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Atmospheric
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Interactive comment on “Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates” *by* J. R. Pierce and P. J. Adams

Anonymous Referee #2

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Pierce and Adams present a global model study of CCN sources with a focus on uncertainties arising from simulation of nucleation and primary particle emissions. This topic is a highly relevant one as the concentration of climate active particles is one of the main outputs expected from large scale aerosol models. While several studies have previously looked at uncertainties from specific CCN sources (such as sea spray or mineral dust emissions), it is at least as important to investigate how the uncertainties related to these different sources compare with each other so that we can direct our research efforts to the most relevant unknown questions.

I am therefore happy to note that several research groups have picked on this im-

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portant topic recently. Just a few weeks before the appearance of this manuscript in ACPD, Wang and Penner reported in the same journal of their model results to very similar research questions. I also heard last month a conference presentation by a third American group tackling the same questions of relative climate uncertainties from nucleation and primary emissions. While the nearly simultaneous publication of these three studies is slightly puzzling, they must all be welcomed as valuable contributions to an important and current question - the models and the details of aerosol processes in these studies are different, thus giving some verification to the similar results obtained.

I have four major criticisms (listed below) concerning the current study. However, if careful consideration to these criticisms is given and the manuscript is modified accordingly, I believe the results of this study can help the modeling community to target its model development efforts better in the future. The methodology used is scientifically sound (with reservations detailed below) and the conclusions follow logically from the presented results. The manuscript is also well structured and written. I therefore recommend its publication in ACP after the authors have addressed the concerns raised in this review.

Major comments:

1) Although the exact nucleation mechanism(s) occurring in the atmosphere are not fully understood, it is very likely that nucleation rates and most certainly new particle and CCN formation rates depend non-linearly on vapor concentrations and other atmospheric conditions. Therefore I disagree with the authors' straightforward assumption that choosing two fairly extreme nucleation mechanisms gives meaningful bounds on the effect of nucleation on CCN. Binary mechanism is known to produce particles only in the FT and thus to severely underestimate nucleation rates in the BL. On the other hand, ternary nucleation has been shown to produce too high particle formation rates pretty much everywhere. The problem with ternary nucleation as a CCN source is that a very high number of nucleated particles compete for a limited amount of condensable vapor and thus only a small fraction of the nucleated particles gain enough mass

to become CCN. In other words, neither of these two mechanisms is optimal for CCN production (nor do they correspond well with observations).

It is therefore possible that a more realistic particle formation mechanism with a moderate nucleation rate both in the BL and FT can lead to a higher contribution of nucleation to CCN. This is because enough new particles are formed (in contrast to binary mechanism) but competition for condensation growth is not as fierce as when using the ternary mechanism.

To test this possibility, the authors should repeat at least their "basecase" run with a third nucleation mechanism that gives atmospherically realistic nucleation rates also in the BL. They can apply either recently proposed kinetic or activation mechanisms (e.g. Kuang et al., 2008, JGR; Sihto et al., 2006, ACP), or alternatively scaled down ternary nucleation rates. In my opinion this additional simulation is needed although Spracklen et al. (2008) and Wang and Penner (2008) both used activation nucleation in global models and obtained fairly similar numbers with the current study for nucleation derived CCN. Both previous studies used a model that is different in many details from the current one and thus their activation nucleation results cannot be generalized for the current model.

2) Nucleated particles are introduced to the model at a size of 10 nm by using Kerminen et al. (2004) formulation to relate the formation rate of particles at this size to nucleation rate at ˜1 nm. There are however some important assumptions behind Kerminen et al. (2004) equations that very likely are not met when they are applied to formation at 10 nm size. First is the requirement of constant growth rate of nucleated particles. As it typically takes at least several hours - and in many parts of the atmosphere more like a half a day - for particles to reach 10 nm, the concentration of condensing vapors is bound to change during this time (e.g. daily cycle of H2SO4). Second is the requirement for fixed pre-existing particle size distribution which is likely not to be the case at least in the boundary layer where particle sources and sinks as well as condensation growth modify the properties of the size distribution.

I am also concerned how the instantaneous growth of nucleated particles to 10 nm affects the nucleation rate at subsequent time steps as the treatment lowers the concentration of nucleating vapors unrealistically fast.

All in all, I have doubts whether reliable simulations of nucleation can be made with a model using a 10 nm cut-off size. I suggest the authors try to prove me wrong by making simple box model calculations with two set-ups: 1) current set-up in 3-D model (10 nm cut-off, Kerminen et al. formulation, long time steps), and 2) detailed size distribution down to 1 nm with sufficient number of bins, explicit simulation of condensation and coagulation growth of nucleated clusters from 1 to 10 nm, fairly short time steps, varying condensable vapor concentrations.

3) The model SO2 emissions seem to be from GEIA inventory for the mid-80's. However, almost all of the measurement data that the model has been compared against is from the late 90's or the current decade. SO2 emissions have decreased dramatically in the industrialized world since the 1980's (e.g. latest inventory from European Environmental Agency shows ˜60% decrease in Europe from 1990 to 2005). I therefore do not see any point in presenting the comparison of model output to the selected North American and European data. Furthermore, if the decision to reduce the fraction of primary sulfate emissions from 3 to 1% has been made based on a similar biased comparison, it can hardly be called an improvement in the model. One would assume a model with 80's emissions to overestimate the aerosol mass and number measured in the late 90's.

The best options would be to either compare the model output to measurements from the 80's (but sufficient data may not be available) or to switch to e.g. AEROCOM emissions for year 2000 and rerun the simulations (but this is very time consuming). If neither of these options is practical, my recommendation is that the detailed comparison to measurements (section 3.2 and figure 2) should be left out as it does not give reliable information on how well the model and the different nucleation mechanisms perform against observations.

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4) The results are presented as annual global and zonal averages. However, changes in CCN are most relevant on local to regional spatial scales and on much shorter than annual time scales. Because of this it can be misleading to make conclusions about the significance of a process or about uncertainties related to it on the basis of large scale averages. Therefore, the authors should discuss the spatial and temporal variability in their results and preferably present some of the figures as global maps at a model level that corresponds to a typical (low-level) cloud altitude.

Other remarks:

1) A very recent paper by Wang and Penner (2008) (ACPD, 8, 13943-13998) addresses some of the same topics than the current manuscript. The authors should compare their results and conclusions with those of Wang and Penner and discuss possible differences and similarities.

2) Change Vehkamaki to Vehkamäki.

3) p. 16300, lines 24 -27: The fact that a parameterization predicts nucleation onset correctly at one measurement location does not automatically mean that it has skill in predicting nucleation rates - or even onset of nucleation events in the atmosphere in general. This is the case with e.g. activation nucleation parameterization for boreal forests (Sihto et al., 2006, ACP) which cannot be applied to a Central European site without an order of magnitude modification of its prefactor term (Riipinen et al., 2007, ACP).

4) top of p. 16301: It is true that strong self-coagulation grows the nucleation mode particles and this decreases their coagulation coefficients with larger particles. But simultaneously particle number is lost in self-coagulation. Coagulation coefficient for self-coagulation is of course very low but coagulation loss depends also strongly on the particle number of both colliding particle sizes. Therefore, I don't think it is self-evident that neglecting self-coagulation in Kerminen et al. (2004) causes underprediction of 10 nm formation rate.

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5) p. 16306, lines 11-18: One could get an estimate of the relative importance of these factors by making quick box model calculations.

6) What is the motivation for including REDBINARY simulation? The standard binary mechanism produces particles only in the FT and doesn't severely overestimate the concentrations there.

7) Figures 6e and 6f should be discussed in the text.

8) Figure 2: some of the run labels do not match the run names given in table 1 (however, see major comment 3)

9) Figure 7: Change either the order of the panels or the sentences in the text. The text discusses first panel 7c.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16291, 2008.