

Reviewer 1

The overall quality of the preprint by Lüttmer et al. is high with good focus, detail, and scientific impact. The authors revisit the accuracy of current models in assuming a homogeneous temperature cutoff for the formation of ice in the atmosphere and instead they use Monte Carlo simulations of droplets freezing under different conditions in the atmosphere, including updraft velocity, super particle number, and CCN concentration. They consider two sets of simulations with and without vapor deposition and two parameterizations for J_{hom} . This work provides a valuable contribution to the field in untangling the importance of homogeneous nucleation albeit under relatively idealistic assumptions. The following specific comments and technical corrections should be considered before publication.

Specific comments:

line 100 and 174. The authors claim that J_{hom} should not depend on the ambient vapor pressure because the ice embryo is not in contact with it. However, an alternative explanation could explain this dependence. If the liquid water is in a subsaturated environment, then the activity of the liquid water changes correspondingly. This change in the liquid water could then have an impact on the homogeneous nucleation rate (similar to how a solute would reduce water activity), so that a change in ambient supersaturation ultimately affects J_{hom} in an indirect manner through the water activity. This interpretation aligns with the authors' Fig. 1b, where a water saturation < 1 leads to reduced J_{hom} , while water saturation > 1 leads to higher J_{hom} . At subsaturated Sw conditions, there may be a driving force for the liquid water molecules to evaporate that competes with J_{hom} . At supersaturated Sw conditions, the driving force reverses, such that there could be condensation to enhance J_{hom} . Thus, the authors should provide additional justification of their stated hypothesis or consider alternative explanations.

Response: For pure-water droplets, the JHOM formulation of Koop and Murray (2016) holds true and is independent of water activity. However, the reviewer's point is somewhat valid, as in the model droplets are initialised as CCNs and we assume that the CCN consists of a solute. Hence, when the droplet size decreases, the concentration of solute increases. This foreign substance can inhibit the formation of ice embryos, which might indeed explain the behaviour of JHOM under subsaturated Sw conditions. On the other hand, the homogeneously freezing droplets are in the micrometre size range, and thus the amount of foreign substance is low. The initial wet radius of the CCN droplet is 15 nm for $\kappa=0.64$. Hence, for micrometre-sized cloud droplets, the concentration and thus the influence of the foreign substance is likely negligible. Note that the stochastic freezing approach presented here is already very sensitive to droplet size, as it depends on volume. Hence, the freezing probability already decreases strongly as the droplet size decreases during evaporation. For subsaturated Sw conditions, the impact of the solute then depends on the timescale of freezing versus the timescale of evaporation.

This argument can, of course, also explain the behaviour of enhanced JHOM under supersaturated Sw conditions, as the droplets grow and the concentration of foreign substances decreases.

We expanded the "homogeneous freezing" subsection with a discussion of this effect and adjusted the discussion in the conclusions section accordingly.

We acknowledge that the effect of solutes on homogeneous freezing is not fully represented in the model here. However, this is associated with large uncertainties and is beyond the scope of this manuscript, and would require a study more focused on the physical chemistry behaviour of water.

line 138. The authors claim that the KP00 parameterization shows that J_{hom} is "independent of the amount of solute". To my knowledge, KP00 showed that J_{hom} is directly independent of the type of solute, not the amount. It could be better to instead state that J_{hom} depends on water activity and that water activity decreases with the addition of more solute.

Response: The reviewer is correct and we addressed the mistake.

The organization of the manuscript could be improved slightly. For example, the first two paragraphs of the Results before 3.1 appear to be detailed descriptions of the parameters used, which could be better placed in a new subsection at the end of the Methods. The end of the Methods section could conclude with an overview, such as a table, of all the simulations performed and ensembles studied to help the reader obtain an overall impression of which variables were studied in which subsection of the Results. Similarly, the beginning of Section 3.3 could be moved to the Methods section to define σ prior to its use in the Results.

Response: We reorganised the description of the model setup into its own subsection in Methods, as suggested, including a brief overview of the ensemble simulations conducted. However, a short text block turned out to be more concise than a table. We also include a definition of the σ parameter in this section; however, we kept most of the introductory discussion of the σ -ensemble in Section 3.3, as this makes the section more self-sufficient and improves the flow of reading and reasoning.

Technical corrections.

line 63. typo 'an' to 'a' stochastic process

Response: We corrected the mistake.

line 110. the notation in Equation 1 and the text could be more consistent. The text defines $P_{\text{frz}}(\Delta t)$, but Equation 1 uses the notation $P_{\text{frz}}(T)$, and the sentence afterwards states that P_{frz} does not depend on the time interval, so the use of $P_{\text{frz}}(\Delta t)$ could instead be replaced by P_{frz} alone or $P_{\text{frz}}(T)$.

Response: We reordered and rephrased the paragraphs about P_{frz} . We now clearly distinguish between the threshold formulation for $P_{\text{frz,thr}}$ only dependent on T and the general formulation of P_{frz} dependent on T , Δt and J_{hom} .

line 189. 'capacity' may be better as 'capacitance'

Response: We changed the term "capacity" to "capacitance," which also matches the name used for this variable later in the manuscript.

line 196. 'were,' can be deleted

Response: We corrected the mistake.

line 222. typo 'a' to 'an' adiabatic parcel model

Response: We corrected the mistake.

Figure 2 (a) and (e) are missing x-axis labels. In the caption, the number of simulations performed could be stated. Was it just one simulation each for JHOM-T and -DWA? The reader could be directed here to section 3.2.1 for the discussion that one simulation is sufficient given $nsd = 1000$.

Response: We fixed the bug in the plotting script; the x-axis in Fig. 2(a,e) now display the correct label. We added the number of super-droplets to the caption and included a sentence in the discussion highlighting that showing a single simulation is sufficient including a reference to the relevant section, as suggested.

line 274. typo ΔT to Δt for time

Response: We corrected the mistake.

line 287. in both panels (d) and (h), the blue line is solid. Should the red line be solid in panel (h)?

Response: We fixed the bug in the plotting script; the inactive JHOM formulations are now correctly displayed as dashed lines.

line 443. missing reference in parentheses

Response: We fixed the label in the tex file and the reference is now displayed correctly.