Response to the Comments of Referees

Journal: Atmospheric Chemistry and Physics
Manuscript Number: egusphere-2023-331
Title: Exploring aerosol-cloud interactions over eastern China and its adjacent ocean using the WRF-SBM-MOSAIC model
Author(s): Jianqi Zhao, Xiaoyan Ma, Johannes Quaas, and Hailing Jia

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 #1

Review of "Exploring aerosol-cloud interactions over eastern China and its adjacent ocean using the WRF-SBM-MOSAIC model" by Zhao et al.

The study coupled WRF-Chem with SBM and used it to study aerosol-cloud interactions for a stratiform cloud case at 15-km resolution. There are a few serious problems as detailed below. Here is a high-level summary: (1) there is no clear motivation, particularly in terms of coupling MOSIAC with SBM since this was done and applied to many studies already, (2) the coupling that the authors did is not correct, (3) applying supersaturation-based activation in SBM to 15 km resolution is not appropriate. These are serious methodology problems, so I recommend a rejection of the study.

We thank the reviewer for taking the time to assess the manuscript and providing helpful comments and suggestions to improve the manuscript. Our point-by-point responses are as below: (1) we acknowledged that some previous studies have coupled SBM and MOSAIC, but such version is not available to public for use, and currently we don't see a version with the coupling of SBM and MOSAIC from the latest public versions of WRF-Chem. To better understand the physical mechanisms of aerosol-cloud interaction from modelling studies, it is necessary for us to couple SBM-MOSAIC in WRF-Chem since both the detailed cloud microphysics including cloud droplet size and aerosol microphysics (for example aerosol size distribution and chemical component) are key factors. We have included more statement to clarify our motivation in the revised manuscript. (2) The coupling system used in our submitted manuscript was based on particle size and ignored the differences in hygroscopicity of different aerosols. According to the reviewer's suggestion, we have imported the MOSAIC default hygroscopicity for each aerosol into SBM and treated the activation of each species of aerosol separately in order to make aerosol activation parameterization capable of resolving aerosol species. (3) This study focuses on liquid-phase clouds with wide range and long duration, and mainly investigates the general characteristics of aerosol-cloud variations rather than a specific process, so a larger temporal and spatial range needs to be adopted, and only a relatively coarse spatial and temporal resolution can be used under limited computing power. Although at 15km resolution mostly using bulk cloud microphysical scheme, our evaluation in Sect. 3.2 shows that the SBM has better simulation performance than the bulk scheme at 15 km resolution for both macrophysical and microphysical cloud properties. In addition, two sets of tests (see response to detailed comments below) are performed to examine the SBM simulation ability differences between the simulations of coarse temporal and spatial resolutions compared to fine resolutions. The test results show that the use of coarse temporal resolution only differs somewhat from the fine resolution simulations in the variation of N_d due to the

difference in resolving meteorological fields, but in general both show consistent variation and do not cause neglect of aerosol activation and clear underestimation or overestimation of N_d by using coarse resolution. In addition, the use of coarse spatio-temporal resolution shows consistent characteristics in both the spatial distribution of aerosol and cloud parameters and the variations of cloud parameters with aerosol. Only the lack of accuracy in resolving the meteorological field leads to relatively few strong activation samples and relatively low average N_d, which can be compensated by 1) optimization of the meteorological field simulation by four-dimensional assimilation, 2) resolving of sub-grid convection by cumulus parameterization,3) inclusion of more abundant atmospheric processes at a larger spatial and temporal scale, 4) selection of winter period dominated by cloud types with large temporal and spatial ranges such as stratocumulus, and 5) filtering out of ice-phase processes when analyzing. Due to a) the better simulation performance of SBM compared to the bulk scheme for cloud macroscopic and microscopic properties at 15 km resolution demonstrated in Section 3.2, b) the consistency of the simulated aerosol and cloud variations at coarse resolution with fine resolution revealed by the tests, c) the aforementioned measures to compensate for the lack of meteorological field simulation ability at coarse resolution, and d) the convenience of SBM for resolving aerosol particle size, aerosol composition and droplet size on aerosol-cloud interaction, it is appropriate to use 15 km resolution in this study. Please see the following detailed point-by-point responses.

Detailed comments:

Abstract:

1. The advantage of WRF-Chem is to study aerosol effects from aerosol properties. It would allow us physically to study how N_d is affected by different aerosol properties. However, the study did not take this advantage and they only examine the relationship of cloud properties with N_d but did not even connect with aerosol properties. Aerosol effects starts from how N_d are changed through aerosol properties.

   In Sec. 3.3, we examined the response of cloud droplet size spectra to aerosol size (Fig.9) in the submitted manuscript. In addition, we have included the analysis of response of cloud droplet to aerosol composition (Fig.10) in the revised manuscript. The detailed analysis and discussions are presented in Sec.3.3 and 3.4.

2. The sentence “more bursty atmospheric supersaturation and lack of subsequent water cause cloud liquid water content (CLWC) in EC to increase explosively with N_d”, the sentence is confusing. How would lack of water contribute to increase N_d?

   Thanks for the reminder. What we meant is that when water content is limited, CLWC exhibits rapid increase with N_d if there are few cloud droplets, and then slow down. We have modified this sentence for clarification in the revised manuscript.

Introduction,

1. The introduction is all about aerosol impacts on stratocumulus and warm clouds. Is this the cloud you study in this work? If so, you need to clarify this at the beginning of the introduction so that people would know your target since the mechanisms of aerosol effects are very different with different types of clouds.

   Yes, our study focus on liquid-phase cloud. We have clarified this in the revised manuscript.

2. Line 81-82, “WRF-Chem currently only provides the coupling of bulk microphysical schemes with an online aerosol module (Gao et al., 2016)”: This is totally wrong, what Gao et al. 2016 documented is the WRF-Chem coupled with spectral-bin microphysics (SBM). I noticed this is later stated. But here correction is needed.

   Corrected.

3. Line 85-90, these statements were good for the motivation of Gao et al., (2016) because bin microphysics scheme was never coupled with chemistry/aerosol module before. However, it does not apply anymore now since Gao et al., (2016) already built the capability and it has been used in many studies such as Fan et al., 2020, https://doi.org/10.5194/acp-20-14163-2020, Zhang et al., 2021, https://doi.org/10.5194/acp-21-2363-2021.; Lin et al., 2021https://doi.org/10.1175/JAS-D-20-0106.1, , Lin et al. 2022https://doi.org/10.5194/acp-22-6749-2022). Thus I have a difficulty to understand the motivation of this work, which repeats Gao et al. (2016) without a justification.

   We acknowledged that some previous studies have coupled SBM and MOSAIC, and used it to examine aerosol-cloud interaction, specifically, for convective clouds. Some good examples are listed above by the reviewer. In our study, we focus on liquid-phase cloud, and attempt to explore how liquid-phase clouds response to aerosol physical properties as well as meteorological conditions. As we understand, although bin microphysics scheme was coupled with chemistry/aerosol module and also used in many studies, but such model version is not available to public for use, and currently we don’t see a version from the latest public versions of WRF-Chem. To better understand the physical mechanisms of aerosol-cloud interaction from modelling studies, it is necessary for us to couple SBM-MOSAIC in WRF-Chem since both the detailed cloud microphysics including cloud droplet size and aerosol microphysics (for example aerosol size distribution and chemical component) are key factors.

Section 2,

1. I am very confused by the writing of this section. It starts from describing “This study is based on WRF-Chem v3.9, the full version of the SBM scheme coupled with the aerosol module (Khain et al.,2009)”, and provided detailed description of SBM, then saying SBM is not coupled with chemistry/aerosols instead of using prescribed CCN. First, in the introduction, Gao et al., 2016 which coupled with SBM with WRF-Chem is described. This writing is not only misleading but I’d ask what’s the logic and point you want to deliver? The authors are totally ignoring Gao et al., 2016 here and repeating what was done in Gao et al. 2016 but did not provide any justification why you are doing this.

   Thanks for the comment. What we wanted to express is that current public versions of WRF-Chem model, even the recent WRF-Chem v4.4, SBM still uses the prescribed CCN, so we need to couple it with online aerosol in order to improve its ability to simulate aerosol-cloud processes. The original text is somewhat misrepresented. We have revised “This study is based on WRF-Chem v3.9, the full version of the SBM scheme coupled with the aerosol module (Khain et al.,2009)” to “This study is based on WRF-Chem v3.9, we use the full version of SBM (Khain et al., 2009) to couple with MOSAIC” and revised “In the current SBM scheme of WRF, the CCN…” to “In the current SBM scheme of the public versions of WRF-Chem, the CCN…”. In addition, in the revised manuscript, we use an improved version of the aerosol activation parameterization compared to the previous manuscript, as detailed in the response to the next comment.

2. Page 5-6, there is a significant problem I see in the coupling SBM with MOSAIC. MOSAIC

predicted various aerosol composition and hygroscopicity over different size ranges and it is physically wrong to map the aerosols from MOSIAC into aerosol bins in SBM based on size only, since activation of aerosols in SBM is only based on simple composition (default sea salt) to get the critical supersaturations for each bin. This is the key but most tricky part for this coupling. That is why Gao et al. (2016) implemented an interface code to set up the 33 critical supersaturations bins so that aerosols in the interstitial size bins in MOSAIC are mapped (i.e., distributed) to the 33 preset aerosols size bins through the interface module, and the mapping is based on particle critical supersaturations calculated from MOSAIC-predicted aerosol properties (size and hygroscopicity) rather than dry size. This was clearly described in Gao et al., 2016 and provided clear explanation why it is done in this way, i.e., "Since MOSAIC-predicted aerosol compositions vary with bins and over each bin a lognormal size distribution is assumed, mapping of aerosols from MOSAIC into SBM bins based on S_{crit} is the easiest and most precise way to connect aerosols between the two models. That is, all aerosol types are represented on a single set of 33 CCN bins, but each aerosol type may distribute over different CCN sizes." Besides this, I see the authors follow Gao et al. 2016 on all other coupling treatments.

Thanks for the suggestion. The hygroscopicity is very important and was neglected in our submitted manuscript. In the SBM scheme, the aerosol activation parameterization is based on Kohler theory, which relies on the critical activation radius of aerosol calculated based on aerosol hygroscopicity and other parameters to treat aerosol activation. We have modified the coupling system based on the differences of aerosol hygroscopicity (the specific values are referred to the default values of the MOSIAC aerosol module, making the coupling system compatible) so that SBM can handle each aerosol composition and particle size, as described in lines 164-171 of the manuscript.

3. They use 15 km resolution for this study. The advantage of using SBM is diminished at such coarse resolution. On the contrary, the original WRF-Chem with the bulk schemes is more appropriate since supersaturations cannot be resolved much so activation is parameterized with Abdul-Razzak and Ghan parameterization. However, the activation of aerosols in SBM is based on supersaturations (not parameterized as Abdul-Razzak and Ghan scheme) but supersaturation would be poorly simulated at 15 km resolution (only very limited supersaturation can be resolved at such a coarse resolution). Therefore, this makes the effort of coupling SBM with WRF-Chem and the use of such physics-complicated model meaningless

   Thanks for the comment. In previous studies, SBM was mostly used for individual convective system simulations, and the spatial and temporal resolution were mostly 1-3 km and 6-18 s. Our study focuses on liquid-phase clouds with wide range and long duration, and mainly investigates the general characteristics of aerosol-cloud variations rather than a specific process, so a larger temporal and spatial range needs to be adopted, and only a relatively coarse spatial and temporal resolution can be used under limited computing power. Although at 15km resolution mostly using bulk cloud microphysical scheme, our evaluation in Sec. 3.2 shows that the SBM has clearly superior simulation performance than the bulk scheme at 15 km resolution for both macrophysical and microphysical cloud parameters.

   In addition, to understand the simulation ability of SBM at coarse resolution compared to fine resolution, we conducted two sensitivity tests trying to answer below questions: (1) whether coarse temporal resolution ignores many activation processes and causes overall aerosol-cloud-water

vapor simulation errors, and (2) whether reasonable aerosol-cloud variation information can be simulated at coarse spatio-temporal resolution. Two sets of tests are set up, focusing on the precipitation process on 2 February 2019, Test 1 is a 6h simulation (0:00 to 6:00 on 2 February) with 15 s and 60 s integration at the same grids (15km resolution) of this study. Test 2 is a 24-h nested simulation (from 00:00 on 2 February to 00:00 on 3 February) using 9 km (54 s) and 3 km (18 s) spatial (temporal) resolution, with the model domain shown in Fig. R1 (because the 15km:3km resolution 1:5 scale nested simulation cannot run under the model setting of this study, the 1:3 scale is used to ensure the stability of the calculation). The tests adopt the same model settings as the SBM-NEW experiment except that four-dimensional assimilation is not used.

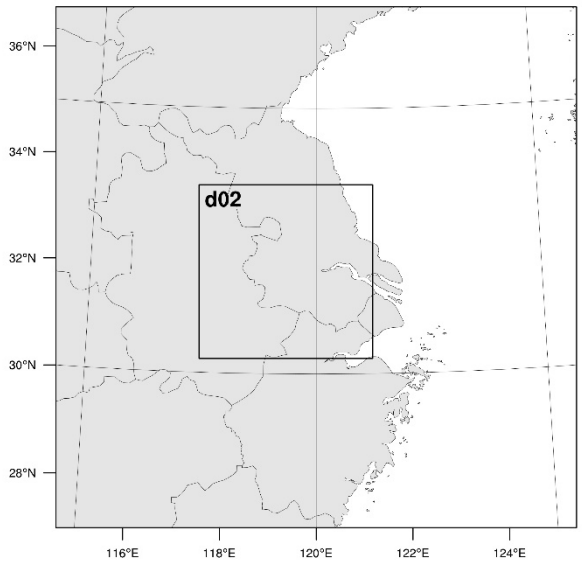

Figure R1. The model domain of the test

Fig. R2 presents N_d, cloud liquid water content (CLWC), water vapor content and supersaturation at 925 hPa with active aerosol-cloud processes simulated at different temporal resolutions from Test 1. The SBM simulations at 60s and 15s resolutions show the overall similar distribution, with only minor differences in N_d due to differences in resolving meteorological conditions and aerosol activation. The differences between the simulations of aerosol activation at the two temporal resolutions are specifically analyzed by selecting nine grid points according to N_d from high to low (locations shown in Fig. R2a). The analysis shows (Fig. R3) that there are some differences in the simulated N_d at different temporal resolutions, but the 60s resolution does not show clear negligence of activation processes and differences in the overall variation compared to the 15s resolution, and the differences in the simulated aerosol activation at the two resolutions mainly lie in the differences in meteorological and aerosol conditions at different time steps.

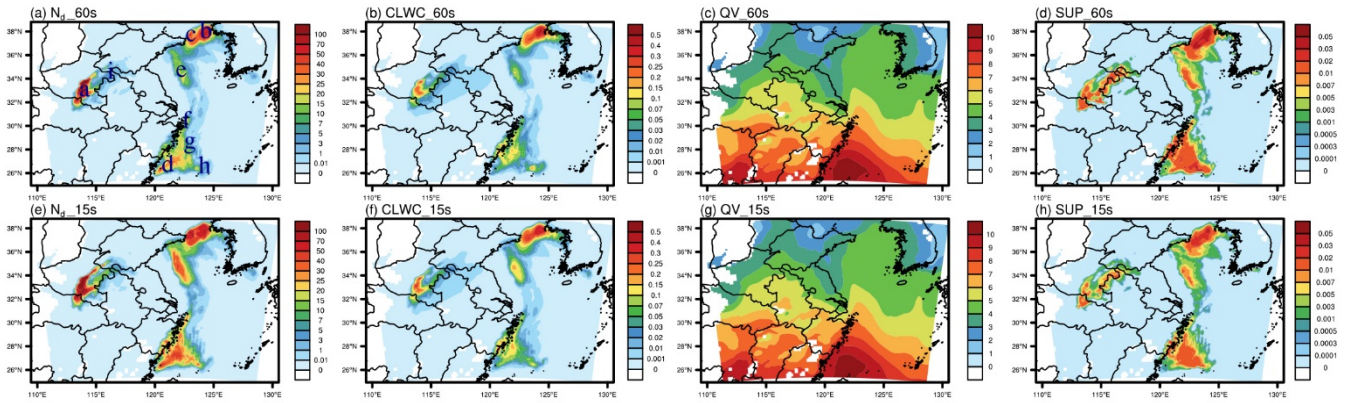

Figure R2. N_d (in cm^{-3}, a and e), cloud liquid water content (CLWC, in $g \cdot m^{-3}$, b and f), water vapor content (in $g \cdot m^{-3}$, c and g) and supersaturation (in %, d and h) simulated with 60 s (a-d) and 15 s (e-h) integration and 15 km spatial resolution at 925 hPa.

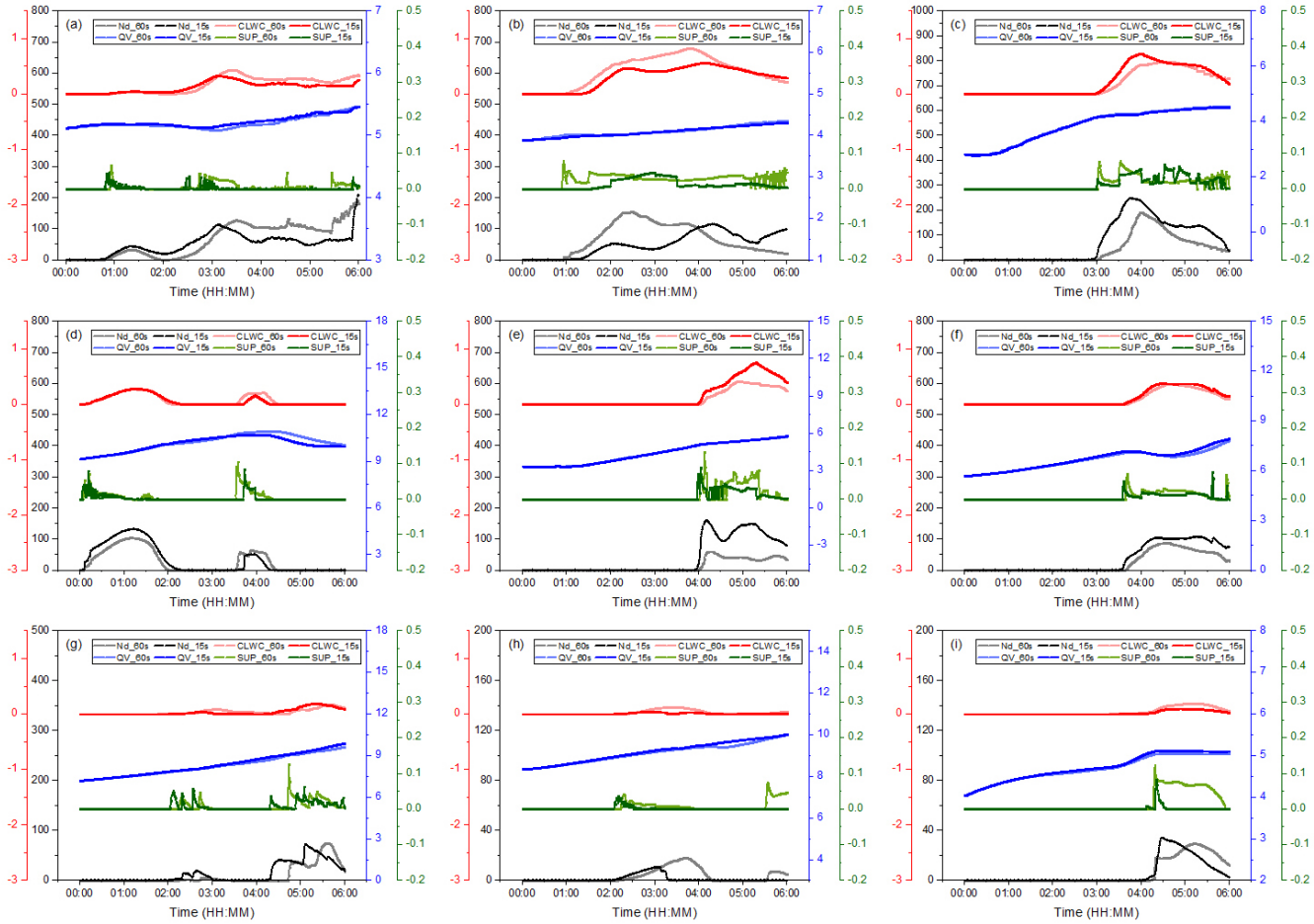

Figure R3. N_d (in cm^{-3}, black lines), supersaturation (in %, green lines), water vapor content (in $g \cdot m^{-3}$, blue lines) and CLWC (in $g \cdot m^{-3}$, red lines) simulated with 60s (light-colored lines) and 15s (deep-colored lines) integration (the locations of a-i are shown in Fig. R2a)

Test 2 then examines the ability of coarse spatio-temporal resolution to simulate aerosol-cloud variations. In test 2, only the data of d01 in the same area as d02 is selected to make the two match. As shown in Fig. R4, similar column AOD, N_d and precipitation as well as 925 hPa (layer with

active aerosol-cloud processes) CLWC, N_d and supersaturation distributions are simulated at 9 km and 3 km resolutions, while the 3 km grid relies on its finer resolution to simulate more detailed aerosol and cloud distribution information and stronger central intensity of precipitation. The analysis of the temporal variations of cloud parameters at each point on 925 hPa (sampled from high to low by N_d, locations shown in Fig. R4h) presented in Fig. R5 also supports the conclusion of Test 1 that the coarse resolution simulation differs only slightly from the fine resolution simulation in resolving specific processes due to differences in resolving meteorological fields, but both show consistent overall variations. In addition, the statistics on the model grid indicate that N_d with aerosol (Fig. R6) and CLWC with N_d (Fig. R7) show consistent trends at 9 km and 3 km resolutions, with the difference that the high-resolution simulation provides richer detail as well as a relatively larger number of intense activation samples and higher average N_d due to more fine resolving of the meteorological field. The lack of detail is compensated by more samples at a relatively larger spatial scale and longer time scale, and the lack of accuracy in resolving the meteorological field can be compensated by (1) optimization of the meteorological field simulation by four-dimensional assimilation, (2) resolving of sub-grid convection by cumulus parameterization, (3) inclusion of more abundant atmospheric processes at a larger spatial and temporal scale, (4) selection of winter period dominated by cloud types with large temporal and spatial ranges such as stratocumulus, and (5) filtering out of ice-phase processes when analyzing.

Due to (1) the better simulation performance of SBM compared to the bulk scheme for cloud macroscopic and microscopic properties at 15 km resolution demonstrated in Sect. 3.2, (2) the consistency of the simulated aerosol and cloud variations at coarse resolution with fine resolution revealed by the tests, (3) the aforementioned measures to compensate for the lack of meteorological field simulation ability at coarse resolution, and (4) the convenience of SBM for resolving aerosol particle size, aerosol composition and droplet size on aerosol-cloud interaction, it is appropriate to use 15 km in this study.

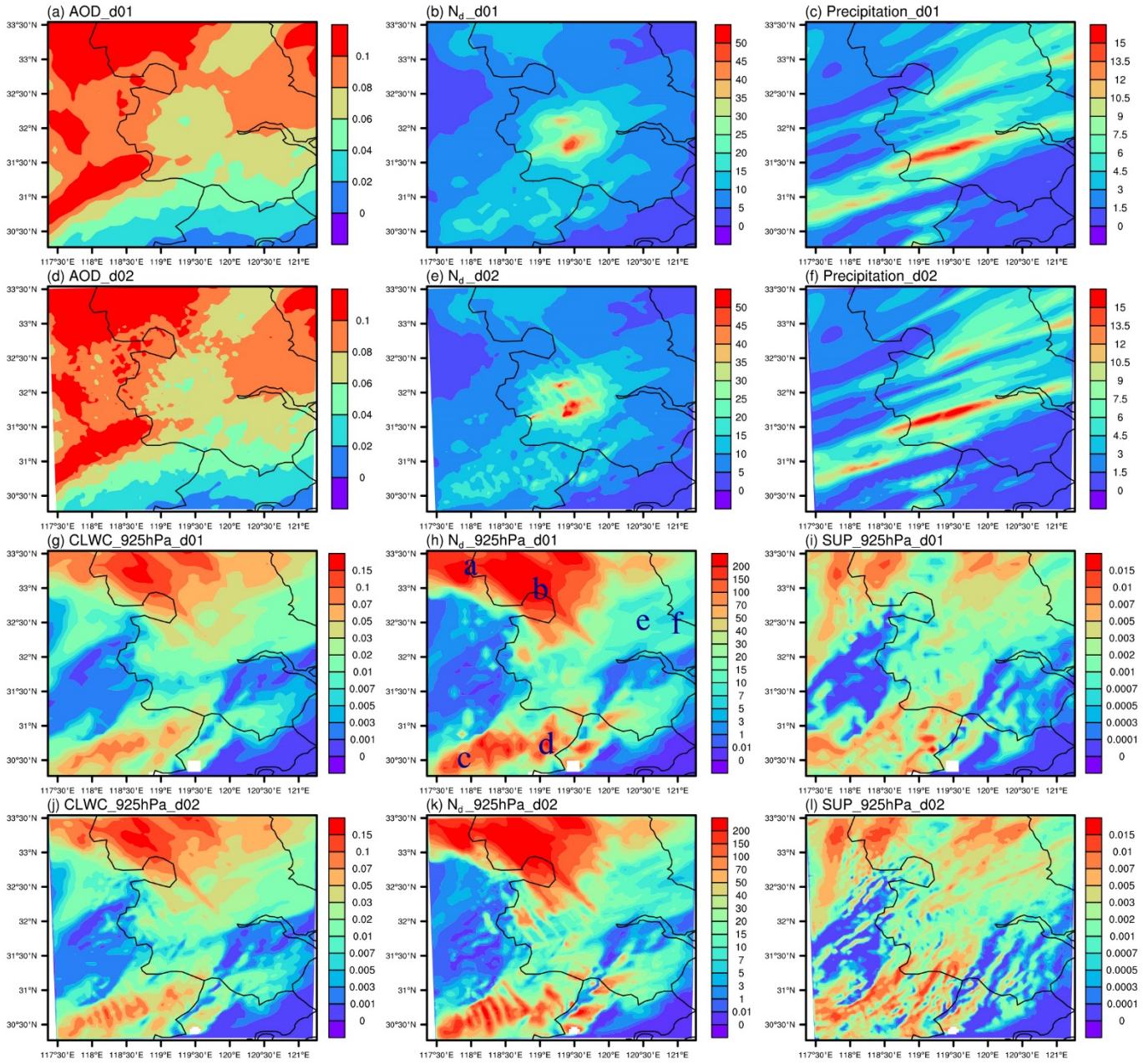

Figure R4. d01 (a-c and g-i) and d02 (d-f and j-l) simulated column AOD (dimensionless), N_d (in cm^{-3}) and precipitation (in mm) as well as 925 hPa CLWC (in $g \cdot m^{-3}$), N_d (in cm^{-3}) and supersaturation (in %)

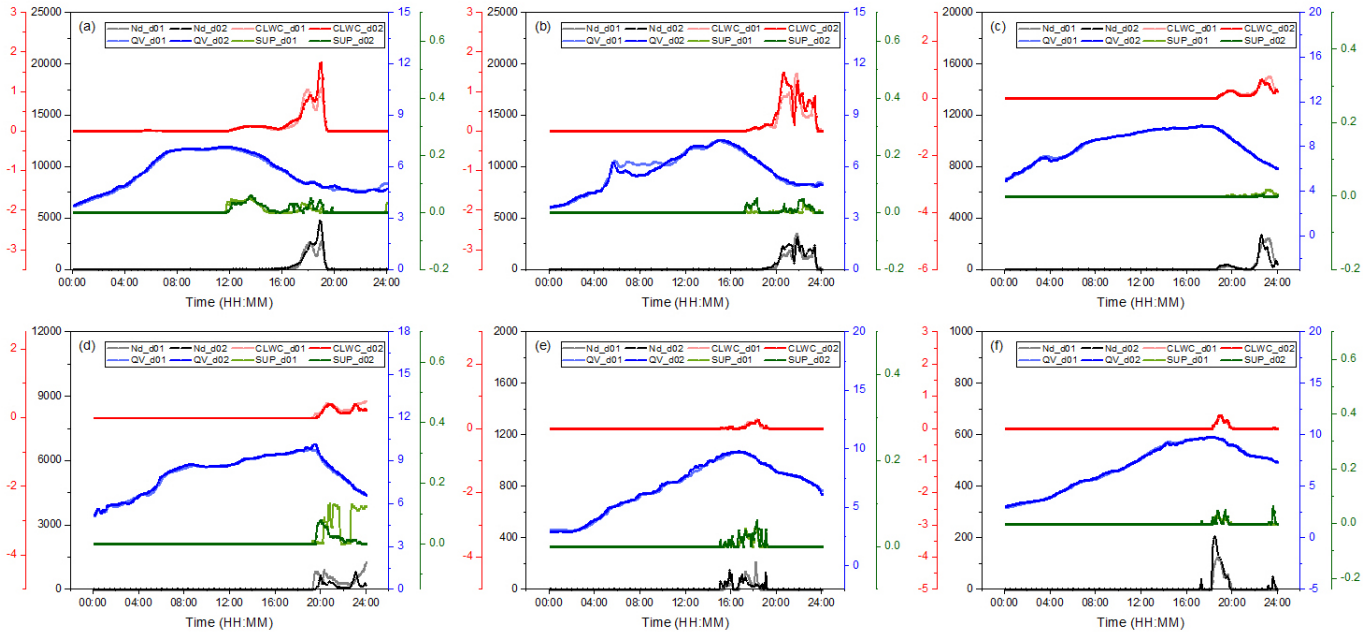

Figure R5. d01 (light-colored lines) and d02 (deep-colored lines) simulated 925 hPa N_d (in cm^{-3}, black lines), supersaturation (in %, green lines), water vapor content (in $g \cdot m^{-3}$, blue lines) and CLWC (in $g \cdot m^{-3}$, red lines) variations (a-f selected from highest to lowest according to N_d, as shown in Fig. R4h)

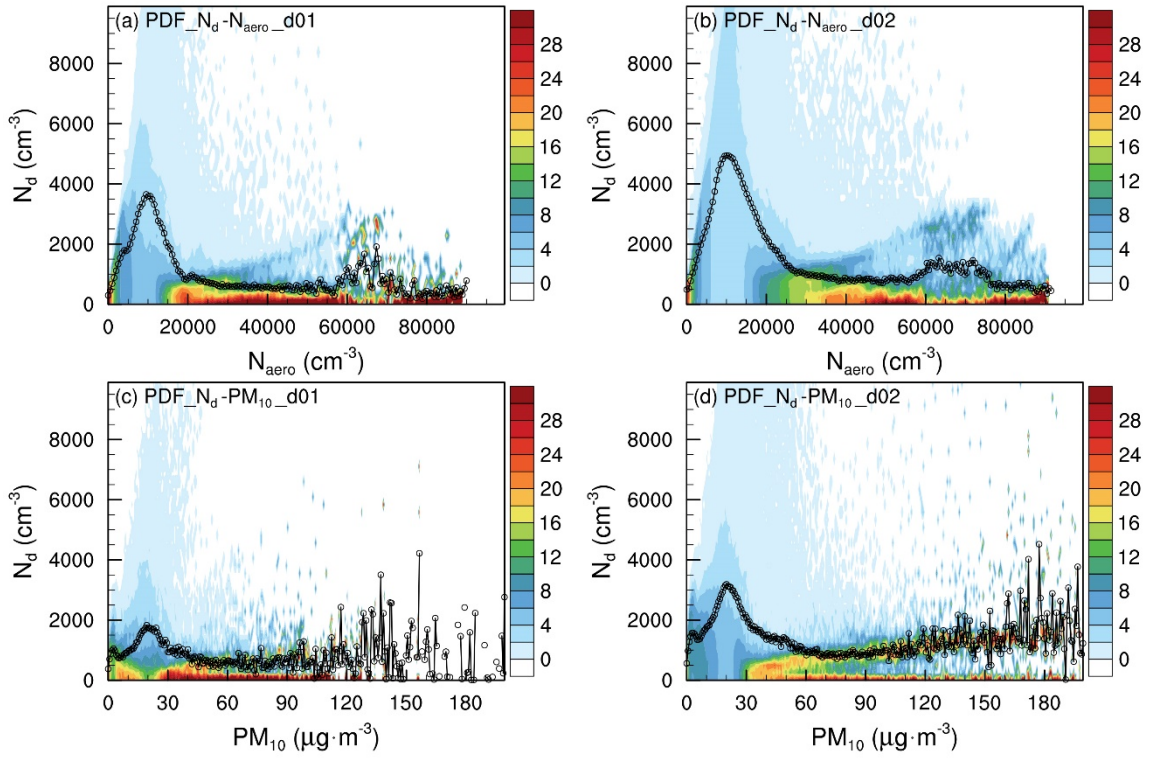

Figure R6. Probability density distribution functions and means (lines in the figures) of the simulated N_d relative to aerosol number concentration (N_{aero}) and PM_{10} (sum of probabilities corresponding to 1 for each N_{aero} or PM_{10} value) in d01 (a and c) and d02 (b and d)

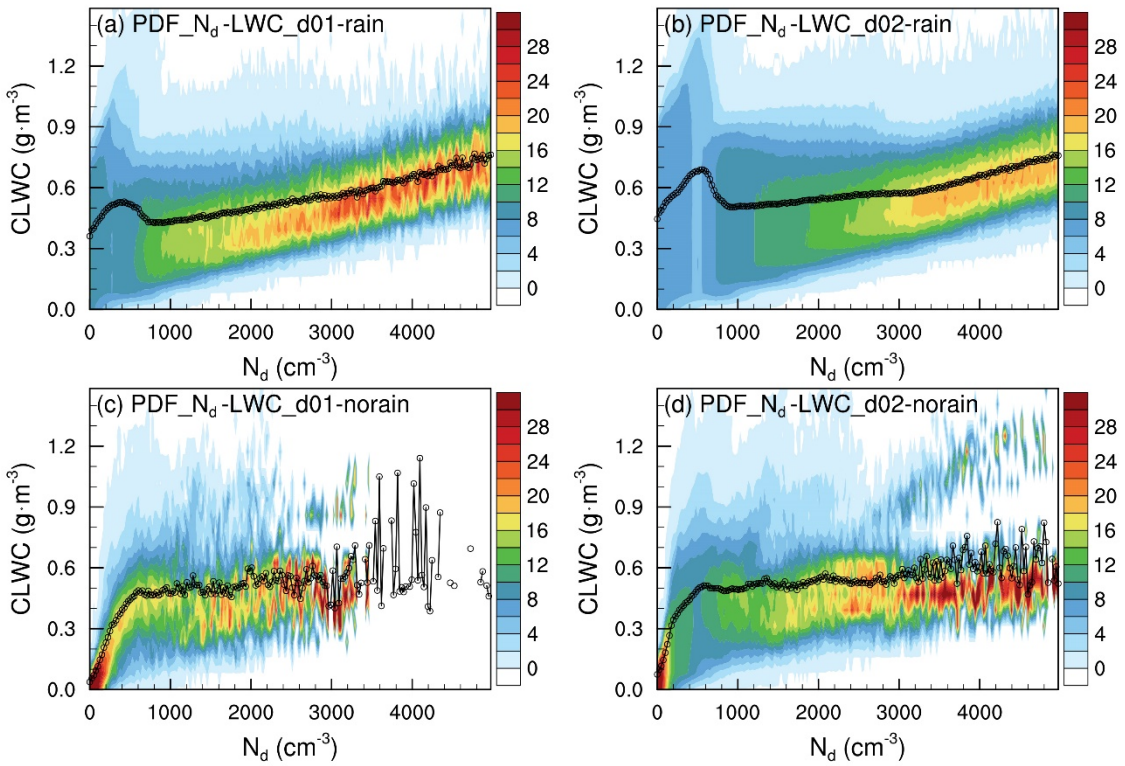

Figure R7. Probability density distribution functions (sum of probabilities corresponding to 1 for each N_d value) and means (lines in the figures) of CLWC relative to N_d of precipitation clouds (a-b) and non-precipitation clouds (c-d) in d01 (a and c) and d02 (b and d)