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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Numerical simulations of homogeneous freezing processes in the aerosol chamber AIDA" *by* W. Haag et al.

Anonymous Referee #2

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This paper is well written and informative. The success in the comparison of cloud chamber studies with box model simulations indicates that homogeneous freezing of sulfuric acid aerosols can be modeled confidently, at least for large cooling rate conditions. Uncertainty is carefully discussed.

In the companion paper, Möhler et al. denoted that the optical particle counter PCS2000 measures particles with diameters between about 0.5 and 20 micron and the overall count rate above 1.5 micron is converted into a number concentration of ice particles n_i. The paper also noted that the sampling effciency is significantly below 100% for particles D $>$ 10 micron. The sampling efficiency for D $<$ 10 micron is not mentioned. Maximal n_i measured during each experiment is then compared with the number concentration (take A3_1 for example, n_i of APSC at around 350 s) predicted by the box model (Table 1).

I am somewhat concerned about the detecting limits and sampling efficiency of the optical particle counter. Take A3_1 for example (Figure 7), the modeled nucleation starts at around 330 s and ends at around 350 s. The time that the optical counter detects signals compares well with the model; however, the measured n_i gradually increases with time and reaches its peak value at around 900 s. Furthermore, at 380 s, a big portion of predicted ice crystals have grown to sizes greater than 1.5 micron (Figure 8). The above 2 observations seem to indicate that, for the small end of the size spectrum, either the actual detecting limit of the optical counter is probably not as small as stated or the sampling efficiency is much less than 100%. This issue has to be addressed and discussed in the paper.

With this uncertainty in the measurement, in order to reach the conclusion of section 6.3, the authors have to demonstrate that the sizes of the predicted ice particles at 900 s for the α=0.05 case are large enough to be measured.

Technical corrections

Page 1472, line 19-21: "All particles are moved to the same size bin and are not partitioned amongst two or more size bins" is a disadvantage rather than an advantage. From time to time, the size distribution will be segmented (a hole in the spectrum) due to the above artificial transferring of particles from one bin to another. For models using this type of bin scheme, usually a companion interpolation scheme is used to interpolate the size spectrum and partition particles back to the empty bins.

Page 1475, line 17: Probability = 1 - exp (-JVt). "proportional" is not the correct word.

Page 1476, line 8: The decrease of number density of H2O (number of molecules/volume) is not directly related to the discussion of increase in RHi (e/e_s, where e and e_s are vapor pressure and saturation vapor pressure, respectively). I think that the authors meant vapor pressure rather than number density of H2O for this sentence.

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Page 1479, line 5: Add "at" around 330s.

Page 1480, line 1: "... do not take up H2O significantly". Although the large aerosols have lower H2O mass fraction, they take up larger amount of H2O mass compared to the smaller ones.

Page 1481, line 7: Consider to replace "mean free path of H2O molecule in air" by "the effective mean free path for diffusion of H2O molecule in air"; line 22, "To obtain lognormal aerosol size spectra" or "To obtain parameters for lognormal aerosol size spectra"?

Page 1490, line 5: In the context, "limited influence" in the paragraph seems to refer to their influence on the conclusion of this work. However, the phrasing of the last sentence of this paragraph seems to describe a general aspect. Take A3_1 for example, α = 0.2, n = 171; α = 1.0, n = 60. That is about a 3-fold difference and is hardly a limited influence. Please rephrase.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 1467, 2002.