

***Interactive comment on “Sensitivity of aerosol concentrations and cloud properties to nucleation and secondary organic distribution in ECHAM5-HAM global circulation model” by R. Makkonen et al.***

**Anonymous Referee #1**

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The paper describes the results of the introduction of additional aerosol processes on aerosol and cloud droplet number concentrations. It nicely shows the consequences of the changes of the original ECHAM-HAM parameterization. Such sensitivity studies are useful and deserve publishing - but should be a little bit better documented, so that other scientists can repeat the experiment with a different model eventually and understand the tests.

Here are my specific comments:

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Are the mass emissions equal in all simulations? I guess so, however

How do the mass budgets change by introducing an additional nucleation mechanism?  
Is there less SO<sub>2</sub> entering the free troposphere due to the additional nucleation mechanism?

Did the aerosols and CDNC concentrations change the meteorology in between experiments? How much has the sulphur life time changed in between the experiments?

Section 2.2: How many of the particles greater than 0.035  $\mu\text{m}$  radius are considered CDNC within the clouds? How do you get to grand averages comparable to eg MODIS? Do you compute these concentrations just in the cloudy fraction? (I hope so)  
In Lohmann et al 2007 it was mentioned that a minimum CDNC concentration of 40  $\text{cm}^{-3}$  is set. How can you have then CDNC concentrations almost near 40 and even lower than 40 as reported in your table 3. Lin and Leach are not in the reference list.

Section 2.3: This section should be reordered and generally revised to better explain the different nucleation schemes. What exactly happens in the original ECHAM nucleation mechanism? Why are there points 1-5 listed in the last part of the section? Do these refer to any experiment? It is written 'we evaluated the sensitivity of the model to the choice of the implementation'; I do not really find the corresponding chapter. The parameters  $\chi$  and  $\gamma$  are not well explained, while others are nicely explained. Is the binary nucleation mechanism of the original ECHAM model changed at all? If the time step of the model is 30 minutes, how does this impact the evolution of the size distribution for a process as fast as nucleation? Please make this chapter clearer and let it correspond to the experiments actually undertaken.

Is it realistic to just add the activation-type nucleation process? Shouldn't there be competition for the available vapour among the processes to form new particles? How do the H<sub>2</sub>SO<sub>4</sub> concentrations differ across experiments? Did the parameterization of the original ECHAM model implicitly include also activation-type nucleation, or is strictly referring only to binary nucleation?

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What is the role of the condensational sink parameterisation on the nucleation rates?

Section 3:

upper troposphere and lower stratosphere; which is the only region where binary nucleation rates in average exceed activation type nucleation; => In the model or in the real world? Citation?

the difference between B and A simulations comes from the fact that Experiments with act type nucleation use Eq 1 to scale nucleation rate to  $J_{3nm}$ ; => I cant follow the argument, can you please explain better.

since only a fraction of nucleation mode particles grow to larger sizes; since average nucleation mode radius is already closer to Aitken mode radius; This also demonstrates that nucleation mode particles are effectively growing into the Aitken mode; => What is the real mechanism in the model which let particles grow into the Aitken mode? Can you quantify the role of condensation, coagulation and purely numerical diffusion between the modes? Why are activation type nucleated particles and binary nucleated particles different in size? Please explain better. What is the average median size radius of the nucleation mode? It should be easy to give these numbers for the different experiments.

Fig 2a shows that binary nucleation produces large numbers in low temperature and aerosol surface area regions and where the relative humidity is high; I don't think that Fig2a shows this. Is the relative humidity really high; in the upper troposphere, respectively in those regions where nucleation is largest?

In the upper troposphere activation type nucleation decreases nucleation mode particle concentrations; => Do you mean adding a nucleation process suppresses nucleation mode particle concentrations? Do you mean that the same mass of  $H_2SO_4$  forming new particles is formed on fewer, but larger particles? Are the

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H<sub>2</sub>SO<sub>4</sub> formation rates from gas phase oxidation the same in experiment B and A1 ?

Section 3.2:

As expected, the sensitivity of the concentration to A is lower than to the particle formation rate;=> First, why? Second, the scales in figures 3 and 4 are not equivalent. In one case you span 5 orders of magnitude (figure 4), in the second 3 orders of magnitude (figure3). No wonder that the figure 3 looks more colourful.

Section 4:

The comparison to the figures from Bennartz is difficult to follow. Please include a figure with the data from Bennartz, plotted with the same routine. Looking at that figure: There are some significant differences between model and MODIS. It would be nice to have these commented instead of stating that there is a reasonably good agreement;. There is a slight confusion in figure subtitle and text with respect to the period in the original Bennartz paper. July 04- Dec05 are not two and a half year in my counting. Is it Fig 5 or Fig 7 in the Bennartz paper?

the resulting CCN numbers; => do you mean CDNC ?

Section 5:

The comparison to the observations show that activation nucleation is a very promising way to improve the ECHAM model;. The direction is clearly towards a better representation of aerosol number ; => If you would have increased emissions of SO<sub>2</sub> or photo-oxidants, or reduced wet and dry removal of SO<sub>2</sub>, you would also have produced a similar increase in aerosol number concentration. Some more modest conclusion would do the paper good. The observations used are not really suited and not very specific for the problem of judging the nucleation mechanism.

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 10955, 2008.

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