Atmos. Chem. Phys. Discuss., 6, S795–S799, 2006
www.atmos-chem-phys.org/acpd/6/S795/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol activation and cloud processing in the global aerosol-climate model ECHAM5-HAM" *by* G. J. Roelofs et al.

G. J. Roelofs et al.

Received and published: 9 May 2006

Author comments for acpd-2005-0369

"Aerosol activation and cloud processing in the global aerosol-climate model ECHAM5-HAM" by G.J. Roelofs, P. Stier, J. Feichter, E. Vignati and J. Wilson

We thank the reviewers for their constructive comments and suggestions. In the following we will address their questions and indicate which changes have been made in the manuscript

Referee 1

1) The referee mentions that "the CDNC due to activation should be different from the CDNC in aged clouds due to self conversion of cloud droplets. Hence the CDNC sim-

ulated by the parameterization should not necessarily match the observations". Due to autoconversion in clouds the number concentration of cloud drops decreases as precipitation drops are formed. Therefore, if collision/coalescence significantly influence the average CDNC then our simulated CDNC can not be compared directly with observed values. However, generally only one cloud in ten actually produces precipitation so that it can be assumed that on average the observed reff is only for a small part affected by autoconversion. Another issue is that it is not clear at what time during the cloud evolution the measurement is made and whether autoconversion or other processes as turbulence and entrainment have affected CDNC significantly. We therefore are confident that the comparison between simulated and observed reff in our study is a good semi-quantitative evaluation of CDNC. As the referee mentions, the simulated CDNC is already smaller than observed, and this discrepancy can not be explained by autoconversion.

2) The referee asks for more information on the assumptions made in the model, why they are necessary and how they affect the results.

i) Ammonium bisulfate: Because a representation of ammonium chemistry is not included in the model we assumed that sulfate is present as ammonium bisulfate. Other options would be sulfuric acid or ammonium sulfate. All three species are well soluble so that the solute term of the Koehler equation for this aerosol does not change much, and the activation characteristics of the aerosol are comparable. However, the different levels of acidity for the three species will have an effect on the distribution of in-cloud produced sulfate over the activated modes thereby indirectly affecting the aerosol size and lifetime, similarly to sensitivity test B1 where HNO3 was decreased.

ii) Feedback of CDNC on precipitation formation: The CDNC is not fed back to the radiation code and the precipitation formation parameterization of the GCM in order to obtain the same meteorology (transport, temperature, cloud occurrence, precipitation patterns) in the sensitivity studies. This enables to explain the differences between the sensitivity studies directly in terms of the input aerosol and cloud physical and chemical

ACPD

6, S795–S799, 2006

parameters.

iii) In-cloud scavenging of interstitial particles: In-cloud scavenging of interstitial particles moves number and mass from the small unactivated modes to the accumulation mode. It therefore has the same effect on the aerosol size distribution as impaction between aerosols. The module HAM considers this collisional growth of unactivated particles. It is likely that a compensating effect occurs. The negligence of in-cloud scavenging of interstitial particles in our model may lead to a stronger uptake of nucleation and aitken mode particles by the accumulation mode through collisional growth. It may be expected that some effect on CDNC will occur, so it is a point that certainly merits more investigation.

iv) HNO3 and activation: The general effect of dissolution of HNO3 in aerosol is to enhance its activation potential. HNO3 concentrations are relatively large in polluted areas. Our previous work indicates that the increase in CDNC is of the order of a 5-10 percent at low updraft velocities but the effect decreases or reverses even for updraft velocities above 50 cm/s (see Roelofs and Jongen, 2005). We conclude that the impact of the HNO3 on aerosol activation is therefore far less than that of, for example, errors in updraft velocity or missing aerosol (precursor) emissions.

vi) Accuracy of simulated LWC: Evaporation and precipitation amounts agree relatively well with observations except in tropical regions where the hydrological cycle appears too strong (Hagemann et al., 2006). Our study therefore strongly underestimates aerosol particle concentrations in that area. For the extra-tropics it can be assumed that the simulated strength of the hydrological cycle is more realistic. Nevertheless, discrepancies between modelled and observed SW and LW cloud forcings suggest that the amount of low clouds may be overestimated in the model (Wild and Roeckner, Radiative fluxes in the ECHAM5 general circulation model, J. Climate, in press, 2006). It is not clear whether this is caused by liquid or ice clouds, or due to an overestimated cloud cover, cloud LWC or too long cloud lifetimes.

ACPD

6, S795–S799, 2006

3) In-cloud chemistry parameterization: We have used detailed cloud parcel model simulations to calculate in-cloud produced sulfate amounts for different cloud types and pollution levels. We have compared these values with calculations with a simple cloud representation which considers only two drop classes with different acidity levels. Generally, an approximate 75%-25% distribution of the liquid water associated with both classes resulted in in-cloud produced sulfate masses within 5% of the detailed model. The optimal distribution varied only several percent around this 75%-25% regardless of vertical velocity or aerosol type. Therefore this distribution was applied in the model.

4) Effect of precipitation formation on sulfate production: Although in individual cloud events the impact may indeed be significant, on the spatial and temporal scales considered here the effect of precipitation formation on in-cloud sulfate formation is relatively insignificant with respect to the simulated sulfate yield and distribution. This is due to the fact that the SO2 lifetime is much shorter than the averaging time of three months used in our study. On the other hand, it may affect the distribution of in-cloud produced sulfate over the different modes, so that CDNC may change somewhat.

All minor comments of referee 1 have been dealt with.

Referee 2

The model predicts masses of sulfate, sea salt and organic matter as soluble aerosol species. The referee mentions that for activation calculations the molar quantities are needed, and not the masses which are provided by HAM. In our study we proceed as follows: From the lognormal parameters predicted by the model for each aerosol mode we derive a size distribution over which the associated aerosol masses are distributed. Using the appropriate molar mass, the masses can be converted to molar quantities that are used for the Raoult factor in the Koehler equation. A problem here form the organics which in reality are a mixture of many different species with different molar masses and solubility. We have arbitrarily assumed that the molar mass is represented by that of oxalic acid. We have described in more detail how we proceed to calculate

ACPD

6, S795–S799, 2006

the Raoult term in the text (section 2) and in the Appendix.

The referee mentions further that unknown chemical composition and chemical processes inside particles make it difficult to realistically calculate particle activation. We note that the model in this version assumes an “ideal set” of aerosol species: all sulfate is ammonium bisulfate, all sea salt is NaCl, organic matter is composed of a single species with fixed solubility. Based on these idealized aerosol species the activation calculations are performed. In the discussion we argue that the impact of these assumptions on calculated CDNC is much smaller than, for example, that caused by inaccuracies in emission rates.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 519, 2006.