Atmos. Chem. Phys. Discuss., community comment CC1
https://doi.org/10.5194/acp-2022-434-CC1, 2022

Comment on acp-2022-434

Bernd Kärcher

Community 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-CC1, 2022

I take the opportunity to comment (italicized) on a few issues in the manuscript that relate to my work for clarification and in the hope to support the overall review process.

[I] Abstract line 1: "Ice formation in cold temperature regimes is most probably dominated by homogeneous
freezing of aqueous solution droplets."

The first sentence in the paper is a strong statement demanding justification. Clearly, homogeneous freezing of solution droplets is a fundamental atmospheric ice formation process that operates when ice-nucleating particles are either absent or present in very low concentrations. Any assertion whether and in which circumstances this pathway dominates 'ice formation in cold temperature regimes' (an active area of research) needs to be supported by appropriate references. What about the role of mineral dust in cirrus cloud formation [Froyd et al., 2022], with dust particles long known to be very efficient heterogeneous ice-nucleating particles?

[II] line 33: in-situ vs liquid origin formation

I don't quite understand the categorization in-situ vs liquid origin formation. In the absence of cloud water droplets, freezing agents include solution droplets, and also in this case, it is the liquid water that freezes. Plus, situations where cloud droplets freeze are also in-situ events. Does liquid origin formation distinguish between ice formation in convective detrainment zones and in conveyeor belts, in which either solution and cloud droplets may freeze?

[III] Abstract line 3: "... idealized nucleation events as modelled with a state-of-the-art ice
microphysics, ..."

Why would the nucleation event be idealized? I think it is the modeling that is idealized. Which of the microphysics schemes referred to / used in the manuscript is 'state-of-the art' and how is this attribute defined? The numerical scheme used in Kärcher & Lohmann (2002) does not employ Lagrangian particle physics, as claimed in line 771.

In that context: could you please motivate / explain more clearly why "... details of the nucleation rate are less important for simulating ice nucleation in bulk models ..."

(abstract line 6) only relates to bulk models?

[IV] line 779-785: "In the study by Kärcher and Lohmann (2002) the impact of latent heat release on the diffusional growth is not considered. It is argued, that for cold temperature this effect is negligible. However, we found in our investigations, that this is only true for temperatures well below 220K. ... using parameterisations based on Kärcher & Lohmann (2002) might lead to moderately enhanced ice crystal number concentrations in the warm temperature regime."

Latent heat release enhancing temperatures during homogeneous freezing of cloud water droplets roll off around 234K depending on the CCN spectrum [Kärcher, 2017]. Here, the authors assert to find moderate (significant?) reductions of homogeneously nucleated ice number concentrations down to much colder temperatures due to latent heat release, this should be demonstrated by numerical simulations. Please analyse the individual diabatic contributions due to water phase changes to the temperature budget with the dry adiabatic tendency at 220K and show a quantitative comparison of homogeneously nucleated ice crystal number concentrations between simulations across a range of updraft speeds with and without including effects of latent release.

[V] Abstract line 11: "In contrast, the maximum supersaturation and thus the nucleation thresholds reached during an ice nucleation event depend on the vertical updraft velocity or cooling rate. This feature might explain some high supersaturation values during nucleation events in cloud chambers and suggests a reformulation of ice nucleation schemes used in coarse models based on a fixed nucleation threshold."

The point that homogeneous freezing thresholds depend on cooling rate and droplet size is known [Kärcher et al., 2022]. I am aware of the difficulty to oversee the onslaught of manuscripts in the scientific literature. In Kärcher et al. [2017], we have analysed the microscale characteristics of homogeneous freezing events in detail. Both articles are relevant for the present manuscript and therefore the authors may want to update their reference list.

I express strong doubts that a w-dependence of the freezing supersaturation threshold might explain low temperature cloud chamber data; if the authors have indications for this to be a valid explanation, it should explicitly be included in the study, focussing on the region T<205 K. Also, the treatment of the whole issue in the manuscript is somewhat confusing, as the authors admit in line 631 that "This might be interpreted as a hint that the formulation by Nachbar et al. (2019) is the more appropriate formulation for the saturation vapour pressure.", i.e., invoking a low-T correction of the currently used saturation vapor pressure as the main cause of high freezing thresholds and not deviations from a fixed freezing threshold in parameterizations of cirrus cloud formation.

Also, it remains unclear why this suggests a reformulation of ice nucleation schemes used in coarse models. In cirrus, the dependence of nucleated ice numbers on the threshold supersaturation is relatively weak (less than linear, see Kärcher & Lohmann, 2002), let alone the logarithmic dependences on the cooling rate, the liquid water volume in solution droplets, and the total droplet number concentration. These dependencies are consistent with / well explained by the self-terminating homogeneous freezing-relaxtion mechanism;

recently your team recognized based on numerical work that an increased nucleation threshold has little impact on ice crystal numbers (Baumgartner et al., 2022).

Please note that the purpose of cirrus parameterizations is to estimate total nucleated ice crystal number concentrations, not to resolve the region around the ice supersaturation maximum. The main differences between homogeneous freezing parameterizations (Barahona & Nenes [2008], Dinh et al. (2016), Kärcher & Lohmann (2002)) may be traced back to how the integration over the supersaturation history is approximated.

[VI] line 603: "The nucleation threshold assigned to the frequently used value j0 = 16 is completely arbitrary
chosen; there is no convincing physical justification for using this particular value; ..."

I am not sure if this is actually a true statement. My understanding re Koop et al. (2000) has always been that a given choice for j0 is not 'completely arbitrary' but chosen as it relates to a nucleation timescale appropriate for analysing the laboratory experiments. This relevant observation time is roughly given by the inverse of the product of the freezing rate coefficient and the volume of supercooled liquid water in the small droplets investigated. Please check with Koop et al. In Kärcher & Lohmann (2002), we have determined T-dependent freezing thresholds by imposing j0 = 10 for (wet radius) 0.25 µm solution droplets, a typical mean size of freezing droplets in cirrus levels, ignoring the comparatively small dry aerosol core volume.

[VII] Abstract line 9: "The use of just one distinct nucleation threshold for analysis and model
parameterisation should be reinvestigated."

In the light of the above , I am not aware of any cirrus parameterization scheme that uses a single value for this threshold, at least a temperature dependence is employed. Please check.

[VIII] line 290: "We use the simple bulk ice physics model as described by the set of ODEs ..."

I would argue that bulk models are not the tools of choice to carry out ice nucleation studies. In the case of homogeneous freezing, early freezing large droplets form a cohort of ice crystals that grows ahead of those forming later at higher supersaturation and on smaller droplets. In this way, a size-dispersed ice crystal spectrum is generated allowing for deposition growth of early formed ice crystals to change (reduce) the supersaturation conditions for later nucleation, as growth rates of µm-crystals are very rapid. It is this self-terminating freezing-relaxation mechanism that eventually determines the total homogeneously nucleated ice numbers, irrespective of updraft speed. Due to the sensitive dependence of the freezing rate coefficient on temperature and water activity (ice supersaturation), it is key to model size-resolved homogeneous freezing, if only approximately analytically, as in our parameterization scheme (Kärcher & Lohmann, 2002). It is also needed to robustly model heterogeneous nucleation events, as ice-nucleating particles activate into ice crystals across a range of supersaturation.

The point is, of course, that, by design, bulk models (aka two-moment schemes) are not capable of resolving this competition for available water vapor and therefore do not correctly represent the nucleation pulse and may not robustly predict nucleated ice numbers. Doing so requires a size/mass bin (spectral) approach or particle-based microphysics. For the purpose of studying nucleation in clouds, why not use a detailed model describing all relevant processes properly?

Bulk models can only be expected to deliver accurate total homogeneously nucleated ice *numbers if they simulate bulk water vapor uptake on freshly nucleated ice crystals accurately. Moreover, modal bulk models based on a fixed functional form of the ice crystal size spectrum (via a constant distribution width such as given in line 717) cannot reproduce the rapid change of the size spread of ice crystals during a freezing event that is ultinmately responsible for the quenching of the supersaturation and the shutting off of the freezing pulse.*

If one wishes, one might view nucleation as parameterized, i.e., constrained / tuned by assumed parameters in bulk models. Arguably, the "overall good agreement of our simple model with the more sophisticated models" (line 774) might be coincidental.

[IX] Short summary: "However, the maximum supersaturation during nucleation events shows strong changes. This quantity should be used for diagnostics instead of the popular nucleation threshold."

I have a hard time supporting the 'should' in the above summary statement. Rather, the question arises how accurately the bulk model used in the manuscript is able to simulate the degree of overshooting. Overshooting will always constrain the maximum supersaturation close to (within a few percent) the freezing-relaxation threshold because of the freezing-realaxtion feedback: the higher ice supersaturation overshoots (due to faster cooling), the more droplets freeze, the faster the freezing events terminates. The peak supersaturation attained during a freezing event must be distinguished from the characteristic supersaturation where freezing-relaxation sets in, see discussion in Kärcher et al. [2022].

[X] line 148: "Remark: As shown in Spreitzer et al. (2017), it is possible to determine and characterize
the steady states of the reduced system, which additionally includes sedimentation. This leads to a nonlinear
oscillator with a bifurcation diagram, depending on the updraft velocity w, and on the temperature T."

I am not sure why this remark features prominently here. There is no connection made to the topic of the manuscript. Could you better motivation this insertion? Also, Spreitzer et al. (2007) seem to describe a numerical artifact (the occurrence of nucleation cycles in a nucleating air parcel triggered by sedimentation and sustained cooling) that is tied to a coarse spatial resolution (box height) relative to the shallow depth of homogeneous freezing zones. The cycles occur when the timescale of vapor loss due to sedimentation (depending on the layer depth) matches the time scale of supersaturation production that scales in proprtion to the imposed updraft speed. High resolution models (meter resolution in the vertical) show that homogeneous freezing at the top of nucleation layers is a continuous process, see e.g., Lin et al. [2005].

Additional references included in this commentary:

Barahona, D. & Nenes, A. (2008).
Parameterization of cirrus cloud formation in large-scale models: Homogeneous nucleation.
J. Geophys. Res., 113.
doi: 10.1029/2007JD009355

Lin, R.-F., Starr, D. O., Reichardt, J. & DeMott, P. J. (2005).
Nucleation in synoptically forced cirrostratus.
J. Geophys. Res., 110, D08208.
https://doi.org/doi:10.1029/2004JD005362.

Froyd, K. D. et al. (2022).
Dominant role of mineral dust in cirrus cloud formation revealed by global-scale measurements.
Nat. Geosci. 15.
https://doi.org/10.1038/s41561-022-00901-w

Kärcher, B. (2017).
Homogeneous ice formation in convective cloud outflow regions.
Q. J. R. Meteorol. Soc. 143.
https://doi.org/10.1002/qj.3069

Kärcher, B. & Jensen, E. J. (2017).
Microscale characteristics of homogeneous freezing events in cirrus clouds.
Geophysical Research Letters, 44.
https://doi.org/10.1002/2016gl072486

Kärcher, B., DeMott, P. J., Jensen, E. J. & Harrington, J. Y. (2022).
Studies on the competition between homogeneous and heterogeneous ice nucleation in cirrus formation.
Journal of Geophysical Research, 127.
https://doi.org/10.1029/2021JD035805