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Interactive Comment

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

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Response to Anonymous Referee 1

Referee comments in *italics*

Referee 1, thank you for the detailed read that you gave our paper and the comments that you made on it. Particularly helpful was the point you made regarding equation 4.

"This clear and well-written paper presents a new model (PUG), developed in order to predict probabilities for ultrafine particles to grow to CCN size. In addition to presenting the model features, the example calculations address two intresting questions regarding CCN formation: 1) What is the effect of emissions uncertainties? and 2) Can one single moment of the size distribution used to predict CCN concentrations? The paper is definitely interesting to the atmospheric research community and deserves publication, after some points are addressed in more detail.

Major comments: 1) As the authors explain, coagulation may be an important growth mechanism in

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polluted conditions. I am worried that the 'book-keeping-procedure' presented (eq. 2, page 10997) might be inaccurate (or even incorrect?). If one wishes to determine the growth rate at a certain size of a coagulating aerosol, is it enough to look at the collisions at that size with all smaller particles? Did the authors check this method with a case in which coagulation dominates (or, causes all) growth, by using their more detailed model? ."

We are not entirely sure what the reviewer's concern with the book-keeping procedure is. We believe it is a misunderstanding due to us not explaining ourselves well enough. No process has been omitted from the model and the "book-keeping procedure" is necessary to avoid double counting coagulation events. The close agreement between the PUG model and the more detailed box model microphysics calculations is evidence of the correctness of the formulation. The "book-keeping procedure" is necessary for the following reason. During any coagulation event, you start with two particles and when they coagulate you are left with one particle that has the mass of the two original particles combined. We assume that in all coagulation events that the smaller particle is removed and that the larger particle is not removed and grows by the mass of the smaller particle. In the case of two particles of the same mass coagulating, one particle is removed and the other doubles in mass. This procedure is consistent with the definition of what occurs during a coagulation event. The growth of a particle due to coagulation is thus equation 2 and equation 7 where all of the particles smaller than (or equal to) the particle in question are considered for coagulation growth. We changed text to help clarify this. The corresponding part of Section 2 now reads "The second process that contributes to the growth of ultrafine particles is coagulation with smaller ultrafine particles; therefore, coagulation is both a growth and a loss process. When two particles coagulate, a single particle is formed with the mass of the two combined particles. For book-keeping purposes, we define that, during a coagulation event, the smaller particle is removed and the larger particle survives and grows by the mass of the smaller particle. The growth rate of a particle with respect to coagulation is then a function of the rate of coagulation with particles smaller than it and the mass of those smaller particles."

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“Minor comments: 2) In the beginnig of section 3, it is stated that a constant background is assumed. This might be severe in some cases, since growth to CCN size can take a while. In addition, loss to background is very sensitive to the background distribution. Were any test simulations done with a varying background? (with TOMAS?) .”

This is an important point that perhaps was not discussed enough in the paper. Although we have not tested simulations with varying backgrounds, we have added the following caveat to Section 5.1, “It should be noted that the timescales plotted are for specific 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.”

“3) Based on the paper, it remains unclear, if PUG is intended to be used as a ’separate’ tool for quick estimation purposes or can it be implemented into a larger code to speed up things etc. Please clarify. .”

This was not discussed enough in the paper. Towards the end of the introduction, “This model was developed to be used as a stand alone model for quick calculations of the CCN formation efficiencies for a given set of atmospheric conditions” was added.

“4) Equation 4 is by no means straightforward. Please derive or explain. (Growth and scavenging is expected to result in exponentially decaying concentrations, why is this not visible in eq. 4?) .”

We are glad you pointed out that this equation needed an explanation. On further thought, we realized that modeling the growth properties with an e-folding timescale was incorrect. The correct form calculates the number of particles that were lost through the first order loss processes during the time its taken for the particles to double in mass through growth processes. Actually, our old Equation 4 gave the same results as the corrected one in the limit of the growth timescales being faster than the removal times. However, the predicted efficiencies are reduced when the removal times are faster than the growth times. We have updated all of the figures and the values given in the text to reflect the changes; however, none of the main points of the paper have changed. A comparison of the old and new figures shows that the effect of the correc-

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tion is noticeable but minor. Please see the modified equation 4 and the text around the equation.

"5) On page 11004, when discussing the differences in PUG and TOMAS results, it is stated that the differences are only from initial distributions. Is this really true? Does this also apply to cases in which coagulation dominates growth? ."

It does not state that the differences are only from the initial distributions, it listed that as one of the reasons.

Based on comments by the other reviewer we have somewhat changed this section anyways and are now comparing the PUG model and the TOMAS box model using the exact same ambient distributions. The differences between the two models are now from numerical error from the discretized size distributions in the two models.

"6) In fig. 4, is loss of number by self-coagulation taken into account? ."

We aren't sure if you are referring to coagulation with particles with very nearly identical sizes (which we do take into account) or if you are referring to coagulation with other particles that are emitted in the same place (which is irrelevant here since we are calculating the CCN efficiency of a single particle).

"7) In fig. 4, 'mass doubling lifetime' is given as a variable. This should be made more clear by giving some examples of e.g. corresponding growth rates (in nm/h). ."

An example is added to the text towards the beginning of Section 5.1. "For reference, when a 30 nm diameter particle doubles in mass, it is then 38 nm in diameter (assuming the density doesn't change). Therefore, the growth rate of the 30 particle in nm hr^{-1} may be approximated by dividing 8 nm by the mass doubling timescale (e.g. for a 10 hour timescale the growth rate would be 0.8 nm hr^{-1})." We considered several metrics for condensation rates and decided that we preferred gas-phase concentrations and growth timescales. We felt that particle growth rates (in nm $^{-1}$) suffer from two drawbacks: (1) they seem to be familiar mostly to the subcommunity of scientists who work

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in the area of nucleation and (2) the particle growth rate is a strong function of particle size selected, so it is not a unique or unambiguous value like gas-phase concentration is.

“8) On page 11006, it is stated that a single size distribution shape is used. What is this shape? .”

We added it in Section 5.1. “Here we have used the PUGC model to create a smooth efficiency surface. Using various size distribution shapes will have only a minor effect on the predicted efficiency.”

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