Atmos. Chem. Phys. Discuss., 6, S6453–S6459, 2007
www.atmos-chem-phys-discuss.net/6/S6453/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Efficiency of cloud condensation nuclei formation from ultrafine particles" *by* J. R. Pierce and P. J. Adams

J. R. Pierce and P. J. Adams

Received and published: 24 January 2007

Response to Anonymous Referee 3

Referee comments in *italics*

Referee 3, thank you for the detailed read that you gave our paper. There are many comments that you have made that have helped make the paper more clear and understandable.

"This paper describes a model that calculates the efficiency to form CCN from size distributed ultrafine particle numbers, with no reference to particle content. The paper is an important input to the understanding of the effect from ultrafine particles onto the climate system, as the efficiency of CCN formation from ultrafine particle varies a lot. It uses particle size and is based onto standard particle dynamics, i.e. coagulation, condensation and deposition, but also onto availability of condensable gases, affecting the

condensation rate; background particle size distribution, affecting the coagulation process; and content of the particle, affecting particle hygroscopic growth etc. The model assumes the same content of all particles, i.e. ammonium bisulphate, and only includes one of the condensable gases, i.e. sulphuric acid. The influence onto the result by not including effect from other condensable gases, and from size distribution change during the process is not totally transparent."

In most of the uses of the PUG model within the paper sulfuric acid is used to either test the PUG model (against the TOMAS box model) or to help illustrate a point (e.g. in order to predict the number of CCN generated from an ultrafine emission, the exact size of the emission must be known). The PUG model is completely general, however; these same things could be done with any condensing gas or mixture of gases. We chose sulfuric acid because it is easy to use (its equilibrium vapor pressure is essentially zero). We have added text to clear up this point. At the beginning of Section 4.2, we added "H_2SO_4 is chosen here as a representative condensable gas to facilitate the comparison of the two models. This choice is not meant to imply anything about its atmospheric importance relative to other condensable gases (e.g. SOA)." In Section 5.2, we added "Again, H_2SO_4 is used here as a representative condensable gas to help illustrate the uncertainties in CCN generation."

In the case where we are showing the predictions of CCN efficiency in various parts of the world and have only included sulfuric acid, not including growth from other condensing gases. We have added text to show that this does not give the complete picture of the CCN efficiency. In Section 5.1, "In the simulation used, there are no carbonaceous or dust particles and H_2SO_4 is the only condensable gas; thus, the timescales and efficiencies shown here are not completely representative of the real atmosphere. However, we expect that the main points determined from this exercise regarding the variability of CCN formation efficiency within regions of the atmosphere and between regions will still be valid when all of the species are included."

We have added text to address the changing of size distributions throughout the aerosol lifetime. In Section 5.1, "It should be noted that the timescales plotted are for specific

ACPD
6, S6453–S6459, 2007

EGU

locations (grid cells in the GCM) and the corresponding CCN efficiency applies only for that location. As particles are transported throughout their lifetime, the efficiency will change with their location."

"The model is compared to another model, developed by the same researchers, and then used to decide the CCN formation efficiencies in different regions of the Earth. The resulting efficiency for a particle to become a CCN often lies between 5 and 40%. Different regions of the atmosphere have different probabilities. The reason for this is a bit vaguely described, even if the regions dealt with are described, the parameters included into the model to differentiate between the regions are not totally clear. The paper addresses relevant scientific questions within the scope of ACP. It also presents a novel tool to describe and calculate the CCN formation efficiency. In some parts the description of input variables to the model should be more specific. The results from the particle dynamics seem OK to me. This paper describes a comparison between models. What about comparisons to real life or lab data?"

We have added text to address the inputs into the PUG model for the different regions of the atmosphere. In Section 5.1, "Time-averaged aerosol size distributions and H_2SO_4 concentrations from various parts of the atmosphere, taken from GCM grid cells within the spatial coordinates described below, were used to find the growth and removal times of the 30 nm particle."

We have added a description of the input variables to the abstract to increase clarity. "The inputs to the PUG model are the concentrations of condensable gases, the size distribution of ambient aerosol, particle deposition timescales and physical properties of the particles and condensable gases."

We have not done any comparisons to real life or lab data. The physics used in the model (coagulation, condensation) are time-tested and should be sound.

"The paper is in principal clear and well written."

"Abstract: Would prefer to have the input variables specified already in this context, as this implies an important description of the of the model, e.g. that the variable hygroscopic growth factor is not taken into

ACPD
6, S6453–S6459, 2007

account, only size."

This is a good suggestion. See the text above for how we changed this; however, the hygroscopic growth factor is taken into account (this is why we assumed a composition and a relative humidity when we were illustrating points).

"p. 11001 r. 6: "the CCN deposition lifetime, which typically will be nearly equal to the aerosol mass lifetime computed by global models." Please show or give reference."

Added a clarification by listing the assumptions necessary to reach this conclusion. In Section 3.2, "It is convenient to re-express ΔCCN in terms of the CCN deposition lifetime, which typically will be nearly equal to the aerosol mass lifetime computed by global models. This happens because most aerosol mass is in particles of CCN sizes, and the lifetime of these particles is dominated by their wet deposition lifetime, which does not vary much with size of CCN particles."

"Section 4.1 and 4.4 The compared models: The choice of the model to compare PUG with is probably due to practical reasons, as the model has the same developers as the PUG model. The possibility to use another model should be further discussed, as the compared models have very different approaches that affect the implementation of the size distributions between the models. The PUG model bases the calculated coagulation rates etc. onto the initial size distribution. How much does this differ from the obtained size distribution, if that would be calculated? The size distributions in each of the models, i.e. PUG's initial size distribution as well as the end point one calculated from the processes taken part in the simulation, and for the other model both the initial size distribution and the equilibrium size distribution should be shown in Figures for the reader to decide if the differences are big or small. Please add figure, e.g. size distributions!"

To simplify the PUG/TOMAS testing we have modified the comparison by using the equilibrium size distributions from the TOMAS model as inputs to the PUG model so both models are calculating the CCN efficiencies using the same ambient aerosol size distributions. We have split the old figure 3 into two figures (new figures 3 and 4). Figure 3 compares the PUG model to the TOMAS box model using the same distributions

ACPD
6, S6453–S6459, 2007

and Figure 4 compares the PUG model to the PUGC model the same way as before. Section 4.3 discusses only the results of the PUG/TOMAS box comparison and we have added section 4.4 to discuss PUGC and the importance of the various growth and sink processes.

"p. 11003 r. 12- The reasons for the choices in the case used for comparison is not clear to me, e.g. why was all particle content ammonium bisulphate? There are available papers that recommend that the content of particles is of minor importance to CCN formation efficiency compared to size that could be included as reference (e.g. Dusak et al, 2006)."

See our response from earlier. For testing the model against the box model, the choice of condensing gases is an arbitrary one of convenience, and we are not trying to imply that sulfate is the only species that condenses and that all particles are inorganic. It was only chosen so we could have aerosol properties to use.

"p. 11004 r.10 Why remove aerosol microphysics when choosing the emission rate?"

This is somewhat of a misinterpretation of what was said. What we mean is that in the TOMAS box model that aerosol microphysics (condensation and coagulation) affect all of the particles in the box. The shape of the size distribution in the box is therefore different than the shape of the size distribution flowing into the box (because the particles are changed by these processes). If we turned these microphysical processes off in the box the size distribution in the box would be the same as those in Jaenicke (1993). This sentence was removed during our reworking of Section 4.

"p. 11004 r.2 "The 90 nm cut-off was used because it corresponded to one of the mass doubling sizes in the model" What would happen if you used e.g. 120 nm? Would it be possible to use something close to 90, i.e. not a specific size bin? How would that affect the result?"

The model can be made to calculate efficiencies at intermediate diameters; however, choosing a diameter that corresponded to a specific size section simplified calculations. If the diameter the particle must grow to is increased the CCN efficiency decreases. We

ACPD
6, S6453–S6459, 2007

EGU

have addressed this point. In Section 4.3, "A CCN cutoff size in between the doubling sizes of the model may also be used by modifying the equations in Sect. 3; however the 90 nm cutoff was used here for simplicity."

"p 11006 r.4: "does not change much" What is much, please put number!"

We appreciate the suggestion. It is assumed that the aerosol composition does not change at all during this calculation. In Section 5.1, "It is assumed that the aerosol composition does not change as it grows such that the aerosol water mass per unit dry mass remains constant and the aerosol density remains constant."

"p. 11006 r. 14: Boundary layers values; please use a more specific word than "values", e.g. "time-scales"."

Thank you. "Boundary layer aerosol size distributions and H_2SO_4 concentrations"

"The PUG model seem to be a first step towards a model that can be used to describe the CCN efficiency, based on more variables than present in the model today, and thus more realistic."

This is correct. For example, future work could estimate CCN formation efficiencies again for different parts of the troposphere but updated to include the effects of SOA condensation, presence of carbonaceous aerosol, mineral dust, etc.

"p. 11008 r. 23: The traffic emitted aerosol normally has even larger variation between sizes, according to my knowledge: I'd probably test 10-60 nm. Would that make a big difference? Here a possibility to understand the effect from content onto the CCN formation efficiency is large, as the smaller particles (<20 nm) mostly consist of oil droplets and the 60 nm particles are soot, so why assume ammonium nitrate? Why assume anything if the content is unimportant? Traffic emitted particles also comprise a large part of the anthropogenic emissions present, and thus the CCN formation of the traffic exhaust would be of large interest."

The range of sizes tested refers specifically to the median diameter (geometric mean diameter) of a lognormal size distribution. Based on all the literature of which we are aware, we feel that the size range we used amply represents the uncertainty/variability.

ACPD

6, S6453–S6459, 2007

Moreover, it is unclear from the comment whether the reviewer is specifically recommending 10-60 nm for the range of median diameters or merely as representative of the range of all particle sizes. If the latter is meant, our assumed lognormal size distributions do already span this space. If the reviewer specifically intends 10-60 nm as the range for median diameters (and can point us to an appropriate reference), we will happily test the larger range. Of course, a wider range would only serve to strengthen our main point that knowing the emitted size distribution (as well as total mass) is necessary for predicting eventual CCN formation.

We realize that these particles are not ammonium bisulfate and have added further clarification on this point. In Section 5.2, "Although traffic emissions are primarily composed of carbonaceous species, for simplicity we have assumed that all particles are ammonium bisulfate. This does not affect our conclusions about the uncertainty of CCN generation." The reason why ammonium bisulfate was assumed was strictly to get properties. It has no effect on the main point in that you need to know more than just the total mass of aerosol emissions in order to determine the number of CCN that the emission will generate.

"Figure 6: I'd suggest to put also the "urban aerosol" into the figure, on another y-axis naturally."

We understand the aid of visualizing the size distribution here; however it also makes the figure busier and less concise. We have given the parameters of the 3 lognormal modes that describe this distribution in Table 1.

ACPD
6, S6453–S6459, 2007

EGU