

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.

R. Makkonen et al.

Received and published: 14 October 2008

We thank the referee for an extensive list of comments and suggestions, and we will answer here the questions raised. Please see also author’s comment “general comment”.

- 1) Mass emissions are kept equal between experiments. However, BSOA yield determines the organic mass partitioned to aerosol phase.
- 2) Merely changing the nucleation mechanism doesn’t affect the mass budgets significantly. The burden of SO₂ changes only by few percents between experiments. Hence

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the mass of SO₂ entering the free troposphere is not significantly altered.

3) Aerosols and CDNC are coupled to the climate model and they affect the meteorology. However, changes are quite small in the timescale of the experiments (one year), and therefore the changes in sulphur lifetime are rather insignificant.

4) CDNC values in the discussion paper were calculated over both cloudy and non-cloudy grid boxes. In revised version of the manuscript, averages are calculated only over cloudy fraction. This explains the low values (below 40 cm⁻³) in original figures and tables. Missing reference was added.

5) This section was slightly rewritten to clarify the points 1-5, and parameters in equation 2 are now explained better. Experiment B is original ECHAM5-HAM, where binary nucleation is not changed. In experiments with activation-type nucleation, binary nucleation rate is converted to formation rate of 3 nm particles.

6) We tried to approach this problem by using different ways to implement activation-type nucleation in ECHAM5-HAM, and came into the conclusion that for this sensitivity study, it is enough to implement activation-type nucleation as explained in Section 2.3. Original ECHAM5-HAM includes only parameterization for binary sulphuric acid-water nucleation.

7) In our parameterizations condensational sink is used in Kerminen-Kulmala equation with growth rate to estimate initial growth of aerosol. This way we can evaluate formation rates of 3 nm particles instead of actual nucleation rates.

8) The statement applies for model calculations. The text was revised.

9) Experiment B calculates the nucleation rate of critical clusters, which are usually less than 3 nm in diameter. Therefore, average radius of nucleation mode is lower in experiment B compared to that of experiments A0-A2.

10) Details on aerosol model dynamics are given in Vignati et al. (2004). In our setup, nucleated particles grow from nucleation mode to Aitken mode by condensation of

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sulphuric acid. In experiments with hybrid BSOA also biogenic organics can help this growth. While the yearly global average radius of nucleation mode in experiment A1 is about 2 nm above 400 hPa, in experiment B the average radius reaches values below 1 nm.

11) This sentence was revised to be clearer on what Figure 2a actually shows.

12) This issue is merely technical. We wanted to keep our base case (experiment B) identical to original ECHAM5-HAM. In simulations where activation nucleation was modeled, we calculated the formation rates of 3 nm particles, instead of actual nucleation rates (as in experiment B). For this reason, nucleation mode particle numbers between experiment B and others are not directly comparable, since their average radius is different. The text was revised to use more correct terms.

13) Actually, the span in Fig. 3 is 6 orders of magnitude and 5 orders of magnitude in Fig. 4. The particle formation rate is proportional to the coefficient A, whereas particle concentrations depend on various other parameters. The color scale in Fig. 3 is now changed.

14) We have now included satellite data in the paper to make comparison easier, and figure text was rewritten. Text was modified to include also discussions about differences between model and observation.

15) CDNC is correct, not CCN. The sentence was fixed.

16) We have partly rephrased the discussions and conclusions sections. New particle formation is certainly not the only error source with current global aerosol models, and when other processes and emissions are improved, the role of aerosol nucleation will change from what is stated in the paper. However, we already have evidence that the binary sulphuric acid-water nucleation is not able to explain the observed nucleation events in the boundary layer; hence we need to include an additional particle source. We don't have detailed information on the process itself, but we can still evaluate

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the climate sensitivity by varying related parameters.

References:

Vignati, E., Wilson, J., and Stier, P.: M7: An efficient size-resolved aerosol microphysics module for large-scale aerosol transport models, *J. Geophys. Res.*, 109, D22202, doi:10.1029/2003JD004485, 2004.

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

ACPD

8, S8221–S8224, 2008

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