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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol nucleation and its role for clouds and Earth's radiative forcing in the aerosol-climate model ECHAM5-HAM" *by* J. Kazil et al.

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We would like to thank the reviewer for the helpful comments which have lead to substantial improvements in the manuscript.

1. I've been quite uncomfortable with modal aerosol schemes with 4 size modes (representing 4 orders of magnitude in diameter) being used to address the impact of nucleation on CCN in 3D models. I don't recall seeing M7 tested for this purpose (I may have missed it). Given that nucleation events can vary greatly in nucleation rate, duration and growth rates, it seems unlikely that modal schemes with 2 modes between 1 and 100 nm could adequately predict the growth all events. The large sensitivity of

cloud droplet burden to maximum geometric mean diameter of the nucleation mode confirms this.
If there has been published work on the use of modal schemes for nucleation and growth, please cite them. If not, I recommend that someone from the modal community do this (maybe the Helsinki group would be the best to do this since they are using ECHAM5-HAM with an emphasis on nucleation/growth), though this definitely doesn't need to be done for this paper.
In response to what you are probably thinking, I'm not trying to be an annoying sectional modeler making the lives of modal modelers harder (I used the Kerminen and Kulmala approximation of the sub-10 nm nucleation mode in the sectional scheme until recently, a similar computational-time saving assumption). This is just something that should be quantified at some point given the sensitivity of CDB to max median diameter shown in the paper.

Modal aerosol schemes have drawbacks, which include, as the reviewer correctly points out, a limited ability to resolve the aerosol size distribution. Nucleation events in short succession, e.g., may result in more peaks in the aerosol size distribution than there are modes in the modal scheme. However, state-of-the-art sectional schemes in climate models are not highly resolved, either (8-10 bins), and should not be a priori expected to perform better than the modal scheme employed here (see Kokkola et al., Geosci. Model Dev., 2, 97-112, 2009). Only chemical transport models have comparably high resolutions (30-60 bins), but are limited in their use for climate research. Sectional schemes suffer from having a lower size cutoff, meaning that a gap needs to be bridged between the size (mass) of the particles that a nucleation scheme provides and the smallest particle size (mass) in the aerosol scheme, as mentioned by the reviewer. Modal schemes have no lower size cutoff and can therefore accommodate nucleating particles of different size (mass) from different nucleation schemes in a consistent way, a handy feature that allows using different nucleation schemes concurrently, as is done in this work. In addition, modal schemes are computationally far less expensive than more highly resolved sectional schemes, and the benefits of exe-

cution speed may outweigh a high resolution of the aerosol size distribution in climate simulations (decades to centuries), in particular in climate runs with coupled aerosol, cloud, surface, and ocean processes.

We have added the reference to Kokkola et al. (Geosci. Model Dev., 2009, doi:10.5194/gmdd-2-209-2009) in the revised manuscript, who compare M7 with several sectional models, although for stratospheric conditions. Tests of the M7 module against a sectional model in tropospheric conditions have been conducted by Vignati et al. (JGR, 2004, doi:10.1029/2003JD004485).

2. (See also my specific comment about Figures 9a and 7c) A very related point, but dealing with cloud activation is modal schemes. It seems like the "Hoppel gap" (the minimum in the size distribution between the Aitken and accumulation modes caused by cloud processing of aerosols) in modal schemes may be shaped greatly by the numerics of the modal scheme as much as actually due to cloud processing. This could have an effect on cloud droplet number concentrations since the cloud scheme could be repeatedly activating a large fraction of the Aitken mode, but the Hoppel gap might not shift to lower sizes accordingly (perhaps it does shift correctly). I could be completely incorrect with this concern, but I was curious to if its been looked at. As with the previous point, I don't think anything needs to be added to the paper (unless its as simple as citing some previous work that explored these). Just something to think about in the future.

The Hoppel gap may not be located at the correct particle size in modal aerosol schemes, as it depends, among other things, on the (fixed) mode width (geometric standard deviation). However, the Hoppel gap may not appear at the proper size in sectional aerosol schemes either, e.g. when activation of aerosol particles is determined based on a fixed size cutoff (e.g. Spracklen et al., ACP 2005, doi:10.5194/acp-5-2227-2005). In ECHAM5-HAM, the number of particles that are activated from the different aerosol modes does not depend on the location of the Hoppel gap: If a particle is sufficiently large to be activated, it does not matter whether it is in the Aitken or

the accumulation mode. Independently of that, the Hoppel gap does move to a smaller size in the model when activated Aitken mode particles are placed in the accumulation mode.

p. 12267, l14: Single kappa parameters (per mixture) begin to have errors for water uptake when RH becomes low. Is this a problem?

We justify the use of the κ-Köhler theory to calculate aerosol water content in ECHAM5-HAM by a significant improvement in the simulated aerosol optical depth in comparison with observations over previous versions of the model, which, however, is not discussed in the present manuscript. Efforts are underway to investigate and better specify hygroscopic growth and its role for AOD in ECHAM5-HAM (D. O'Donnell).

We are not using a fixed κ for all RH. The basic equation of the κ-Köhler theory set out in Petters and Kreidenweis (ACP, 2007, doi:10.5194/acp-8-6273-2008) is equation (11) in that paper:

$$RH \cdot exp\left(\frac{-A}{D_d f}\right) = \frac{f^3 - 1}{f^3 - (1 - \kappa)} \quad , \tag{1}$$

where D_d dry particle diameter, f the growth factor, and A the Kelvin curvature term

$$A = \frac{4\sigma M_w}{RT\rho} \quad , \tag{2}$$

σ being the surface tension of water and ρ its density. This equation is solved for the growth factor f offline - to avoid iterative solving online - for atmospherically relevant RH, T, D_d and κ; the results are stored in a lookup table. We calculate the κ of the mixture at each grid point and timestep as per equation (7) in Petters and Kreidenweis (2007) and then look up f.

However, even if a fixed κ was used at all RH, the errors would be limited: Kreidenweis et al. (Env. Res. Lett., 2008, doi:10.1088/1748-9326/3/3/035002) find that using a fixed κ leads to water contents that are within experimental errors over the full range

of RH, except for marine-type aerosol, for which the parametrization under-predicts water content for RH $<$ 90%. However, at locations where marine-type aerosol are most abundant (near the ocean surface), RH is $>$ 70% in the annual average of our simulations, which would limit the errors. We derive from figure 3 in Kreidenweis et al. (Env. Res. Lett., 2008, doi:10.1088/1748-9326/3/3/035002) that at a water activity of 0.7 ($\sim$ 70% RH), using a single κ under-predicts the radius of marine-type aerosol by $\sim$ 19%; for higher RH, the error is smaller.

We have replaced the expression

"This approach assigns a single hygroscopicity parameter κ ..."

with

"This approach assigns a hygroscopicity parameter κ ..."

p. 12269, l24: A density of 2 g cm^{-3} seems high for an average density assuming that most aerosol will contain a good deal of water. Even if the density is more like 1.2 g cm^{-3}, would it not make a large difference?

In order to address this, we have re-calculated the table of particle formation rates from aerosol nucleation using an assumed mass density of 1.2 g cm^{-3} (instead of 2 g cm^{-3}) for the pre-existing aerosol population, and repeated the simulation $\mathcal{S}_{\mathrm{ref}}$. This change has no effect on the annual mean ultrafine aerosol concentrations that are compared with the Clarke and Kapustin (2002) data. We have added a corresponding sentence in the manuscript.

p. 12272, l5: This issue is more for the people working on the implementation of lifetime effects in ECHAM5, but relevant here. Does the nudging affect the aerosol cloud-lifetime indirect effect, or are the nudging timescales much longer than the timescale of cloud dynamical changes imposed by the aerosols?

Nudging imposes a given general circulation on the simulations, while on smaller scales, clouds are free to vary in terms of optical properties and lifetime. This means

that the effect of aerosol on cloud lifetime is limited by large scale dynamics, enforced by nudging. However, this is precisely the case in the actual atmosphere. Nonetheless, it is a valid question to ask whether nudging (on the large scale) may in some other way limit the response of clouds to aerosol on smaller scales. There is no simple answer to this, as it is not straightforward to compare the effect of aerosol on clouds in nudged and non-nudged simulations: for a robust comparison, the non-nudged simulations would have to have the same large scale dynamics as the nudged simulation. What is commonly done is to compare a one-year nudged simulation with a non-nudged, multi-year simulation. Lohmann and Hoose (ACP, 2009, doi:10.5194/acp-9-8917-2009) have found that there is a limited difference in the effect of anthropogenic aerosol on net short-wave top-of-the-atmosphere radiation when comparing a nudged ECHAM5-HAM simulation for the year 2000, and the average of a 10-year climatological simulation (with no nudging). This difference may, however, be due to a difference in the large scale dynamics of the two simulations, as opposed to a limitation of the response of clouds to aerosol in the nudged simulation. The relaxation times we used for nudging in the prognostic equations are 48 h for divergence, 6 h for vorticity, 24 h for temperature, and 24 h for surface pressure. These are longer (less constraining) than used in other global model studies, such as in Hauglustaine et al., JGR, 2004, 10.1029/2003JD003957, or Telford et al., ACP, 2009, doi:10.5194/acp-9-4251-2009.

Section 4.1: Other possible reasons for too many CN3 are the coagulation sink being too low or below-cloud and in-cloud impact ion scavenging being too low.

We agree, and this is part of what we mean when we write " ... other model components are likely responsible for the overestimation of the ultrafine particle concentrations ...".

p. 12276, l13: I would not consider one order of magnitude change in nucleation rate a "large error" considering the huge uncertainties in nucleation rates. However, I am more a user of nucleation schemes rather than an expert in how uncertainty in thermodynamic parameters affects the rates of a given mechanism, therefore one order of magnitude may be quite large.

We have revised the corresponding passage of manuscript to read as follows:

- "However, nucleation parametrizations may exhibit errors in excess of one order of magnitude due to underlying uncertainties. We have repeated simulation $\mathcal{S}_{\text{ref}}$ with neutral and charged H_2SO_4 /H_2O particle formation rates reduced to 1/10 of their original values."

p. 12279, l14: "...aerosol nucleation is required...". I realize you mean "of the simulations ran, the one with nucleation gave the CDB agreement". I'd be careful about making general statements outside of the simulations tested since it would be quite possible to get agreement with CDB without nucleation by changing primary emissions or other aspects of microphysics (not that I disagree that nucleation is contributing to CDB).

We have removed the sentence

"At the same time, this test shows that aerosol nucleation is required in the model to obtain the observed CDB at the considered continental site."

Figure 9a vs. Figure 7c: I was surprised to see that the sensitivity of CDB to charged nucleation was significantly higher than the sensitivity of the accumulation mode to charged nucleation (this is true for activation nucleation, but most obvious for charged). The specific regions that made me concerned are the subtropics near the surface. In 7c, the sensitivity of accumulation mode # to charged nucleation is 1-5 % around 30 N or 30 S. In 9a, the sensitivity of CDB to charged nucleation is 5-15 % (and 10-15 % in oceanic regions). I would guess that in these oceanic subtropic regions, the accumulation mode is going to dominate the CCN, and if Aitken mode particles are being activated, they will quickly become part of the accumulation model through cloud processing. Therefore, I would have assumed that the sensitivity of CDB and accumulation mode to nucleation would be similar (CDB would be a small bit higher because there

has to be some flux of Aitken mode into the accumulation mode, but not this much). This was the cause of my general comment #2. My gut feeling here could of course be wrong and the flux of Aitken mode particles into accumulation mode is significantly large enough to cause the large difference in sensitivity of CDB and accumulation mode to nucleation.

We agree, the model does show a stronger response to nucleation in the cloud drop number than in the accumulation mode number. However, there typically are more Aitken mode than accumulation mode particles in the atmosphere, and a higher number of activated particles from the Aitken mode compared to the accumulation mode is not a priori inconsistent. The model may, however, overestimate the number of particles that activate from the Aitken mode. This may be because the model retains particles in the Aitken mode too long before placing them in the accumulation mode, or because the activation scheme activates particles that are too small. We anticipate that the continuous model development process will resolve these issues in the future.

Section 4: I'd be interested in hearing about the sensitivity of CDB and SW forcing to the solar cycle and its effects on charged nucleation. If this is not a paper in the queue, could a paragraph or two be added here?

A separate publication on this topic is planned.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 12261, 2010.