Reply to Anonymous Referee #3 Comments

We appreciate you for thoughtful and helpful comments. We tried to answer the referee's comments. Our 'Reply' is embedded below. We hope we provide the appropriate answers, and if there are more questions, please let us know.

The article presents simulations of marine stratocumulus clouds forming off the coast of East Asia using a bin microphysics scheme coupled to a mesoscale dynamical model from the Japan Meteorological Agency. The representation of aerosols in the model has been improved to account for more realistic multi-modal size distribution and multiple chemical components. The model simulations are driven by initial and boundary conditions from the meso-analysis dataset of the Japan Meteorological Agency for meteorological variables. Initial and boundary conditions for the aerosol are taken from the SPRINTARS model. The authors conduct two case studies with contrasting meteorological context and compare the improvements in the representation of aerosol and clouds with a previous version of the model and with observations from satellites and ground-based observations. Sensitivity experiments are conducted for both case studies by interchanging the initial conditions for aerosols or moisture. These sensitivity studies are used as a tool to investigate the effect of aerosols on marine stratocumulus in the East Asian region. It is found that the improvement of the aerosol parameterization and/or cloud microphysics in the model greatly improves the cloud microphysical and macrophysical properties when compared with the observations. In the sensitivity experiments, size distributions of cloud droplets showed a more narrow distribution if aerosol concentrations are increased, thereby also affecting precipitation development. Both findings are in agreement with findings from many other studies. Clouds growing in the polluted environment also showed the tendency of cloud thinning and higher cloud layers than those developing under maritime conditions. Sensitivity tests suggest that quantities such as effective radius and cloud droplet number concentrations are predominately affected by changes in aerosol number concentrations whereas changes in liquid water path and cloud optical depth are more influenced by changes in the meteorological conditions than changes in aerosols.

While the paper contains some interesting (rather technical) aspects such as the improvement of the microphysics in the model and comparison to observations, scientifically there is little in the manuscript that is really new and revealing. Although the general contents of aerosols, cloud microphysics and aerosol-cloud interactions would fit the scope of ACP, there is doubt that the manuscript meets the standards of ACP regarding novelty and scientific merit. Some of the details about the modeling framework are unclear, which leaves the reader with many open questions. Due to this lack of explanation it is also not clear if the modeling framework is really capable of realistically simulating the physics of aerosol effects on stratocumulus clouds. Although the manuscript generally has a good flow the grammar and wording would benefit from revisions. Some figures need improvements.

→ I appreciate your detailed comments. The main objective of this study is the application of complicated aerosol size and chemical composition into the 3-dimensional realistic model, which made the model to distinguish from different aerosol conditions (e.g., maritime or polluted aerosol), better simulating CN/CCN and cloud microphysical properties. Especially, the consideration on the complexity of aerosol conditions is very important in the investigation of aerosol-cloud interaction over this region, because the enhancements of aerosol loading from various source regions (e.g., dust storms from deserts located on the continent, anthropogenic pollutants transported from industrialized regions over the coastal area of China, and sea salt particles emitted over the oceanic area under strong wind conditions) have been predominant. With the distinction of aerosol characteristics, the investigations on low-level shallow clouds are performed under various environmental conditions by case studies on East Asia region. In particular, the simulation of low-level shallow clouds over East Asia is the first attempt with focusing the aerosol effects on cloud with high-resolution modeling. In the revised manuscript, we rewrite several sentences to emphasize the originality and the findings in this work.

We tried to provide clearly the details of modeling framework and to improve some figures as you pointed out. We also rechecked the grammar and wording in the whole manuscript.

A detailed list of review points is listed below.

1. Section 2.2: It is unclear how the aerosols are treated in the model. Prognostic or diagnostic? Clearly, in reality, aerosol size distributions can change significantly, regionally as well as temporally. How is this taken into account? Are aerosol dynamical processes such as coagulation, condensation, aerosol nucleation, aerosol scavenging treated in the model. How is the activation of aerosols to cloud droplets treated. Please clarify.

→ In our simulation, the size distribution function for CN (summation of five aerosol species) is discretized into 17 size bins, with the number concentration in each bin acting as a prognostic variable; the advection, diffusion, and activation to cloud droplets of CN are calculated at every time step.

Aerosol size distributions can surely change regionally and temporally mainly because aerosol chemical species are changing significantly. In this study, we determined the number concentration and aerosol size distributions for each of five different aerosol species. Hence, we can apply different aerosol size distributions and aerosol loading under inhomogeneous aerosol field from SPRINTARS simulation to different cases.

Unfortunately, aerosol dynamical processes are not considered in the present simulation. Regarding the activation of aerosol into cloud droplets, the critical radius of CN is calculated with maximum supersaturation for droplet radius from the Köhler equation, and then all CN larger than the critical radius are assumed to be nucleated to cloud droplet. We discussed those in Sec. 2.2 of the revised manuscript.

2. Section 2.2.1 and Fig. 1: As can be seen from Fig. 1, the representation of the aerosol size distribution and comparison to observations is clearly improved in the new version of the model. However, for some species such as sulfate, black carbon and organic carbon there is no observational evidence that would support the bi-modality in the aerosol spectrum. Thus, the magnitude of the fine mode aerosol is speculative at this point and there is no reasonable argument that it should exist at all. Also, mass closure would not be sensitive to fine mode aerosols. However, the cloud parameterization is likely to be sensitive to the fine mode aerosols especially in cases with higher updrafts that create enough supersaturation to activate these particles. For dust, the size distribution is overestimating the number of accumulation mode aerosols by one order of magnitude while underestimating the size. I think a better representation of the aerosol size distribution could be obtained by fitting log-normal size distributions to the observations and using the estimated parameters of the fitted size distributions in the model.
→ Newly applied aerosol size distribution parameters are determined as all three-mode log-normal form, fitted based on the previous observational studies (e.g., sulfate from Chuang et al. (1997) and Herzog et al. (2004), OC/BC from Penner et al. (1998) and Herzog et al. (2004), and sea salt and dust particles from d'Almeida et al. (1991)). Unfortunately, it is impossible to fit aerosol size distributions from Aitken to coarse mode aerosol particles using observational dataset from the campaign over East Asia due to the limited cut-off size, as shown in Fig.1. If we fit log-normal size distributions over only available size range from the observation in East Asia, CN and CCN number concentrations should be exceedingly underestimated at a given SPRINTARS mass concentration because the mass closure is sensitive to coarse-mode aerosol particles. Since the CCN nucleation process is sensitive to the fine-mode aerosol particles as you mentioned, we should adopt the fitted values from previous observation, supporting bi-modality in the aerosol spectrum. We believe that this is the best way currently.
It should be also noted that the determined size distribution of dust particles seems to be overestimating the number of accumulation mode aerosols by one order of magnitude while underestimating the size, possibly because of the contamination of other aerosol species. This should be upgraded to simulate the real world if there will be available and sufficient observation dataset over East Asia. We included it in the revised manuscript.

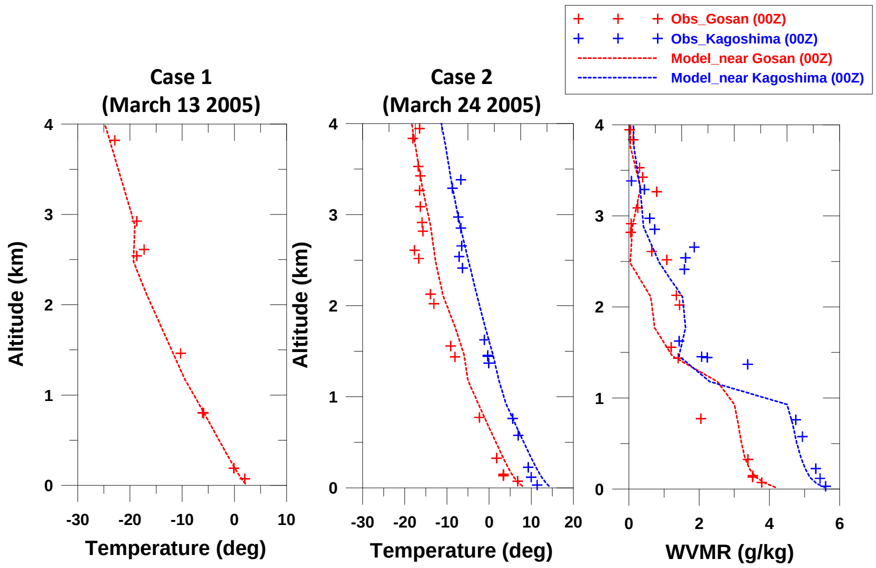
3. Section 2.2.2: The assumption of insoluble BC is certainly a limitation of this study and unrealistic. Many studies show that BC can be coated with sulfate, which renders mixed BC-SU aerosols generally good CCN.

→ BC-SU aerosols are internally mixed aerosol particle. Regarding the statement that BC can be coated with sulfate, therefore, it is difficult for us to apply this assumption in our simulations because our current model suppose that aerosol particles should be mixed with each other externally, not internally. BC insolubility in the present simulation should be definitely improved in further work.

4. Section 3: While a horizontal resolution of 3 km seems to be a reasonable compromise between the LES scale and the synoptic scale, both of which are important for stratocumulus clouds, the vertical resolution of the model seems to be insufficient to realistically capture features of the boundary layer. It would be interesting to see comparisons of boundary layer structure with observations to be able to judge how well the model can handle the boundary layer dynamics at this coarse resolution. Otherwise, it is unclear to what extent the model is capable of simulating aerosol effects on boundary layer clouds through microphysical feedbacks that are ultimately linked to changes in the PBL dynamics.

→ Basically, the cloud development was sensitive to the vertical resolution in cloud modeling study, especially for stratocumulus clouds. However, Guo et al. (2008) found that basic features of the integrations (cloud liquid water path and optical thickness) are similar for vertical resolutions of 40 m or finer. In addition, Lee et al. (2009) reported that the qualitative nature of simulation results in robust to the vertical resolution with nearly identical results between the vertical resolutions of 40 m and 15 m.

The vertical profiles of meteorological parameters shown in Fig. 3 are the horizontally averaged values for each level using JMA-MANAL dataset, thus the profiles of meteorological parameters must be varying for every grid point. We compared the averaged vertical profiles near observation sites with sonde dataset at Gosan and Kagoshima (observation sites in the target area in our simulation) where the sonde observation has been performed continuously (Please see Supplement 1 in the next page). Unfortunately, the dataset is not available for case 1 except the temperature profile at Gosan. Overall, the simulated vertical profiles look well matched with observation dataset, though the temperature profiles do not show clear PBL structures. Due to the insufficient observational dataset, we could not discuss it in the revised manuscript.



Supplement 1. Comparisons of simulated vertical profiles (indicated as dotted line) of temperature and water vapor mixing ratio (WVMR) for both cases 1 and 2 with sonde observation at Gosan (red cross) and Kagoshima (blue cross)

5. P. 23460, L. 12ff: Even if the wind directions are similar in both cases there are clear dynamical features (e.g., cut-off low type feature, vorticity) that are completely different and clearly impact cloud development. However, these features are not discussed by the authors. I also recommend to show only the relevant parts of the weather charts in Fig. 2 (e.g., only part that overlaps with model domain).

→ Differences of cloud development between cases 1 and 2 resulted mainly from the distinction in moisture amount near boundary layer, rather than from different dynamical features (e.g., wind direction and speed, and vorticity, etc.) in our case simulation. The obvious difference in moisture amount between cases 1 and 2 was already discussed with Fig. 3.

Also, we truncated the all four edges of weather chart in Fig. 2 and enlarge it. In fact, the part of weather chart is a little bigger than you recommended, covering the whole the northeast Asian region, because it is difficult for us to distinguish between high and low pressure system in only relevant part of model domain.

6. P. 23461, L. 15ff: I think one fundamental question that has not been addressed here is to assess how the SPRINTARS model compares with observations for the particular case studies. E.g., one could compare the aerosol fields against observations from the ground based sites to get a first clue on how SPRINTARS compares. This is important because later in the text the authors compare the regional model with observations but it is unclear if

the regional model does a better job than SPRINTARS.

→ The SPRINTARS model have been performed during the springtime over East Asia and compared with satellite observations, suggested by Fig. 8 of Nakajima et al. (2003) and Fig. 4 of Nakajima et al. (2007), in terms of aerosol optical properties. For aerosol number concentration, the model has not been fully evaluated mainly because of limited observation. Based on the previous studies, it is obvious that SPRINTARS simulations over East Asia have underestimated aerosol optical thickness (i.e., aerosol loading), resulting in the less mass and number concentrations than those in the real world.

7. P. 23463, L. 14ff: What does the uncertainty range mean for the model simulations? Why is there uncertainty and why is it so large? Why do the authors compare averaged values instead of comparing observations from the ground-based site against nearest grid point values? If the model has a horizontal resolution of 3km a point to point comparison should be reasonably fair.

→ The uncertainty ranges stand for the standard deviation of CN and CCN number concentrations over the region of 32.5-33.5°N and 125.5-126.5°E with a size range of D_p larger than 10 nm to compare with the single-point observation dataset at Gosan. However, the averaged area looks large because the spatial resolution was set to 3 km, as you mentioned. Therefore, we reset the averaged area to the smaller size (33.1-33.5°N and 126.0-126.4°E), centered on the observation site, than the previous work. Then, we compared the values at the nearest grid point from the ground-based site with the averaged value for small area, which shows very similar values between them (e.g., 2129.5 cm^{-3} for the nearest grid point, and 2117.3±44.89 cm^{-3} for the area-average in CN number concentration of case 1). In fact, model simulation can generally lead to the uncertainty spatially and temporally, thus we decided to use the area averages for 33.1-33.5°N and 126.0-126.4°E in this study. Newly calculated values of CN and CCN number concentrations are included in the revised manuscript.

8. Although the simulations with improved microphysics are closer to observations, which is encouraging, the overall comparison with observations could not be much worse. The aerosol concentrations are off by more than 30% in both cases and the CCN concentrations by more than 300% in case 2. This leaves the reader wondering if the "good" comparison in case 1 is just by chance or just pointing to deficiencies in the model's microphysics to get the correct number of CCN from erroneous number of aerosols.

→ There is still a gap between calculated and observed CN and CCN concentrations in spite of the improvement, especially for polluted case (case 2). Firstly, this discrepancy possibly resulted from the underestimation of aerosol mass concentration by SPRINTARS especially in polluted condition or from imperfect size distributions of pollution aerosol such as sulfate and organic carbon (too much shifted to Aitken-mode particle which is difficult to be activated to cloud droplet), rather than observational uncertainties. Secondly, inhomogeneous spatial distributions of aerosols can be attributable to this discrepancy, because direct comparison of simulated CN/CCN values with 3-km horizontal resolution with the observed data at a point (i.e., Gosan site) was made in this study. This is discussed in the revised manuscript.

9. P. 23465, L. 13: Fig. 6 suggests that there are clear regional differences. I am not convinced how meaningful a comparison of averaged values is in this case. Also, comparing the model with MODIS observations shows large discrepancies in the spatial patterns. Yet the authors argue that the comparison is reasonable. It would be good to compare also cloud droplet number concentrations and cloud fraction with corresponding MODIS products. Fig. 6 would greatly benefit from a different color scale. It is difficult to discern differences if almost all values have very similar shading.

→ Fig. 6 displays the simulated variables over whole model domain, showing inhomogeneous cloud field as you mentioned. We focus on low-level shallow clouds over the oceanic area which the dotted box in Fig. 2 indicated. The comparison of averaged values was performed for the limited area of 125–132°E, 28–33°N and 125–129.5°E, 30.8–33.2°N for cases 1 and 2, respectively.

The distributions of simulated variables are slightly shifted to the southeast, compared with MODIS observation, but the spatial patterns look similar with MODIS retrievals in case 1. However, the simulation for case 2 does not reproduce well very shallow low-level cloud which is shown in MODIS retrievals. This is possibly because NHM tends to simulate less low-level cloud with its PBL parameterization for the condition of shallow boundary layer. The resolving the problem in NHM is not regarded to be out of scope in this study. We included these spatial discrepancies additionally in the revised manuscript.

Regarding the comparison of other variables from MODIS product, cloud droplet number concentration is estimated by several assumptions using cloud effective radius and optical thickness, not directly retrieved from MODIS radiances. Because we already showed the comparisons of effective radius and optical thickness, thus we don't think CDNC comparison is required between model simulation and MODIS observation.

As you recommended, we modified Fig. 6 in a different color scale to distinguish the differences between model simulation and MODIS observation.

10. P. 23466. L. 9: So far, the manuscript exclusively deals with the evaluation/comparison of the newly implemented microphysics parameterizations and not at all with aerosol effects on Sc cloud as suggested by the title. Section 5 covers the aerosol effects by means of sensitivity studies but fails to provide any content that is scientifically new. At this point

I wonder if it would be better for the manuscript to focus on the implementation/evaluation part only, change the title accordingly and resubmit the manuscript to a different journal.

→ As we reply to your general comment, our implementation of the new realistic aerosol size distribution and chemical components resulted in a better simulation of cloud microphysics, which can also make the model to distinguish from different air mass (e.g., maritime or polluted aerosol). Especially, the consideration on the complexity of aerosol conditions is very important for the investigation of aerosol-cloud interaction over East Asia. This study also covers the investigations on low-level shallow clouds performed under various environmental conditions by case studies on East Asia. In particular, the simulation of low-level shallow clouds over East Asia is the first attempt with focusing the aerosol effects on cloud with high-resolution modeling. In the revised manuscript, we rewrite several sentences in the introduction and summary to emphasize the originality and the findings of this work.

11. P. 23468, L. 7: In my view a significant advantage at simulating aerosol-cloud interactions with a regional model rather than a LES is to capture mesoscale dynamical feedbacks that stem from the cloud microphysics and feed back to the cloud dynamics. However, it is unclear if a simulation time of 9 h would be sufficient to capture these effects. Can the authors comment on what motivated the simulation time of 9 h?

→ Some studies using a LES model for idealized cloud simulations (e.g., Khain et al., 2005, Khain et al., 2008) already reported the feedback effect from cloud microphysics to cloud dynamics through latent heating/cooling; this feedback effect depends on the types of clouds and cloud systems (Khain et al., 2008, Fig. 17). Their most simulations were conducted for less than 5 hours. In addition, Iguchi et al. (2008) discussed this kind of effect using a regional model and an integration time of 9 hours is sufficient to capture the effect as shown in their Figs. 18 and 19. Thus we believe that 9 hours is enough in this case. However, other feedback effects with a long term may exist and should be resolved in a regional model simulation, e.g. a feedback effect through radiation. We will address such kind of effects in the future works because a high computational cost of using a spectral bin microphysics restrict the integration time now.

12. P. 23474, L. 15: The authors conclude that clouds in polluted aerosol conditions show tendencies towards cloud thinning and higher cloud layers and speculate that this may be related to updrafts. However, differences in updrafts and/or boundary layer dynamics have not been looked at so far. How can the authors arrive at this conclusion?

→ In the manuscript, we explained phenomenon due to the strengthened updraft (the maximum difference between M_{humid} and P_{humid} is about -3.1 cm s^{-1}) and a little wider convective area with updraft by 2.2 % near the cloud layer. The target cloud is not convectively active, thus we suppose these small differences can cause the cloud thinning and higher cloud layer in polluted condition (P_{humid}).

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