

Interactive comment on “CCN activation and cloud processing in simplified sectional aerosol models with low size resolution” by H. Korhonen et al.

Anonymous Referee #3

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This paper examines the relative merits of different approaches that represent the activation of cloud condensation nuclei and their effects on cloud drop number, distribution of sulfate among the cloud drops, and surface area of the aerosol size distribution. The authors determine that aerosols represented with a few aerosol size bins adequately characterizes cloud drop activation especially when the shape of the distribution within a size bin is accounted for.

I) Comments on Philosophy of the Approach in the Context of Previous Reviewers' Comments

The study introduces alternative approaches to describing the aerosol size distribu-

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tion within a size bin. These approaches include a) assuming the size distribution is monodispersed within the bin, b) assuming the size distribution changes size linearly, and c) assuming a piece-wise linear distribution. In a way, these methods are similar to what is done in the multi-modal approach to describing aerosols, but this study focuses on more correctly describing the CCN (cloud condensation nuclei) distribution, the critical aerosol distribution for cloud drop activation and subsequent cloud properties. Thus, performing the same analysis on other aerosol modes is not as important to determining the effect on cloud properties. However, one may ask what the sensitivity of cloud properties is to non-linear descriptions of the aerosol size distribution within a size bin. It is easy to imagine applying a parabolic curve as the distribution of the aerosol size bin.

One of the motivations of this study is to reduce the uncertainties of climate forcings associated with aerosol-cloud interactions as predicted by large-scale climate models. One of the major difficulties in these large-scale models is parameterizing processes occurring on subgrid scales. Activation of CCN to form cloud drops is clearly one of these subgrid processes (even for clouds resolved by the model) and the parameters, e.g. vertical velocity, that influence the cloud drop formation process are also occurring at subgrid scales. For example, the large-scale model predicts the mean vertical velocity for a grid cell while in reality the grid cell contains updrafts and downdrafts. Thus, it must be recognized that there are several aspects contributing to the uncertainties associated with aerosol-cloud interactions. The one discussed in this paper is just one aspect and is applicable to not only global models, but also regional (1000s km), cloud (<500 km), and boundary layer (i.e. large-eddy simulations $\mathcal{O}(5)$ km) scale models. In addition to the questions brought forth by Reviewer 1 regarding the applicability of the methods presented in this study to large-scale models, it would also be nice to know how much do the alternative approaches reduce the uncertainties in aerosol-cloud interactions relative to other factors (e.g. subgrid-scale vertical velocities, organic aerosol effects; see e.g., Lance et al., 2004).

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II) Specific Comments

1. One of the interesting aspects of the paper is the results from multiple cloud cycles. However, I did not find any description of processes that occur (or not) when the air parcel is moved from cloud top to cloud base. Are the aerosol size distribution and its characteristics at the top of cloud from one cloud exposure used for the next cloud cycle? Or does the model simulate the air parcel in the downdraft so that physical and chemical processes continue to modify the aerosol? If it is the latter, there are many more questions regarding what occurs as the air parcel returns to cloud base that need to be answered. Clearly there needs to be more explanation on this aspect of the numerical experiment.

2. One assumption that the authors make is that the dominant reaction of aqueous SO_2 oxidation is by H_2O_2 because simulations of acidic cloud drops are performed. Indeed, previous studies have shown that the pH ranges across the cloud drop spectrum (e.g., Ogren, 1989) while others have shown that this pH spectrum across drops affects the production of sulfate (e.g., Roelofs, 1993). The smallest sized drops may have $\text{pH} > 5$ where oxidation by O_3 is important.

Furthermore, one of the experiments simulated is for urban conditions when SO_2 concentrations are high and H_2O_2 concentrations are low. It is not shown, but I would expect that the H_2O_2 is titrated and certainly oxidation of SO_2 by other oxidants should proceed.

However, because the paper is focused on the effect of the method and not on relative sulfate production, and because the results are compared to a detailed cloud scheme with the same reactions, ignoring other aqueous chemistry does not alter the conclusions of the paper. I do suggest that the authors emphasize that the results should be considered relative to the detailed model and that the magnitude of the results have some uncertainty because of the assumptions.

3. The simulations were performed for adiabatic conditions. That is, entrainment of air

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and turbulence within the air parcel were ignored. These processes tend to broaden the cloud drop distribution. How would the results of this paper change if entrainment and turbulence were included?

III) Minor Corrections:

1. Figure 4. Despite the effort to accentuate the simplified approaches from the reference simulation, it is difficult to see the lines for Approach 1, 2 and 5. This is because light colored lines and similar marker styles were used in addition to the markers being at the same location for each line. I would suggest changing the reference line to a wider, gray line and using solid lines with markers for the simplified approaches.

2. Figure 5 is too small. It may be because of the way the journal puts the figure into the article, but something needs to change.

3. Table 2 caption. "Percentual error" should be "Percent error".

4. Page 4881, first full paragraph that has two "On the other hand" phrases. Remove the first "On the other hand".

Line 9, "values is a prescribe" should be "value is a prescribed"

Line 19, "with the moving" should be "as the moving"

Line 26, "In real atmosphere" should be "In the real atmosphere"

5. Page 4882,

Line 8, "principal" should be "principle"

Line 11, "from accurate solution" should be "from an accurate solution"

Line 15, "diagnoses the properties" should be "diagnoses of the properties"

Line 23, "In polluted" should be "In the polluted"

6. I suggest proofreading the text again to be sure other pages do not include similar

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grammatical errors.

References

Lance, S., A. Nenes, T. Rissman (2004) Chemical and dynamical effects on cloud droplet number: Implications for estimates of the aerosol indirect effect, *J. Geophys. Res.*, **109**, D22208, doi:10.1029/2004JD004596.

Ogren, J. (1989) Measurements of the size dependence of solute concentrations in cloud droplets, *Tellus*, **41B**, 24-31.

Roelofs, G. J. H., (1993) A cloud chemistry sensitivity study and comparison of explicit and bulk cloud model performance, *Atmos. Environ.*, **27A**, 2255-2264.

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