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Comment on acp-2022-434

Anonymous Referee #1

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-RC1>, 2022

In this paper the authors revisit formulations of the nucleation rate coefficient for homogeneous nucleation of aqueous solution droplets. The mathematics of their approaches is derived thoroughly and the results of numerical experiments are presented, so that the reader can see the effect of various simplifications. This is a very good paper, but I think, it can still be improved. In particular, the motivation for this work is not expressed clearly. This is my major point of critique, and I think with a minor revision the paper should be publishable.

Specific comments:

"In this study we will investigate the impact of the formulation of the nucleation rate on the resulting ice crystal number concentrations."

I miss the motivation for this re-assessment. Has it turned out recently that the usual formulation (using Koop et al. 2000 essentially) is wrong?

The authors mention that the old Mishima-Stanley theory is now ruled out and that two alternative water theories have survived which give similar results for upper-tropospheric conditions. It is not clear to me, however, how this fact impacts on the Koop-et-al 2000 findings and their formulation of the nucleation rate coefficient. The implications of this finding is not given in the paper, and in fact, these theories are not mentioned again. It is thus unclear why this is mentioned at all.

"However, the representation of these processes contains still uncertain parameters or even the (mathematical) formulation of the processes remain uncertain." Perhaps this is the point where the authors should become more concrete. What are the uncertain parameters and in which way is the mathematical formulation uncertain.

Line 75: It would be good to list the mentioned parameters.

Lines 105 ff: It is not easy to understand the statement "constant temperature and pressure is assumed" when you just before present the T and p tendency equations. My understanding is this: you do not need p and T themselves, all you need is the changing supersaturation which in turn can be prescribed, such that the proper causes for changing S, namely changing p and T, need not to be explicit. If my interpretation is correct, I suggest that the authors change their explanations accordingly. In this case, why do we still need p_{env} and T_{env} , is this because there are p and T dependent parameters in the equations (e.g. diffusion)?

Line 120: You may add that such a high value is mainly chosen in order to let the nucleation process run without consumption of the available aerosol droplets, which would complicate the interpretation of the results.

Eq. 20: As a_w and Δa_w have a special value for water saturation, say Δa_w^* I would prefer to have the equation in this form $J_{sol}(\Delta a_w^*[T])=J_{pure liq}(T)$.

Line 164: Can we understand why this equality fails?

Line 168: strictly speaking, the log of the nucleation rate is shifted, which means that the nucleation rate itself is to be multiplied by a constant (obviously a constant far away from unity).

Figure 1 caption: Not completely clear. I think the blue line is for solution droplets with infinite dissolution, that is pure water droplets. This should be clear in the figure caption.

Line 176: I still don't understand this. Probably I did not follow the most recent literature, but who (reference) has found that the Koop et al 2000 parameterisation is wrong? Can the results of Koop and Murray for pure water be used as a proof for the falsehood of the older formulation when the latter is applied to solution droplets? In order to accept the shifted Koop 2000 line as a reference, one needs to know what is wrong with the unshifted original line. As far as I remember, this was based on measurements as well. Have there been indications that these measurements were incorrect? Later you demonstrate that differences between several formulations and your reference formulation are as small as typical measurement uncertainties and that it can therefore not be decided, which formulation is "correct" (I would rather say, which formulation is most appropriate, since "correct" implies that you know the truth.) So it seems, the motivation for your exercise is not that the original formulation is wrong, but rather that you seek for something quite simple that can be used in analytical studies. If so, this should be clearly stated in the beginning.

Line 202: replace "explanation" by "interpretation".

Line 211: Isn't the degree of $p(x)$ fixed by your ansatz in Eq. 25?

Line 286: not clear, what the vertical axis here represents.

Section 3.5.2: I don't get the point here. Before you have made a best fit, but what is interesting in showing results obtained with worse fits? I think, this section can be shortened without much loss to the reader.

Section 3.6: I must admit, that I cannot follow in this section. This kind of mathematics is not in my repertoire. I fear that other readers can have problems as well with this section. I suggest that a short paragraph be added, either here or as an appendix, where this technique is shortly explained.

Section 5: Please make clearer what is new here to the developments in Section 3. In 3, the reference nucleation rate was the Koop et al 2000 formulation, but shifted such that the Koop and Murray results for pure water were matched in a certain T-range. It is not quite clear to me what the difference of this to the new fit is.

Line 572: "avoid"

Line 634: This is an important remark. Can you give any recommendation how this formulation should be changed in such models to become more realistic?