

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

Anonymous Referee #2

Received and published: 8 March 2006

The work presented by Roelofs et al., presents a parameterization for cloud processing that calculates activation of aerosol particles to cloud drops, cloud drop size, and pH-dependent aqueous phase sulfur chemistry. The parameterization is implemented in a global aerosol-climate model, which feeds the parameterization with the required parameters such as updraft speed, temperature, aerosol size and “chemical parameters”.

Although the simulated CDNC is found to be relatively sensitive to the uncertainties in parameters such as updraft velocity, or the assumptions on the organic solubility, the work presented by Roelofs et al., is new and deserves publication in ACP, if the discussion has been extended with respect to the specific comment given below.

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Specific comment:

The factor B_i (Appendix) that is needed for the Raoult term in the Köhler equation has been derived from the simulated masses of sulfate, organics and sea salt contained in mode i. However, what is needed for the Raoult term is the number of total moles. So the caveat is that without an aerosol composition module that explicitly resolves the estimation of number of moles associated with the distribution of organics, sea salt, etc. is NOT possible - HAM simply does NOT provide this. For instance, how many moles do you assume for sea salt, if you do not know the chemical composition nor the physical state of the aerosol?

Let's simply consider sea salt that is composed out of various and more or less hygroscopic salts, including e.g. NaCl and MgCl₂. Depending on temperature (T) and relative humidity (RH), alone these two salts would provide either 2 moles of 1 mole dissolved NaCl if the RH exceeds approx. 70% (the so-called relative humidity of deliquescence, RHD), or 3 moles of 1 mole dissolved MgCl₂ if the RH exceeds approx. 32% RH (both at 298 K). However, in case sea salt becomes coated/mixed with acids such as sulfuric, nitric or organic acids (e.g. if sea salt particles are affected by coastal air pollution) than the chlorine molecules would be substituted by these acids since - in thermodynamic and chemical equilibrium - they would preferentially and at least partly be neutralized by cations such as Na⁺ or Mg⁺⁺. This would not only affect the number of moles that eventually determine the Raoult term - MgSO₄ would only contribute 2 moles instead of 3 moles of MgCl₂ in case of complete dissociation - but probably even more important is the fact that due to the changed aerosol composition, the physical state of the sea salt particles might be different at the same T and RH; the RHD of MgSO₄ is approx. 86% (instead of 32%). Such shifts in the RHDs are probably most important, as the RHDs determine the aqueous phase, and together with the number of moles of dissolved compounds, the hygroscopic growth and activation of the aerosol particles (besides effects on heterogeneous chemistry due to changes in the surface reactivity). But the RHD - as the number of moles in the solution - depends strongly on

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the aerosol composition. E.g. observations show that aged aerosol particles containing organics do either clearly show a deliquescence behavior or not. i.e. such aged particles might have a lower or higher RHD than their inorganic counterpart.

Hence the picture outlined above becomes much more complicated in case the actual aerosol composition of at least major inorganic and organic aerosol compounds is considered - a pre-requisite for a realistic aerosol/cloud coupling/modelling, however.

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

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