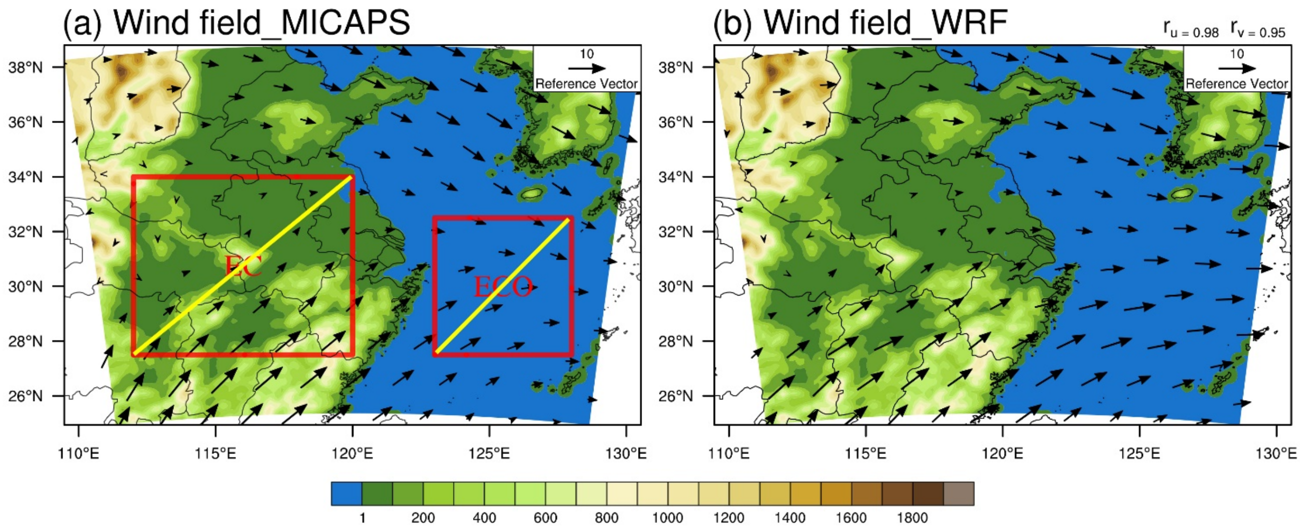

Figure R1. Topography (unit: m) of the model domain, MICAPS (a) and assimilated simulated (b) 850 hPa wind fields (unit: m·s-1) and positions of the oblique lines (yellow lines in Fig. R1a)