

Interactive comment on “An approximation for homogeneous freezing temperature of water droplets” by K.-T. O and R. Wood

Anonymous Referee #1

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This manuscript presents a new parameterization to predict homogeneous freezing temperatures of water and aqueous solution droplets in the atmosphere. Using the number of critical embryos formed in a droplet as a result of critical fluctuations, based on classical nucleation theory, the authors show that the derived temperature at which the number of critical embryo equals one, can reproduce experimental studies including freezing from water droplets and aqueous solution droplets. As a result, it is found that the spread of homogeneous freezing temperatures is largely governed by differences in droplet size (volume) distribution applied in the ice nucleation experiments. As such, this new parameterization is suggested for predicting homogeneous ice nucleation in the atmosphere.

General comments:

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This is a nicely written manuscript dealing with a topic that fits well within Atmos. Chem. Phys. I am in favor seeing this manuscript published. However, before I can recommend this work for publication some general and specific comments should be addressed or sufficiently clarified.

Equation 1 is the foundation of this work. However, as far as I recall, not the mean number of critical embryos is derived but it gives the number of i-mers of certain size formed for a given fluctuation (as given e.g. in Pruppacher and Klett). This reflects the partitioning function of the grand canonical ensemble. More information has to be given why this equation should reflect a mean number of critical embryos and which size of the critical embryo was assumed. The size of the critical embryo may depend on other thermodynamic parameters. Please elaborate.

As stated above, I like this work, but it is not clear to me what is gained with regard to atmospheric application compared to previous parameterization, e.g. by Koop et al. (2000)? Computationally, the formulation by Koop et al., it seems, is still more efficient. Usually in a model, one knows time, either as a model time step or by given updraft velocities, and if not, one could just assume a time constant for the Koop et al. formulation. The neglect of time in this study works because close to the homogeneous freezing limit the nucleation rate coefficient is a very steep function of temperature. As such, in explanation of the spread in ice nucleation experiments, there will always be an effect of time but possibly negligible compared to the volume effect. If the authors could make a case why this parameterization is of advantage in implementing into cloud models, this would strengthen this paper.

It would be interesting to know at which spread in size distribution, time considerations (or vice versa i.e. time versus volume effect) are important. This could help guiding experiments.

Specific comments:

p. 31868, l.5-6: “Without consideration of time dependence and stochastic nature. . .”.

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I understand why you write this here but it could be misunderstood that homogeneous ice nucleation is not time dependent or not stochastic, which it obvious is. In fact, your basic equation is derived from CNT that assumes fluctuations. Here, you can neglect time dependence since the nucleation rate is so steep with respect to changes in T. I suggest to clarify this statement.

p. 31868, l. 16: Would it not be better to call it ice melting temperature instead of equilibrium temperature?

p. 31868, l. 21: ...of temperature and time...? Previous experiments when deriving nucleation rate coefficients interpreted their data using droplet volume and time including Koop et al. (2000).

p. 31868, l. 23 following: Regarding the Riechers et al. study. Do you mean they are the only one who reported droplet size distribution for one given droplet size (i.e. the deviation from a monodisperse droplet distribution)? Maybe clarify.

p. 31871, l. 5: Why should the fluctuation probability be higher in larger volumes? The fluctuation probability is in principle an energy term and thus is independent of volume. It depends on temperature, supersaturation, surface tension but not volume? Since in this parameterization molecular fluxes are not considered, there is no volume dependence. Please elaborate since this statement is not clear from given information.

p. 31871, l. 17: Please add a reference at the end of this statement.

p. 31872, Eq. 3: Why is the decadal log used for the sensitivity of droplet diameter?

p. 31876, l. 6-10: Could you clarify this statement? What is the call for more "potentially important dependencies"? If not, maybe avoid this statement.

p. 31876, l. 19 and following (discussion Fig. 4): There are a couple of points regarding Fig. 4 which may be helpful for the authors: i) I am wondering why the authors did not also plot the data of Swanson, Knopf and Lopez (2009), and Knopf and Rigg (2011), the latter ones being a much more extensive data set? ii) Knopf and Rigg

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(2011) and Riechers et al., argue that J_{hom} by Koop et al. (2000) may be ~ 2 orders of magnitude too high. Does this effect interpretations/derivations of this study? iii) The reasoning for the deviation at lower a_w is not complete. Abbatt and co-workers observed higher freezing temperatures due to heterogeneous ice nucleation. Swanson observed freezing below the homogeneous freezing line, this usually indicates other issues than a heterogeneous nucleation process. For example, the droplets may have possessed less water than indicated by experimental RH (not in equilibrium, mass transfer, etc.). In addition, at lower a_w , the assumption that a_w does not change with decreasing temperature may be less "true". See e.g. E-AIM model by Clegg and co-workers. Deviations at low a_w could be due to our incomplete understanding of a_w for certain aqueous solutions.

Technical corrections:

p. 31872, l. 11: missing space after first comma.

p. 31874, l. 22: Change "sold" to "solid".

p. 31875, l. 10: Maybe instead "by" use "using".

p. 31877, l. 14: Maybe "to" instead "with".

p. 31878, l. 18: ...shifted to...

p. 31879, l. 21: ... higher than...

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