Interactive comment on "Aerosol–cloud interactions studied with the chemistry-climate model EMAC"

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Response to reviewer 3

We thank the reviewer for the constructive and valuable comments, and will revise and improve the manuscript according to your comments.

In response to the comments:

The study by D. Y. Chang et al. attempts to compare two cloud droplet nucleation parameterizations, which differ in their representation of the activity of water in solution in the chemistry-climate model EMAC and a reference model setup without a coupling between aerosol and clouds. The outcome of such an activity would be interesting and welcome but due to several shortcomings such as the missing tuning of model parameters for the different model setups, the study does not present results that would allow a sound comparison. I cannot recommend the manuscript to be published in the present form.

General Comments

1. Model parameters need to be tuned when the model is changed so that the shortwave, longwave and net fluxes, shortwave and longwave cloud radiative effects, cloud cover, liquid water path etc. are within observed ranges. Only then can model results that were produced with different parameterizations be compared to observations. Some simulations in the paper show unrealistic values for global annual mean cloud properties. Large parts of the analysis and conclusions in the paper are not meaningful because of this lack of model tuning. Only after tuning would the attempted assessment of the performance of the different simulations be possible.

→ We agree with your argument that the model has to be tuned to make the assessment of model performance against observations meaningful. Model parameters can be tuned to improve agreements between model results and observations.

But tuning parameters for each model setup is another influencing factor that changes cloud properties and cloud radiative effects. This is not ideal for identifying aerosol physicochemical effects on cloud droplet nucleation and their propagated effects on clouds and climate. We will rewrite section 3.4 with further analyses of aerosol and cloud properties, and aerosol physicochemical effects on cloud droplet nucleation processes and refocus on the explanation how the different critical supersaturation algorithm in the ARG cloud droplet nucleation scheme can propagate into large differences in cloud properties.

2. There are indications that there is an implementation problem in the STN-simulations (cloud droplet nucleation parameterization which uses the osmotic coefficient to describe the activity of water in aqueous solution). There is a difference of approximately a factor 2 between the activated aerosol fraction and CCN between the STN and HYB simulations. This difference is the same for aitken, accumuluation and coarse mode, i.e. independent of size although the solute effect should be more important for smaller particles, i.e. the difference should decrease with particle size. The activated aerosol fraction in the lowest model levels is close to 1 for aitken, accumuluation and coarse mode, CDNC concentrations in the lowest model levels are very high and the global, annual mean CDNC burden is very high.

→ We concur about insufficient explanations for large differences that could also point to implementation problems. We believe that the cloud droplet scheme is well implemented in the model, however, we will carefully check the implementation with corresponding assumptions line by line. The volume-weighted global mean values from the surface to the upper troposphere could also be misread. The solute effect is more important to droplet formation for smaller particles than for larger particles. Note that the activated aerosol fraction is a relative value. The product of the activated aerosol fraction and the available aerosol number concentration determines the nucleated cloud droplet number concentration for each aerosol size mode. Therefore the vertical and horizontal aerosol distribution strongly influences CCN number concentration and thereby CDNC. As shown by the vertical distributions of the activated aerosol fraction (Fig. 5), the largest differences are found in the lowest model level. For accumulation

and coarse modes the vertical distribution of the available aerosol number concentration decreases exponentially with altitude, while for the Aitken mode maximum number concentration are found at relatively high altitudes (about 10km) for both land and ocean and the near the surface over land. These differences in vertical aerosol distribution also influence the total activated aerosol number concentration. We will rephrase the text regarding Table 6 with more description and discussion including how CCN and CDNC are calculated in the model, with the vertical distributions of aerosols separately for land and ocean.

3. The unrealistic cloud properties in the STN-simulations are mentioned in the paper but no sufficient explanation is provided.

→We will try to provide further explanations for why the STN scheme simulates the rather different cloud properties.

4. Section 2 should be extended to describe also the methods used in (e.g. Taylor diagram, skill score) and the observational data used. This information is now in different parts of the paper and not enough details for the observational data are provided

→ We apologize for any inconvenience or confusion caused by the arrangement of the information and not enough details for the observational data. We provided the descriptions of Taylor diagram and skill score in the supplement, and of the observational data in Table 5. As requested, we will provide more details for the observational data in the revised manuscript. Some parts in Section 3.4 about general assessments will be removed, including Taylor diagram and skill score, in the revised manuscript, since the purpose of the study will be more focused on understanding the changes in cloud properties.

Specific comments

Thank you very much for your specific comments. We will revise the manuscript based on your comments.

P21983, L9: Unclear. In M7 the modes are constrained by reallocating aerosol particles that exceed the upper size limit of a mode to the next larger mode.

→ We will rephrase this sentence in the revised manuscript. The current model used the Global Modal-aerosol eXtension aerosol module (GMXe; Pringle et al., 2010a) not M7. In GMXe, the modes are able to reallocate aerosol particles that exceed the upper size limit of a mode to the next larger mode. We will explain this in more detail.

P21999, L21: Why are the STN-simulations so insensitive to the aerosol composition?

→Your question refers to the discussion of Fig.5, the relative difference of the activated aerosol fractions over land and ocean. Our phrasing might have been misleading and will be changed and rephrased. Fig.5 clearly shows that the sensitivity in aerosol activation of STN and HYB above the PBL in the free troposphere is comparable. In the PBL HYB simulates higher activities in the marine environment. We will provide more details on the