Dear anonymous referee #2,

We very much appreciate your constructive comments and your time for RC2. Thanks to your review, our manuscript was substantially improved, especially for the organization of manuscript. We considered all your comments into the revised manuscript.

Point-by-point responses to your comments are written in blue in this letter.

With best regards,
Mizuo Kajino

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[1] The paper of Kajino et al. compares the three aerosol representations (bulk, 3-category, 5-category) which are available in the regional CTM NHM-Chem. The results of air quality related variables (O3, NO2 etc) are compared with observations. Further, CCNs and AOT etc. are compared between the three aerosol representations. The paper is a resubmition of a paper which was rejected earlier in GMD.

I already reviewed the rejected earlier submission of the paper. The focus has now changed from a model description paper to a model evaluation paper. The manuscript fits well into the scope of the journal. The paper is very long and parts of it could be shortened, while some parts could be improved with adding some more discussion.

[1] Thank you for your evaluation and reviewing our manuscript many times. Yes, it was long. We shorten the paper by moving general description of results parts to Supplements according to your comment. Also, additional discussion is made in Sect. 7 based on your comments and in the beginning of Sect. 6 according to RC1. Please refer to RC2 [5] and RC1 [11].

[2] Further, to my opinion the language should be improved at some points. In general, the paper needs some larger revisions before it can be accepted for publication.

[2] The manuscript was sent to language editing just before submission, and so we believe that the sentences are grammatically correct. However, we admit that some parts were not clear. We tried our best to improve the language so that the sentences are clear and concrete so that readers can easily get the meaning. For example, as you commented in RC2 [9] as well as RC1 [20], "The initial and boundary conditions should

be improved before model formulation" was really awkward. We tried to remove such confusing and unclear statements in the revised manuscript.

Major comments:

[3] To my opinion the figures are partly confusing. Some of the figures show Bulk/5-cat, 3-cat/5-cat, 5-cat, other show 3-cat, 5-cat, 3-ca/5-cat. An example are Fig. 3 and Fig.4. To add a little bit of confusion the caption of Fig 4 says (same as Fig. 3). I think it would be much easier if all figures would have the same design. Similar for example Fig 8 and 6/7 or Fig 9 and Fig 10.

[3] Thank you for your comments. We agree that all figures should be harmonized. In the previous manuscript, we had Fig. 8 (fog deposition) because fog deposition scheme is a feature of NHM-Chem. However, because the manuscript was already lengthy and because the fog deposition was not pronounced in this crude grid size, we moved Fig. 8 to Supplement 5. So, previous Figs. 9-12 were shifted to Figs. 8-11. Also, it is meaningless to compare Bulk for climate-relevant variables, because Bulk was not designed to simulate them. AOT is regarded as a climate-relevant variable but, satellite AOT is often used in the data assimilation for the operational forecast of aerosol mass concentrations. Thus, the result of Bulk was also presented in Fig. 8 (AOT).

After all, all figures are harmonized but in two different ways as follows:
Figures 3-8 (O_3-AOT): Bulk/5-ctg, 3-ctg/5-ctg, 5-ctg
Figures 9-11 (AAOT, BC, CCN): 3-ctg, 5-ctg, 3-ctg/5-ctg

[4] Currently, there is no coupling of meteorological variables and aerosol, which is a major shortcoming of the study. This should be clearly mentioned and discussed (see also Reviewer 1).

[4] Yes, we have mentioned it in several locations but it was not appealing. Please refer RC1 [2]. We inserted the following sentences in Abstract and first paragraph of Conclusion:
(implementation of aerosol feedback processes to NHM-Chem is still ongoing)

[5] The authors included a lot of information to the supplement, but sometimes discuss this supplementary material very detailed. As an example see page 20 l5ff. Either this are supplementary information (what is fine) or this are no supplementary information. If this are no supplementary information the figures should be included into the

manuscript. If this are supplementary information the long description should be moved to the supplement to shorten the paper a little bit.

Generally, the paper offers a lot of analyses and information, but the results are of course only valid for the NHM-CHEM model. Therefore, I ask the authors to shorten parts of the general description of the results a little bit and put more empathize on the following points:

1) why are these variables important (see reviewer 1) 2) Discuss similarities /differences of the aerosol representations available in NHM-Chem with aerosol representations available in other models 3) Discuss which findings most important for other aerosol models

[5] Thank you for your useful comments. The general descriptions of the results such as comparison with observations were also presented in our previous paper (Kajino et al., JMSJ, 2019), and thus removed from the revised manuscript. Please refer to the additional statements at the end of Sect. 5 "Observations and model validations", as follows:

"Because the main objective of the paper is the aerosol module intercomparison, the general description of the results, which were already made in our previous paper (Kajino et al., 2019a), are not presented in detail here, but are presented in Supplement 3."

Accordingly, parts of 3rd, 4th, and 5th paragraph of Sect. 6.2, the entire part of 2nd paragraph of Sect. 6.3, and a part of last paragraph of Sect. 6.3, were moved to Supplement 3. One paragraph of Sect. 6.4 describing the comparison for the aerosol optical properties was also entirely moved to Supplement 3. A part of the first paragraph of Sect. 6.5, as well.

General features are presented in Supplement referring the main texts, but main texts do not refer materials in Supplement in the revised manuscript.

In terms of the important points as you raised, 1), 2), and 3), the point-by-point answers to the items are summarized as follows

1) WHY ARE THESE VARIABLES IMPORTANT: "Air quality variables" surface mass concentrations of O_3, mineral dust, and $PM_{2.5}$ are important which negatively impact the health of the population and the environment. Depositions of SO_4^{2-}, NH_4^+, and NO_3^- caused environmental acidification. "Climate relevant variables" AOT, AAOT (dust and BC), CCN, and INP (CCN containing dust and BC, acting as immersion freezing) involves in aerosol-cloud-radiation interaction processes which alters energy budgets and precipitation, but are still uncertain.
   → We summarized this point in the 1st paragraph of Sect. 7

2) DISCUSS SIMILARITY/DIFFERENCES WITH OTHER MODELS: "3-category method is widely used method in air quality model such as CMAQ and WRF-Chem, and so our 5-category approach has an advantage. These aspects (external mixture of BC and dust) are already considered in many of climate models, but chemical mechanisms are usually simplified in the climate models.
   → We summarized this point in the 2nd paragraph of Sect. 7

3) DISCUSS WHICH FINDING IS MOST IMPORTANT FOR OTHER MODELS: Our message is mostly for air quality modeling communities. "Bulk method is no more used in air quality models, but still useful for operational forecast. The 3-category is less accurate when the sea-salt and dust coexist in an air mass (Gobi dust transport in Northwest Pacific or Saharan dust transport in Mediterranean or Atlantic) or when considering the light absorption and INP activity of BC and dust in the aerosol-cloud-radiation feedback system.
   → We summarized this point in the 3rd and 5th paragraphs of Sect. 7

Minor comments:

[6] P4l20ff: The discussion about what schemes/couplings have been used in which publications is were long and could be removed without loosing important information for the paper.
[6] We decided the maximum number of citations to two for each sentence. Please see the last paragraph of Sect. 2.

[7] The description in p12l5ff about the temporal length of the simulations is very confusing and should be rephrased
[7] (1st paragraph of Sect. 4.1) We deleted the dates because it is clear that "simulation

starts from January with spin-up of 5 days" means "simulation starts from 27 December". This modification enhanced the clarity of the way of time integration. Thank you for your suggestion.

[8] The sentence 'We applied the monthly mean values of GFED3 without temporal variations" is unclear (the same monthly means in each month?)
[8] (1st paragraph of Sect. 4.2) We modified it to "without daily and diurnal variations".

[9] The part in the conclusion: 'The initial and boundary conditions should be improved before model formulation. The difference in PM2.5 was large, i.e., up to 20-100%. Improving the model formulation, as well as its initial 25 and boundary conditions, is needed.' is unclear. What exactly and why should initial and boundary information be improved?
[9] Yes, the sentence was very confusing. We intended to say that data assimilation (improving initial and boundary conditions) can be used to enhance accuracy, even though the forward model is not improved. We thoroughly reorganized this paragraph of Sect. 7 "Conclusion and discussion" (3rd paragraph in the revised manuscript) as follows:
"The bulk equilibrium method was evaluated for the eligibility of operational forecast, namely, the surface mass concentrations of air pollutants such as O_3, mineral dust, and $PM_{2.5}$. The differences in the simulated seasonal mean concentrations between the bulk method and the 5-category method were smaller than 5% and 5-10% for O_3 and mineral dust, respectively. The difference in $PM_{2.5}$ was large, i.e., up to 20-100%, due to the neglection of nitrate mixed with dust particles of the bulk equilibrium method. Still, however, the statistical scores of the bulk method regarding $PM_{2.5}$ were not very different from the other methods. In order to fill the gap between observations and simulations, operational forecast is associated with data assimilation and post-process of statistical bias correction. The bulk method is not used in recent air quality models any more. Still, however, as the model performances were similar with each other, the faster bulk equilibrium method can be recommended for the use of operational forecast."
I hope this improvement is clear.

[10] I suggest to rename the conclusion into 'conclusion & Discussion'
[10] We changed it.