Review of "Exploring aerosol-cloud interactions in liquid-phase clouds over eastern China and its adjacent ocean using the WRF-Chem-SBM model" by Zhao et al., submitted to Atmospheric Chemistry and Physics (ACP)

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This report contains general, major, and specific comments from this reviewer on the manuscript.

A summary of the manuscript and general assessment:

Recommendation: Reconsidered after major revisions

This study conducted WRF-Chem-SBM simulations to investigate aerosol-cloud interactions, particularly the relationship between aerosol number concentration and cloud droplet number concentration/liquid water content, focusing on the differences between eastern China (EC) and the adjacent ocean (ECO). The simulations reasonably reproduced the distributions of cloud properties and precipitation amounts obtained from satellite measurements, especially when a four-dimensional assimilation was applied. The authors identified dominant mechanisms for cloud development in the EC and ECO. The different relationships between aerosol number concentration and cloud droplet number concentration/liquid water content in EC and ECO are presented and the mechanisms for the differences are discussed.

This study is within the aims and scope of Atmospheric Chemistry and Physics (ACP), specifically the subject for "Aerosols, Cloud and Precipitation", the research activity for "Atmospheric modelling", the altitude range for "Troposphere", and the science focus on "both Chemistry and Physics".

Although I appreciate the authors' efforts in the model simulations using the state-of-the-art model for chemical-aerosol-cloud-precipitation interactions and the analysis, this manuscript has significant problems in readability and presentation, particularly using figures. It was very tough for me to read the whole manuscript and try to understand the contents in the presentation. I found that this manuscript was resubmitted to ACP after a rejection, and I reviewed the open discussion on the previous manuscript. I have no idea why the previous reviewers did not point out the presentation problems. However, I cannot ignore the problems because they greatly reduce the quality of the manuscript. Also, in my opinion, there are still major problems in the analysis approach of the simulation results and the discussion of the significance of the results with those of other similar studies. More details are provided in the following paragraphs.

First, the presentation using figures has serious problems. The worst figure and caption are in Fig. 9, and I describe my complaints individually in the Major Comments below. The main problem with the figures is that there is little explanation of what is shown in most of the figures and how the values shown are calculated. As a result, almost every time I encountered figures while reading the manuscript, I had to speculate how the authors calculated and produced the plots. This was stressful and confusing. In most cases, I assumed simple spatial and/or temporal averages, not instantaneous values of the variables, because the date and time are not specified in the captions. However, there are some variables, such as positive supersaturation and cloud droplet effective radius (CER), that cannot simply be averaged over all grid points. For example, in Fig. 8, these variables are shown together with Naero and water vapor, which can be simply averaged over all grid points. I am very confused as to whether all variables were averaged over the same sampling or not, yet the authors use the figure to argue for their correlation. In particular, sampling supersaturation needs special care, because it could be negative even in the cloud. The values and the scientific implications of the averaged supersaturation depend heavily on how it is sampled for averaging.

I request the authors to revise the figures, their captions, and the text of the manuscript to explicitly state what is shown and how the values shown in all figures (including Supplementary Materials) were calculated to avoid confusion and misunderstanding by readers. This is one of the minimum requirements for scientific papers.

Second, the approaches to analyze the simulation results in Sections 3.3 and 3.4 have significant problems. It is difficult for me to tell all the problems, especially related to the statistical analysis, and how to improve them to the authors in these review comments, but I hope the manuscript will be improved as much as possible for reconsideration. First of all, the authors would perform statistical analysis to obtain the average states without fluctuations from individual extreme processes (line 318). However, Figs. 9-13 clearly show such fluctuations, probably due to the limited sampling volume for the conditions. The parts with fluctuations are reflected in the discussion and conclusions. If the authors want to exclude the effects of individual extreme processes, the conditions with low frequencies should be filtered out in the calculation of the statistics.

Related to the above problem, there is little information about the probability or frequency of the variables discussed in the sections, except for Nd and CLWP. This makes it difficult to understand how representative each point in the colored contours and lines in Figs.

9-13 is of the entire special and temporal sampling volume. For example, Fig. 10a shows a ratio of Nd to Naero averaged over small ranges (bins) of U and Naero, if my understanding is correct. However, this plot does not have the information of how frequent and representative each point is in the whole sample volume. For example, -20 m/s of U is probably a fairly rare case and not representative of what is simulated in EC. Additional levels of statistical analysis for frequency and sampling are needed to unravel the correlation with different weather conditions and finally understand the relationship between Naero and Nd-LWP.

There are some misleading interpretations of the statistical analysis. Please be careful when writing the explanation. For example, a higher mean does not always mean a higher frequency or probability.

Third, there is no discussion of what and how the use of the state-of-the-art model improved the simulations of aerosol-cloud interactions compared to other previous modeling studies as well as observations. This is necessary to explicitly demonstrate the scientific significance of this research. For example, as partially described in the introduction of the manuscript, conventional global aerosol transport models have had problems, e.g., they tend to overpredict the increase in LWP in response to increases in Na or Nd, compared to global-scale satellite observations (e.g., Quaas et al. 2009). Did the simulations in this study solve the problem or not? How similar or different is the obtained relationship between LWP and Na(AOD)orNd compared to those in high-resolution model simulations in other studies? In addition, there is no direct comparison of the relationship with satellite observations in this case, although the authors sometimes emphasize the advantages in the manuscript. Why was the relationship between LWP and Nd from the simulations not directly compared with that from Terra/Aqua MODIS satellite observations in this case?

Other Major comments:

1. Caveat when evaluating simulation performance based on RMSE and correlation to the coarser resolution gridded dataset (Fig. 2)

There is a caveat when evaluating the simulation performance based on the RMSE against the coarser resolution gridded data set than the model horizontal grid spacing in Fig. 2. The MICAP is horizontally gridded data with a resolution of 2.5 degrees. This basically means that the data does not contain information about variations at horizontal scales smaller than degrees, which can definitely occur in the real world. Thus, when MICAP is re-gridded into the model horizontal grid structure with finer grid spacing, the interpolated data cannot be fully used as the true value for evaluating spatial variations in the simulated fields containing finer-scale variations represented in the model with finer grid spacing. Therefore, the calculated RMSE for Figure 2 has little validity, and the difference in RMSE calculated in such an approach does not guarantee worse or better performance of the simulations.

From this perspective, the RMSE and correlation against IMERG in Fig. 3 are okay, but those against PM2.5 have the same problem. In the case of PM2.5, the improvement of the mean bias by using the assimilation greatly reduces the RMSE, while it does not guarantee the improvement of the horizontal variations, especially at smaller scales, as shown by the similar correlation coefficients between the two simulation results.

2. Figure 9

At first glance, I could not understand what the color contours in these plots were showing at all because of the mismatch between the plots and the caption, as well as the poor wording in the caption. However, after struggling for several hours over days, I finally reached an understanding of what these plots would show to some degree, although I still do not have full confidence. First of all, I was totally confused by the mismatch between the plots of Figs. 9a, 9e, 9i, 9k and the first line of the caption, "Probability density distribution functions (sum of probabilities corresponding to 1 for each Naero or AOD value)". Figures 11 and 12 also have the same problem. The numbers next to the color bars in these plots are much larger than 1, which is clearly inconsistent with the caption that the sum is 1. Also, these plots of Figs. 9a, 9e, 9i, 9k are collocated with the other plots in which the color contours show a different type of variable, not probability, which further confused me. Finally, I came to the conclusion that the color contours in Figures 9a, 9e, 9i, 9k would show the probability distribution in percentage (%) format. However, this information is not included in either the plots or the captions.

Another big problem is the phrase "the first two lines" and "the third line" in the caption and in the text of the manuscript. I could not understand which line(s) in the series of plots corresponded to "the first two lines" and "the third line". Each plot panel has only one line. Finally, I came to the conclusion that "the first two lines" would mean the lines in Figs. 9a, 9b, 9c, 9d, 9e, 9f, 9g, and 9h, and "the third line" would mean the lines in Figs. 9i, 9j, 9k, and 9l. But this wording is rather ambiguous. If my conclusion above is correct, the authors should have explicitly indicated which lines of which plot panels, or at least these words should have been "the lines in the plots in the first and second rows in Fig. 9" and "the lines in the plots in the third row in Fig. 9".

The position of the plot panels is also quite misleading and unfriendly to the reader. If Figure 9k is in the same series plot as Figures 9a, 9e, and 9i, it should be in the same row or column of the plot group and follow Figure 9i. Same for Fig. 9l. And why does Fig. 9 have no plots for the relationship between AOD and Nd vs. supersaturation and QV, even though the same plots are included but against the aerosol mean radius?

3. Causality between AOD and precipitation

The discussion in the manuscript does not take into account that the aerosol-cloud interactions are not one-way, especially when precipitation is involved. Figure 3 shows that there is a negative correlation between accumulated precipitation amounts and near-surface concentrations of PM2.5. The constraint of the meteorological variables by the assimilation does not affect the emission from the land surface, except for dust. Thus, the accumulated precipitation amounts were changed by the constraint of meteorological variables using the assimilation as the first order. The change in the accumulated precipitation amounts modified the aerosol concentrations by the wet deposition process. Then, the modified aerosol concentrations can affect the precipitation amounts through the change in cloud microphysical properties as the second order. Therefore, the effect of wet deposition by precipitation is not negligible in the current simulations.

The discussion in Section 3.4 is sometimes misleading regarding the causality between AOD and precipitation, as noted in one specific comment. If the authors believe that this part, which I consider misleading, is correct, please provide evidence through additional levels of analysis to show its validity.

4. The relationship between the multiple mechanisms for cloud development and Nd

There is a conclusion like " multiple supersaturation pathways and abundant aerosols in EC cause Nd to exhibit a much robust increasing trend compared to ECO at low Naero and strong fluctuations at high Naero" (line 453). I agree that the role of abundant aerosols in EC is important, while the role of multiple supersaturation pathways, i.e., multiple mechanisms for cloud development, is still unclear to me. The manuscript does not show the contribution of each mechanism or pathway to the formation of the Nd statistics. If the authors think this is important, please provide additional evidence.

Specific comments:

Line 122: As I look at the time series plots of near-surface PM2.5 concentrations at the ground sites in Fig. 4, removing the first 24 hours from the analysis as a spin-up seems insufficient, because the simulation clearly underpredicts the concentrations at most sites in a few days from the beginning compared to those at later dates in the period. Should at least the first 48 or 72 hours be removed from the analysis?

Line 123: For the initial and lateral boundary conditions of the gas and aerosol species, were any data sets used or not?

Table 1. Both RRTMG and CAM have their own radiation schemes for both long and short wavelengths. Why did the authors use different packages for the long and short wavelengths?

Line 150: Did the four-dimensional data assimilation (FDDA) in WRF apply only to the simulation in the parent domain with 12 km grid spacing (Fig. 1), or also to the simulations in the nested domains with 4 km grid spacing?

Line. 156: Relative humidity is not included in the data sets (relative humidity is already a function of pressure, temperature, and dew point).

Line 160: Please use the correct full name of the product, the Integrated Multi-satellitE Retrievals for GPM (IMERG). Also, please do not forget to include the following citation in the reference as shown on the web page.

Huffman, G.J., E.F. Stocker, D.T. Bolvin, E.J. Nelkin, Jackson Tan (2019), GPM IMERG Final Precipitation L3 1 day 0.1 degree x 0.1 degree V06, Edited by Andrey Savtchenko, Greenbelt, MD, Goddard Earth Sciences Data and Information Services Center (GES DISC), Accessed: [Data Access Date], 10.5067/GPM/IMERGDF/DAY/06

Line 168: Please add the following citation for MOD06_L2.

Platnick, S., Ackerman, S., King, M., et al., 2015. MODIS Atmosphere L2 Cloud Product (06_L2). NASA MODIS Adaptive Processing System, Goddard Space Flight Center, USA: http://dx.doi.org/10.5067/MODIS/MOD06_L2.061

Line 222: There is no citation and data source information for the MODIS AOD in Section 2.3. And is the simulated AOD a "clear sky" AOD, correct?

Line 264: The phrase "supersaturation pathway" is often used in this manuscript, but this is not popular in the community; at least I did a Google search, but nothing came up for atmospheric research. Please add an explanation of the meaning at this first appearance.

Figure 6. These values are a variable (x, y, z, t) in the model. Please include an explanation of how the authors sampled and calculated the values shown in Fig. 6 from the model output.

Figure 8 and Line 291-368. First of all, it is problematic that there is no explanation of how the vertical cross section of the supersaturation was calculated in Figs. 8e and 8f. Since the shown supersaturation is greater than or equal to zero, I assume that the authors only sampled grid points with supersaturation > 0% (i.e., relative humidity > 100%) and plotted the average there. However, if this is the case, the higher supersaturation in Figs. 8e and 8f does not always mean that supersaturation and clouds occur more frequently, which is probably what the authors would argue around line 296-368.

Lines 298-303: Aren't (2) and (4) identical after all?

Line 351: "temperature and water vapor changes at that time compared to the last time". I do not understand what this means.

Lines 352-359: The plots in Fig. 10 show a ratio of Nd to Naero, not a ratio of locally activated Nd from aerosols to Naero. Since Nd is advected by wind, the existence of Nd is not identical to the existence of aerosol activation.

Lines 360-365: I cannot follow what the authors would argue here. Since supersaturation, i.e., relative humidity, is a function of air pressure, temperature, and water vapor (mixing ratio or specific humidity), the existence of saturation is based simply on whether these three components meet the condition.

Line 385: In the range of Nd above 16,000 cm-3 the sample volume seems to be insufficient to calculate the means of the CLWC for a robust conclusion. Same as in line 388 for non-precipitating clouds. These are inconsistent with the concept of statistical analysis described in line 318, " avoid fluctuations from individual extreme processes and obtain the average state of the liquid phase clouds".

Lines 398-399: The causality in this sentence for Fig. 12d does not make sense to me. As shown in Fig. 3, precipitation has a significant effect on the simulated aerosol concentration through wet deposition. Thus, simulated AOD is low where there is heavy precipitation. Also, high CLWP is needed to develop heavy precipitation.

Line 408: However, each line graph in Figs. 12e and 12h shows a peak near AOD=0.

Line 480: Please submit namelist files of the WRF model simulations in the supplemental materials.

Grammatical problems:

"Strong aerosol activation", this or similar wording is often found throughout the manuscript, but is ambiguous. Please rephrase to avoid misunderstanding.

Line 402: "ADO" => "AOD"