

Response to reviewer #2

This study discussed the representation of ice activation in cloud models and identified a problem with the application of cumulative activation fractions when considering the INP/ice particle budget. The authors formulated differential activation fractions that are consistent with the reduction of INP number after activation and demonstrated that the new representation can prevent the INP overestimation. They applied the new formulation with lab-based soot INP measurements and showed using the differential activation fractions indeed prevents the INP overestimation.

The manuscript is concise but very clearly written. The derivation of the formulation is inspiring. This work will improve the INP representation in cloud parameterizations, especially for considering the competition between homogeneous and heterogeneous ice nucleation processes. I recommend publication after some clarifications. Below please find my specific comments.

We thank the reviewer for the positive assessment of our manuscript. We respond to the specific comments below.

Title: In my opinion, the title is a little bit too general. A more specific title would be better, e.g., something like “Improving the heterogeneous ice nucleation parameterization using differential activation fractions”?

We think the current title: “The representation of heterogeneous ice activation in cloud models” is more precise, as we do not improve nucleation parameterizations, but rather how they are employed in models. We add “Aerosol-cloud interactions:” to put this work into the broader context and to indicate that we are not just addressing a technical detail in our study. More importantly, we increase awareness concerning a pitfall in simulating aerosol indirect effects on clouds that more often than not might pass unnoticed, with potentially wide-reaching repercussions for the robustness of climate change predictions.

Line 31-32: Then for immersion freezing measurements that are reported only as a function of temperature, does the INP overestimation problem also exist?

The same problem also exists for immersion freezing, since INPs only become active below a threshold temperature. This temperature is characteristic for individual particles implying that immersion freezing is largely deterministic.

To make this clearer, we supplement the sentence on lines 31–32 to read: “In the case of immersion freezing experiments, where an ensemble of water droplets with immersed INPs is cooled, frozen fractions are parameterized as a function of supercooling (temperature, T) instead of supersaturation such that differential and cumulative AFs are functions of T instead of s .”

Figure 1 caption (3rd line): “‘no budget’ approach, arrows labeled with ϕ ”. There are three arrows labeled with ϕ , but it seems only two of them indicate no budget approach?

Please note that the third (blue) arrow is labelled with φ , whereas the other two (black) arrows are labelled with ϕ . For clarity, we correct in line 3: “black arrows labelled with ϕ ”.

Page 5, Line 103-105, formula (7): Could you please elaborate how you came up the idea of using such a mathematical form? In other words, why other forms can not conveniently fit cumulative AFs? Does it work for other activation fraction forms other than the one (ns function) reported Ullrich et al.

(2017)? Also, maybe consider showing the measured and fitted curves in a figure as appendix? Just to have an idea about how “reasonable” it is.

In the entire discussion, the specific functional form of s -cumulated ice-active fractions does not matter for our results. The hyperbolic tangent was chosen solely for convenience; it is based on only two parameters, s_ and δs , with clear physical significance (line 104) and allows to easily compare measured and parameterized ice-active fractions (line 105). We presume that it provides reasonable fits to activation curves of other INP types as well, which show a similar s -dependence, including contrail-processed aviation soot particles shown in Fig.4. Please note this choice does not imply that other functional forms (involving, e.g., the error function) cannot provide such fits as well.*

We applied the hyperbolic tangent approximation to fit the activation curve for monodisperse ($1\ \mu\text{m}$) dust particles from Ullrich et al. (2017), using $s_ = 0.352$ and $\delta s = 0.0175$. To illustrate how well the dust parameterization is approximated by equation 7, we add a figure in a short appendix with the title “Analytical representation of ice-active fractions” and refer to it in line 108. The new figure shows that the tanh-fit approximates the parameterization very well, especially in the crucial part around s_* , where Φ rises steeply from low to significant values. Using the original parameterization, which is more cumbersome to evaluate, would not alter the discussion of results in section 3.*

Page 5, Line 108-110: It seems that the choices of s^* and δs (a factor of 3 changes) are a bit arbitrary (or did I miss something important?). If $s^*=0.352$ and $\delta s=0.0175$ are used for plotting figure 2, how will the results look like? (I assume δs here is not the grid spacing Δs).

We believe this comment refers to our choice $s_ = 0.35$ and $\delta s = 0.05$ stated in line 110. The reviewer is correct: the slope parameter δs is not to be confused with the grid spacing Δs varied in Fig.2. The choice $s_* = 0.35$ corresponds to the value for the $1\ \mu\text{m}$ dust particles rounded to two digits. We have chosen a larger slope parameter (relative to dust) spreading ice activation over a larger range of supersaturation than that shown in the above figure, mainly to illustrate the ice activation events shown in Fig. 4 more clearly.*

We add in the novel appendix: “In discussing deterministic ice formation (section 3.2), we have chosen a larger slope parameter, $\delta s=0.05$. This spreads ice activation over a larger range of s -values and illustrates the ice activation events more clearly.”

Page 8, Figure 4: It looks a bit surprising to me that the activation fraction for soot with 400nm size is similar to that for the desert dust particles as shown in Figure 2. Also, do you still need to fit cumulative AFs for this application? If so, how did you choose s_* and δs ?

Please note that the soot particle size (400 nm) is a mobility diameter (line 165) while the dust particle size ($1\ \mu\text{m}$) is the diameter of a volume-equivalent sphere (line 107), so both sizes (and by inference their cumulative ice-active fractions) are not directly comparable. For this application, we did not fit the cumulative ice-active fractions to the hyperbolic tangent, but compute them directly from the physical parameterization for soot-PCF (Marcolli et al., 2021) applied to aircraft soot in order to display them along with their differential counterparts in Fig.4.

We add on line 171: “with $\Phi(s,D)$ taken from Marcolli et al. (2021).”