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Review of acp-2022-434

Anonymous Referee #2

Referee comment on "Impact of formulations of the homogeneous nucleation rate on ice nucleation events in cirrus" by Peter Spichtinger et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-434-RC2, 2022

The manuscript by Spichtinger et al. revisits the impact of nucleation rate formulation on the outcome of homogeneous freezing events. These investigations are motivated by two recent studies reassessing the saturation pressure with respect to supercooled water (Nachbar et al., 2019) and the homogeneous freezing rate in pure supercooled liquid water (Koop and Murray, 2016). Without questioning the crux of Koop et al. (2000) water activity criterion (WAC) theory, the new parameterizations, if correct, would imply changes to the commonly-used formulas of homogeneous freezing rates in Koop et al (2000). More generally, these recent results emphasize uncertainties in the formulation of homogeneous freezing rates, as recognized by the authors of the present study.

In this context, the paper introduces and evaluates meaningful approximations to the nucleation rate in order to identify and discuss the key parameters for the outcome of nucleation. The authors also propose an updated formulation of the homogeneous freezing rate of aqueous solution droplets in the frame of the Koop et al. theory.

The manuscript should be a valuable contribution to ACP, but I have a few concerns about the method and presentation and recommend that the paper be reconsidered after major revisions. My comments, requests for clarification and suggestions to the authors are detailed below.

Main points :

1) Consistency between liquid saturation pressure and nucleation rate: As far as I understand, for consistency with the Koop et al. (2000) method, each change in the saturation formula (and implied change in ice water activity) requires updating the fit to the homogeneous freezing rate of pure water. Was this carried out when switching to Nachbar's formula in section 4 ? It should be specified if yes, done if not.

Related issue: the authors have already published on the impact of Nachbar's formulation in Baumgartner et al. (2022). Differences should be highlighted. If there are none, the whole section 4 (including Figures 10 and 11) can be removed and summarized in just a couple of sentences (in particular since the effects are small).

2) Clarity and organization: I would suggest the authors start by presenting the uncertainties regarding the nucleation rate and the new formulations before introducing their approximations and finally analyze the impact of the updated formulas. This would make the motivation clearer. I also think the manuscript would benefit from shortening. Many equations and comments are redundant (see below).

3) Suitability of the model to study details of ice nucleation: in a comment to this paper, Bernd Kärcher expressed skepticism regarding the ability of a bulk model to study ice nucleation. For such a focused study, expected limitations of the modeling approach should be discussed.

4) What is the reason for the simple adjustment to the Koop et al (2000) formula in Sect. 3.1? This seems to create a 'third' mixed category between the former and updated formula. I would recommend restricting the study to those two cases.

5) I had difficulties following some sections, in particular 3.6. At other places, the text sometimes falls into tautology: in section 3.4, the authors arrive at Eq. (45) which as far

as I understand is exactly the same as Eq. (43) and (42) if we consider that Sc also depends on j0. Eq.(42) is repeated only a few lines below as Eq.(43) on page 11.

6) There are some ambiguities in the notations, for instance pressure and the polynomial are both represented by the same symbol (p) . I suggest clarifying and adding a table with a list of symbols.

7) I am not entirely convinced about section 6. The relevant quantity for atmospheric modeling is the ice crystal number density, not really the threshold which is mostly specific to chamber experiments. Moreover, the 'Koop-line' depends on the aerosols size distribution and is not supposed to represent the maximum supersaturation reached, rather an approximate ice onset.

8) Diffusion growth: it seems that it was originally intended that the paper also treats the sensitivity to diffusion growth (the summary line 644 still mentions it). While I agree this is beyond the scope of the study, could the authors comment in the text on whether the sensitivity to nucleation rate formulation they characterize holds for different growth parameters? Also, it would make sense in this case to have Part 1-Part 2 papers.

Specific comments:

Title: should mention homogeneous nucleation or homogeneous freezing since it is the only nucleation pathway considered in the paper.

P4 Model description should state that the model has two moments.

Line 111 and 132: There is an inconsistency, it is first stated that latent heat release is neglected but this term is still included later.

line 140-141: Same as above, the first term comes from considering latent heat.

Line 164-165: This disagreement at temperatures above 235 K is indeed surprising. Have you confirmed by comparing with the original Pruppacher 1995 data used by Koop et al. (2000) ?

line 181: missing index n

line 258: eq. 41 is the same as eq. 31

line 340, ‘most’: not all ?

line 701: ISO convention for natural logarithm is ln , not log

Line 747-748: Please check formula A11 and please correct if needed. Also define L.

Line 751: for which value of the eccentricity ?