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Atmospheric Chemistry and Physics Discussions

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

J. R. Pierce and P. J. Adams

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We would like to thank this reviewer for helpful comments and recommendations that have strengthened the paper.

Major comments:

The authors find that both CN and CCN concentrations are surprisingly insensitive to the atmospheric nucleation rate. I wonder whether this is a real feature of the system or whether it results from the chosen nucleation mechanism combined with the numerical treatment of the problem. My arguments are the following:

Apparently, the insensitivity of CN and CCN concentrations results from the extremely low Number Utilization Efficiency (NUE) for ternary nucleation, as compared with binary nucleation. The

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authors state that the low values of NUE for ternary nucleation are caused by very high nucleation rates which lead to large condensation and coagulation sinks, as well as underestimation of the contribution of self-coagulation to nuclei growth when using the parameterization scheme. I fully agree with the authors that this is the likely explanation. In the real atmosphere, however, nucleation mode particles rarely give a significant contribution to condensation and coagulation sinks (this can be easily checked out from measured particle number size distributions in different environments during new-particle formation events). Also, nucleation mode particle number concentrations are not usually high enough (except maybe in certain urban centers, power plant plumes) to make nuclei self-coagulation a very important contributor to nuclei growth. Thus, the very low NUE may be an artifact resulting from too high nucleation rates rather than a real feature of atmospheric aerosol dynamics.

I strongly recommend that the authors investigate this issue carefully. The simplest solution would be to artificially reduce the ternary nucleation rate by a fixed amount (for example by factors 10, 100 and 1000). Another option might be use a third nucleation mechanism such as the simple activation-type nucleation that depends linearly on sulfuric acid concentration.

Our point is not that the nucleation mode particles themselves make significant contributions to condensation/coagulation sinks, and we agree with the reviewer that they generally do not. We also agree that the nucleation rates from the ternary nucleation parameterization are likely too high globally. It is possible that the relationship between nucleation and CCN may not be easily extrapolated between the binary and ternary cases that we presented. We have taken your recommendation to run an activation-type nucleation simulation. We use j(1 nm)=1.0E-6*[H2SO4] as the nucleation relationship in the boundary layer (Sihto et al. 2006) and Vehkamäki binary nucleation in the rest of the troposphere. Due to long simulation times were are only able to run the model for 3 months for this scenario (two months of spin up and one month of results). However, we are able to compare the results for this month (June) with the results of the BINARY and TERNARY simulations for the same month. The tropospheric average nucleation rates were 8 times larger than in BINARY, and the CCN increased by 5% both in the boundary layer and across the entire troposphere. The CCN here are about half as sensitive as the BINARY:TERNARY comparison. If in fact the TERNARY

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results are unreasonably high, this would reduce the importance of nucleation uncertainty on CCN uncertainty. Even with reduced primary emissions and high SOA it is not likely that the CCN concentrations from the activation-type nucleation simulations are greater than that of the TERNARY.

We have added this simulation, ACTIVATION, to the text, included its global budgets in Table 2 and shown its effects on CCN in the boundary layer in a new Figure (Figure 5).

Minor comments:

The chosen ternary nucleation mechanism is sensitive to gaseous ammonia, especially at low ammonia concentrations. Is this consistent with how gaseous ammonia concentrations have been calculated in the model? The model predicts gaseous ammonia only when it is present in excess of neutralizing the sulfate aerosol. In atmosphere one would expect to see some gaseous ammonia also in acidic aerosol systems.

This is true. We have added the following sentences to the discussion regarding the addition of ammonia to the model. "Realistically, gas-phase ammonia is present even when the aerosols are acidic; therefore, our assumption about gas-phase ammonia being present only when sulfate is neutralized may lead to an underprediction of gas-phase ammonia and ternary nucleation rates. As will be discussed in the next section, ternary nucleation rates are already very high, and this underprediction does not likely affect the results."

Is it realistic to have any fraction of SO2 to be emitted as primary particles? The model has already primary carbonaceous particle emissions and a large fraction of SO2 is anyway co-emitted with these particles.

A discussion of this is included in Adams and Seinfeld (2003). They represent sulfate particles emitted both directly from a source or those nucleated near the source within the plume that would be very difficult to predict in a global model. These emissions are, of course, very uncertain.

To roughly assess this uncertainty, we have our reduced primary simulations that re-

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duce these (along with all other primary emissions by a factor of 3).

How sensitive the system is to the assumed mean size of primarily-emitted particles?

It is expected to be reasonably sensitive. Unfortunately, with the large number of simulations and comparisons in the paper already, we did not address this issue directly here.

However, there are two good indicators of how sensitive it can be. 1) Between Pierce et al. (2007) and this paper, we changed the number median diameter of all biomass and biofuel burning carbonaceous emissions from 30 nm to 100 nm. The CCN concentrations decreased globally by over 10% from the changes in these emissions alone. 2) Much of the difference in the sensitivity of CCN to nucleation rates between our paper and Makkonen et al. (2008) may likely be from very different assumed primary sulfate size distributions (they emit it almost entirely into the accumulation/coarse mode, adding very few particles).

We now address this point in the last line of the paper. “Furthermore, we did not specifically address the changes in CCN to changes in the primary emission size distribution while keeping the primary mass emissions rates constant, and this will contribute to additional uncertainty in CCN from primary emissions.”

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