Dear anonymous referee #2,

We very much appreciate your constructive comments, useful information and your time for RC2. I am sorry for the inconveniences about all the duplications of the current manuscript against an accompanied paper, submitted to *Journal of Meteorological Society of Japan* (*JMSJ*). The two manuscripts were originally a single manuscript, which was too lengthy, far beyond the limitation of *JMSJ*. I had to divide it into two and submitted separately one to *GMD* (mostly model description and performance differences due to representation) and the other to *JMSJ* (suggestion of model evaluation methods), but the separation was not really complete. I considered *GMDD* as a model description paper, because it is expected to be published as a discussion paper much sooner than *JMSJ*. However, after considering your comments and the other referee's comments (RC1), we feel that it is more natural and favorable to regard the *JMSJ* manuscript as a model description and evaluation paper and regard the current manuscript as the second paper about performance evaluation with respect to different aerosol representations. Hopefully, the *JMSJ* manuscript will be accepted soon. Therefore, we will completely re-organize the current manuscript by carefully avoiding the duplications from the *JMSJ* paper before the resubmission. The title of the revised manuscript will be "Comparison of three aerosol representations in the predictions of mass concentration, deposition, and climate-relevant variables in NHM-Chem v1.0".

Considering the above situations, we would like to finalize the revision of manuscript, after we find that the *JMSJ* manuscript is very close to its final form, in order to avoid the duplications. Four additional weeks are granted for the preparation of the revised manuscript, but I am afraid that we cannot make it within the time. I will try my best but we might extend the due date of the revision, if the review process of *JMSJ* delays. Thank you very much for your patience and kind understanding.

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

With best regards,
Mizuo Kajino

The paper by Kajino et al. 2018 introduces a new regional chemical transport model NHM-Chem based on the mesoscale non-hydrostatic meteorological model NHM of the

Japan Meteorological Agency. It primarily compares 3 existing aerosol representation schemes with varying complexity, namely a simple bulk, a 3- and a 5-category method. As such, the paper fits to the scope of this journal, but however, significant details are missing in order to get a complete picture about the model system as indicated in the title.
Thank you for your evaluation.

In my point of view, the paper has to include the most important features of this new system, core developments, model setup (domain etc.), important schemes and improvements compared to other existing (coupled meteorology-chemistry) models such as WRF-Chem, WRF-CMAQ, EMAC, COSMO-ART. Presented aerosol schemes should be set in context to other existing schemes such as GMXe (Pringle at al. 2010) and the author should further include a statement what makes their most sophisticated aerosol scheme unique compared to others found in literature. The title in a way is misleading, as it assumes a detailed description of the model also including model evaluation. The latter is not represented in the current version of the GMDD paper.
I am sorry for all this confusion. We decided to change the title and the revised manuscript is no longer a model description paper. Still, we will include the description and discussion about the differences with other existing models, which you introduced. Also, please note that we will remove “unique” from the manuscript, as it was overstating.

The authors already made an attempt to provide these information in a similar paper in another journal, which however is not published yet. In my point of view a better way would have been the publication of follow-on papers in the same journal (compare: MESSy, COSMO/MESSy in Jöckel 2004, Kerkweg 2012 and follow on papers). As already mentioned by other reviews, the author should definitely check for text duplication between both manuscripts and provide a suggestion how to tackle this issue.
I agree with your point. As I replied to the Referee #1’s comment and as previously mentioned, we will completely separate the two papers by removing the duplications throughout the current manuscript, before resubmission. Also, thank you for your introduction of the above references.

For these reasons, I unfortunately cannot recommend the paper for publication in GMDD in its current form but am willing to consider to review a revised version. To support this statement I have added further comments below:

Thank you very much. We look forward to your review on the next re-submission, after improvement according your comments.

Introduction

There is a focus on aerosols only. Please highlight other chemical compounds and reactions used in this study (e.g. ozone). As mentioned above, it would be interesting to learn more about the intention of developing this new model, meaning how it sets apart from other existing systems and what does it have in common. Which processes should be improved and which areas of research benefit from this model. These aspects are mentioned briefly (P2, Line 31-32), but however need more clarification. In order to get a feeling of the model performance (P3, Line 8), comparing the model results to observations is a crucial part.

Thank you for your comment. We will improve the Introduction, carefully reflecting your comments.

Model description

This section (particularly 2.1) is too short and needs more details on most important schemes, model setup, configuration, domain (Figure needed) and also details on technical realization of the coupling between meteorological and chemical model. As parts of the above mentioned aspects are mentioned later in Chapter 2.4, Chapter 2 needs to be re-structured.

Even though, the revised manuscript will be no longer a model description paper, we will shortly but clearly describe the model descriptions in Sect. 2.1 together with re-organizing Sect. 2.4.

Some minor points:

P3,L14: specify term ‘acusa’ and provide literature

asuca (Asuca is a System based on a Unified Concept for Atmosphere)

P3,L15: simpler than what?

simpler than the current three categories presented in the manuscript.

P4, L4: identical vertical resolution just for your selected model configuration?

Yes. Rather “common” than “identical”.

P5, L9: You mention two options to calculate the efficiencies. Which option did you chose and why?

We used Kajino and Kondo (2011) here in the study, because of computational efficiency.

P6,L9: short definition of internal and external mixing should be provided before.

Thank you. The terminology of mixing state is slightly different among users. I modified the sentence as follows: “BC is usually hydrophobic when emitted (externally mixed with other aerosols) and become more hygroscopic due to the condensational growth of water-soluble inorganics (internally mixed), such as sulfate, nitrate, and ammonium, during transport.

P6,L11: Are dust plumes a regular feature of that region, how are they linked to meteo-rological conditions and where are they originated from. Please summarize the findings of Zhang and Iwasaka 2004 in this context.

In Japan, massive transport events of mineral dust originated from Chinese arid regions such as Gobi and Taklimakan deserts often occur in the spring associated with the cold front of migrating anticyclones. Zhang and Iwasaka (2004) found 10-20% of number fractions of coarse mode particles were the mixture of mineral dust and sea-salt with similar mass fractions, during the dust events in the spring of 1996. They also found only 5-15% of pure dust particles and as much as 60-80% of the mixture, including dust slightly mixed with sea-salt and sea-salt slightly mixed with dust.

P6,L31: Is any data-assimilation used in your study?

Data assimilation was only applied to meteorological fields and not applied to chemical fields. (It is applied for the current JMA’s operational forecast of oxidant, though).

P7,L11: Numbers have to be provided in respective table.

We will put numbers in the table.

P8,L26: The actual model domain is hard to figure out from Figure 2-10. How many domains where used? Nesting? How was the coupling achieved?

Single domain with 30-km horizontal resolution was used. Each panel of Figures 2-10 is the domain. We will add one figure showing the model domain in the revised manuscript.

P9,L13: Please indicate the grid resolution of the anthropogenic emission.
We will add it.

P9,L19: Please provide more detail on the calculation of the biogenic flux.
We will add it.

P10, L20ff: Where are the values originated from?
The values of $D_{g,n,dry}$ of dust and sea-salt are derived from their original size distributions of Han et al. (2004) and Clarke et al. (2006) by assuming the prescribed geometric standard deviation σ_g as 2.0. We will modify the sentence to avoid confusion.

Model performance

P11,L24: In my opinion the term operational forecast is misleading (see also at other locations in the manuscript) as the study rather discusses a 1-year hindcast simulation for 2006 than an operational forecast. Further it is not entirely clear how the scope of Chapter 3.1 and 3.2 differ. They could potentially be combined.
Yes, it was confusing. We will make a clarification in the revised manuscript that the hindcast of mass concentration can be regarded as an indicator proving that the bulk equilibrium method is durable for the operational forecast. In the revised manuscript, we have two sections but with the different separation, Sect. 3.1 for concentration and Sect. 3.2 for deposition.

General: How does Ozone link to dust and PM2.5 concentration. Please discuss why O3 differs for different aerosol representations. What are the respective pathways and reactions?
Thank you. We concluded that O_3 were not different among the methods, but actually somehow different. It was due probably to different aerosol surface areas (different heterogeneous loss rate of NO_x, as a precursor of O_3 formation) and different aerosol pH (different loss rate of O_3, in the production of aqueous phase sulfate).

P12,L1: due to the small figure size, the prevailing wind patterns are hard to capture
We will emphasize the wind arrows in the figures.

P12,L3: please specify, provide more details on the ‘source region’. Why is concentration highest in spring?

We will modify the sentence as follows: “highest in the summer over China but highest in the spring in Japan due to the long-range transport associated with the travelling disturbances”.

P12,L16: clarify the term ‘operational forecast issue’

This sentence will be deleted as it was confusing. It meant that the 3-category and 5-category are not designed for operational forecast, and thus the comparison between the two methods is not relevant issue for the operational forecast”.

P13,L2: Can you provide proof for the overestimation of NO3-?

Yes, we will do it by comparing the difference of $PM_{2.5}$ between the methods against that of NO_3^-.

P13,L9: How do you explain the large range of 20-100% here?

P13,L11: More information of the discrepancy between simulated and observed PM2.5 needed.

We will quantify the reason of this large range of discrepancy in the revised manuscript. The major difference between the bulk and the other non-equilibrium methods are nitrate with DU category. As nitrate is more mixed with anthropogenic or mineral dust (as $CaNO_3$ or NH_4NO_3), which is not allowed in the bulk method, pile-up $PM_{2.5}$ of bulk method should increase artificially compared to the other methods because NH_4NO_3 have to be mixed with SUB category and $CaNO_3$ cannot be produced.

P13,L13: provide R-values

We will move a part of Kajino et al. (2018a) to the supplement of the revised manuscript and give the *R* values in the main text.

P13,L19f: the term ‘air quality issue’ is too general

Thank you for your comment. As previously mentioned, we modified the separations of Sects. 3.1 and 3.2.

Chapter 3.2

P13,L21: How do you explain the large range of 20-100% here?

Please refer to the previous reply, regarding the difference of simulated $PM_{2.5}$.

Understanding of the differences in simulated PM2.5 needs for a more quantitative analysis as the reader does not have a clear picture if the presented values are realistic compared to observations.

Please refer to the previous reply, regarding the difference of simulated $PM_{2.5}$.

The link to air quality does not come out clearly in this chapter or is too general. In my opinion discussing AQ related questions, needs for inclusion of other pollutants as well such as the CO- and NO-family, Ozone and PM10.

Please refer to the previous reply, regarding the new sections 3.1 and 3.2.

Chapter 3.3, Chapter 3.4

For both chapters, the term 'climate relevant indices' is not very clear. Maybe it is better just to keep with 3.3 Aerosol optical depth and 3.4. Aerosol-cloud interactions.

Thank you. We will modify the title of the sections according to your comment. Because it was difficult for us to define one phrase to include the important variables for aerosol-cloud-radiation interactions, such as AOT, AAOT, CCN, and IN. Therefore, in the revised manuscript, we will use "climate-relevant variables" in the title, define it only twice in Abstract and Conclusion, and avoid usage elsewhere in the manuscript.

P14,L27: Specify why the bulk method is not suitable for climate modeling. The term 'climate modeling' however is misleading here as it is not subject to this study. Please clarify this sentence.

For climate modeling, aerosol size distribution is very important, but the bulk method does not resolve aerosol microphysics such as nucleation, condensation, and coagulation. We will rephrase it.

P14,L30: explain the large range 20-100%

It is due to the treatment of internal/external mixture assumptions of soot/soot-free and dust/sea-salt at the lateral boundaries of the model domain. Because it is very artificial and it is over the region where AOT is tiny, we will remove the colors over the region where AOT is lower than 0.1.

P14,L31: are you referring to the boundaries of the domain here?

Yes. Please refer to the above reply.

L15,L27: General assumption, or do you refer to any meteorological models in particular here?

No.

P16,L1-3: The conclusion of your findings are not clearly presented here.

We will modify the sentence.

Conclusion

The development and description of NHM-Chem is not sufficiently presented in this paper or not the main aspect as the study mainly focuses on the comparison of existing aerosol schemes. Further it should be indicated why the aerosol representation is a unique feature of NHM-Chem. In general, the analysis and the results are not detailed enough to show the models capability to serve operational forecast and air quality needs and observations are missing in order to substantiate your conclusion. Overall the paper lacks of a general conclusion, a discussion of the applicability of the results and the model's capabilities or recommendation for future modeling efforts in this field.

The revised manuscript is no longer a model description paper, but we will re-organize all the contents based on your comments as well as the other referee's, by including the model evaluations but avoiding duplications from the other paper, to be published in *JMSJ*.

The term "unique" is completely removed from the manuscript, as I admit that it was overstating.