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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric aerosol size distributions simulated by three online global aerosol models using the M7 microphysics module" *by* K. Zhang et al.

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Reply to referee 2

We thank the reviewer for his/her helpful comments and suggestions. Our reply to the review is detailed below.

The manuscript presents an inter-comparison of three global aerosol models, which include that same aerosol microphysical module. This manuscript is of interest to researchers using these global aerosol models presented in this paper, since it identifies

the differences in the atmospheric aerosol size distributions resulting from treatment of aerosol processes and transport. Nevertheless, the problem with the manuscript is that it does not identify the reasons for the differences between the model results. I suggest publishing the manuscript after more effort is put into identifying the reasons for the inter-model discrepancies.

In order to provide explanation for the inter-model discrepancies, we performed various sensitivity experiments and presented the results in the manuscript. For example in Section 2 (p5819 - 5820), four simulations were performed to investigate the reason for the considerably high H_2SO_4 production in ECHAM5-HAM. From these simulations, turbulent dry deposition, DMS emission, oxidant concentration, and the possible nonlinear interaction among these factors were ruled out, and the gas- and aqueous-phase chemistry were identified as the main reason. As another example, Fig. 2 in the manuscript showed two sensitivity simulations which revealed that the condensation of H_2SO_4 was the dominant reason for different upper troposphere number concentrations of the nucleation and soluble Aitken mode in CAM3-LIAM and ECHAM5-HAM. In addition, in the revised manuscript we follow the reviewer's suggestion and demonstrate that the sea salt emission parameterization is the main reason for inter-model discrepancies in the low-level concentrations of the soluble accumulation mode and soluble coarse mode.

It is true that there are quite a few differences between the model results which we are not yet able to explain with confidence. Considering the high complexity of the aerosol models, an exhaustive attribution of inter-model discrepancy is difficult to accomplish in a single publication. This manuscript presents our first attempt in this direction, and we are fully aware that future work is needed. Nevertheless we think our analysis provides useful information to the aerosol modeling community.

Specific comments:

One reason for the discrepancies between the model results and observations have been stated to be "probably caused by the fixed mode variance in the mathematical formulations used in the modal approach in the three models". Would it be possible to make simulations using different mode widths. How sensitive are the results to the mode width, if for example coarse mode width is changed from 2.0 to 1.8 (which is said in Summary to be the observed width in China adjacent seas)?

Following the reviewer's suggestion, we added two further sensitivity tests in which the width of the coarse mode is set to 1.8 and 2.5, respectively. Simulations over the China adjacent seas does not show clear improvement. Our initial speculation about the model variance's role turns out to be incorrect. The corresponding text in the manuscript has been revised.

As for the other modes, the number concentrations do not show evident change in these two experiments. This is understandable because coarse mode aerosols have very low number concentration, and their impact on smaller particles is in general small.

Another main reason for discrepancies is said to be differences in emission parameterizations. The sea salt emission parameterizations appear to be very similar in the models. Nevertheless, in Section 6.2, the reason for the underestimation of accumulation model particle number concentrations in ECHAM5-HAM is suggested to be caused by the sea salt emission parameterizations. This should be fairly easy to verify by feeding identical inputs typical to the remote oceans to the emission modules and comparing the number fluxes obtained from the parameterizations. Have the 10 meter wind speeds between the different models been compared? Could this explain the discrepancy?

According to the reviewer's comment, we have carried out additional analysis and sensitivity experiment to quantify the impact of the two factors – emission parameterization and 10 meter wind – separately.

In Fig. R1 (please see the supplement) the annual mean 10m wind simulated by the three GCMs is compared with the 1989-2009 multi-year mean from the ERA-Interim reanalysis. All else being equal, higher 10m wind speed will result in stronger sea salt emission. According to Fig. R1 in the supplement, ECHAM5 produces significantly stronger winds than GAMIL almost all over the globe, and stronger winds than CAM3 in many regions, yet the emission in the accumulation mode is much weaker in ECHAM5-HAM (Fig. R2 in the supplement). Thus the 10m wind can not explain the inter-model discrepancy in the simulated number concentration.

An additional simulation has been performed using the ECHAM5-HAM model, with the sea salt emission scheme replaced by the one from LIAM. Since aerosols have no feedback to the host GCM in the simulation, in the new simulation the LIAM emission scheme receives exactly the same input (i.e. 10m wind) as in the original ECHAM5-HAM model. Results from this additional run are shown in Fig. R3 in the supplement, where we see an evident increase in the number flux of the accumulation mode and decrease in the coarse mode, leading to similar changes in the simulated number concentrations. These results support our attribution that differences in emission parameterization makes significant contribution to the inter-model discrepancies. The new results have been added to the manuscript.

In section 4.4, it is said that diagnostics for aging of the insoluble particles are not available. Wouldn't it be fairly straight forward to calculate the rate of insoluble particles converted from insoluble to soluble modes?

The conversion of insoluble particle to soluble aerosol is caused by (1) condensation of H_2SO_4 on insoluble particles, and (2) coagulation of insoluble particles with soluble particles. Indeed one can diagnose these sinks in the model, but technically they are not available in the current version of these models. This is why we did not perform detailed comparison on aerosol aging in this study. Our original statement was unclear, and has been removed.

From this manuscript it is a bit difficult to distinguish which discrepancies are caused by model meteorology and which are caused by aerosol processes. Comparing the radiative properties of aerosols to e.g. satellite data would shed some light on how much wind driven online emissions (dust and sea salt) differ for different models.

In this manuscript two out of the three models – GAMIL-LIAM and CAM3-LIAM – share the same aerosol module. Their differences in aerosol simulation are caused solely by model meteorology.

The discrepancies between ECHAM5-HAM and the other two models are indeed caused by meteorology and aerosol processes combined. Therefore in the manuscript we have used some "surgery experiments" to isolate different processes. For example, in Section 3 the dry deposition of SO_2 in ECHAM5-HAM is replaced by the scheme from LIAM to investigate the cause of high H_2SO_4 production. In the revised manuscript, similar experiment is performed which quantifies the impact of parameterization scheme on sea salt emission and consequently on aerosol number concentration.

Note that the focus of this manuscript is on aerosol size distribution. The aerosol radiative properties provided by satellites represent the collective effect of particles of different sizes, and do not provide direct information about each individual size range. This is the main reason why satellite data are not used in this manuscript.

Technical comment: - In Table 3, nucleation, condensation, and coagulation overlap.

This problem was caused by the discussion paper form and will be corrected in the final version.