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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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Review of "Aerosol nucleation and its role for clouds and Earth's radiative forcing in the aerosol-climate model ECHAM5-HAM" by Kazil et al., ACPD, 2010.

This paper explores the impact of nucleation on the global aerosol size distribution, cloud droplet number concentration and cloud short-wave radiative forcing. The authors use a combination of charged-enhanced and uncharged H2SO4-H2O nucleation throughout the troposphere as well as activation-type nucleation in forested boundary-layer regions. This is the first time this pairing of nucleation schemes has been used in global models. Although the nucleation field is evolving quickly and we still don't know

all of the details regarding nucleation mechanisms, I feel that this pairing of nucleation schemes is consistent with the current understanding of nucleation (at least more so than previous studies, my own included). Thus, this work contributes very nicely to our field.

This paper fits well within the scope of ACP and is well written. I recommend it being published in ACP after several minor comments have been addressed.

General comments

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.

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.

Specific comments

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

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?

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?

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.

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.

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).

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.

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?

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